出國報告書

(出國類別:會議)

參加 2009 年美國地球物理學會 秋季會議 出國報告

服務機關:行政院環境保護署 姓名職稱:張順欽簡任技正

派赴國家:美國

出國期間:98年12月12日至12月20日

報告日期:99年1月30日

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摘要

2009年美國地球物理學會秋季年度會議綜合了太空、大氣、海洋、水文、地質等等不同學門的研究,以演講、口頭報告及海報等發表方式,提供研究人員發表及交流的平台。據統計共有來自全球 78 個國家,超過 15000 人,參與此次會議,會議場地位於美國舊金山舉行。由於發表論文眾多,爲了節能減碳經省經費,會議並未印發論文集,而是以網路線上登載方式,與會者可以根據自己專長或有興趣之領域、題目選定參與之時段與會議場所。本次奉派出席 2009 年美國地球物理學會秋季年度會議,除汲取國際環境監測新知及最新應用,供作本署環境監測業務推動參考。同時於會中發表 2009 年 4 月 25 日我國受中國大陸沙塵影響空氣品質之監測成果,說明我國受污染物長程傳輸之影響,彰顯我國環境監測成果,有助於國際監測合作之推動。本次重要心得與建議:1.空氣污染物對於氣候變遷的影響日益重要,宜進行分析與追蹤;2.衛星觀測應用於空氣污染物傳輸日趨成熟,值得投入研究;3.遙測技術應用於空氣污染物排放與空氣品質監測,值得國內參考;4.溫室氣體監測與推估技術,值得持續追蹤參考。

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壹、目的

1.1 會議名稱

2009 年美國地球物理學會(AGU, American Geophysical Union)秋季年度會議

1.2 出國人員

環保署環境監測及資訊處 張順欽簡任技正

1.3 出國日期

98年12月12日至20日

1.4 預期效益

- 派員出席 2009 年美國地球物理學會秋季年度會議,汲取國際環境監測新知及最新應用,供作本署環境監測業務推動參考。
- 2.於會中發表 2009 年 4 月 25 日我國受中國大陸沙塵影響空氣品質之監測成果,說明我國受污染物長程傳輸之影響,彰顯我國環境監測成果,有助於國際監測合作之推動。

1.5 2009 年 4 月 25 日沙塵個案背景說明

2009 年 4 月 25 日下午受大陸沙塵影響,全台 76 個空氣品質監測站有 69 站空氣污染指標値(PSI)超過 100,空氣品質達不良等級,為近 20 年影響台灣最強的一次大陸沙塵。沙塵在 25 日下午影響台灣北部,區域 PM₁₀ 最高濃度超過 900 μg m⁻³,是日入夜後影響中南部,區域 PM₁₀濃度達到最高濃度約 700 μg m⁻³,26 日清晨最高濃度影響到達高屏及台東,約為 500 μg m⁻³。藉由本署與美國 NASA合作觀測之微脈衝雷達(Micro Pulse Lidar)觀測資料顯示,本次沙塵通過台灣北部時,分布高度約在 2 公里;抵達台灣中部時分布高度降低,在海拔 500 公尺的埔里或位於 2,862 公尺的鹿林山,沙塵影響相當輕微。沙塵分布在垂直方向的較低高層近地表空間迅速移動,自沙塵源區到台灣僅約 48 小時,造成了前所未有的高濃度沙塵。此外,細懸浮微粒(PM_{2.5})濃度也大幅增加,其中硫酸鹽濃度增加更超過本地污染累積濃度,顯示區域性污染傳輸的影響,對於民眾健康效應更須注意。中國沙塵對我國空氣品質之影響由來已久,但以本次沙塵影響最顯著。由於

沙塵源區資訊以及中國大陸華北、華中甚至東南沿海空氣品質資料獲得不易,難以掌握沙塵暴的動態或工業污染物的傳輸情形,使得沙塵預警不確定性相當高。藉由國際研討會論文發表,可以提供中國大陸沙塵長程傳輸對下風地區影響的資訊,同時與國際類似研究人員進行技術交流,有助我國監測技術提升。

貳、會議過程

2009 年美國地球物理學會秋季年度會議綜合了太空、大氣、海洋、水文、 地質等等不同學門的研究,以演講、口頭報告及海報方式,提供研究人員發表及 交流的平台。

2.1 會議論文投稿

本次參加2009年AGU秋季年度會議以「Unusual PM episode caused by yellow sand over Taiwan on April 25, 2009 _ 摘要先行投稿, 論文經接受爲海報發表, 如 圖 1。

Email Templates: Poster Acceptance

sc3824@gmall.com From: fm-help@agu.org

CC: BCC: Subject:

FM09: Abstract Acceptance



I am pleased to inform you that the abstract listed below was accepted for a poster presentation at the 2009 AGU Fall Meeting, 14-18 December, in San Francisco, California. Poster sessions are held in Moscone South at 747 Howard Street.

Abstract Reference Number: 685750
Abstract Title: Unusual PM episode caused by yellow sand over Talwan on April 25, 2009
Paper Number: A21A-0106

Presentation Type: Poster Presentation Presentation Date and Time: December 15, 2009; From 8:00 AM to 12:20 PM Location: Poster Hall, Moscone South

Poster sessions are scheduled 0800h-1220h in the morning and 1340h-1800h in the afternoon. Although poster sessions are only active for one half day, authors must put up their displays in the morning between 0730h and 0800h and leave them up until 1800h for maximum viewing opportunity. You must be present at your poster for at least one hour during the allocated session time.

All posters must be removed from the Poster Hall at 1800h on the day of your presentation or they will be recycled.

PRESENTER INSTRUCTIONS AND GUIDELINES: Please review the presenter guidelines to prepare your session and order any needed AV equipment. Instructions can be found on the AGU Web site: https://www.agu.oro/meetings/fm09/guidelines/index.php

You may browse the entire program and create your personal itinerary here: http://agu-

Reminder. The pre-registration and housing deadline is 12 November. After 12 November, registration fees will increase.

Should you need additional information, please e-mail m-heip@aqu.org.

Regards,

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圖 1、論文接受發表文件

2.2 海報製作

論文摘要經接受爲海報發表後,即依照給定格式製作海報,如圖2。

Paper Number: A21A-0106

Unusual PM episode caused by yellow sand over Taiwan on April 25, 2009

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c. Department of Natural Resources, Chinese Culture University, Taiwan, R.O.C.

d. Chang Jung Christian University, Taiwan, R.O.C.

Abstract

This study analyzes the unusual particulate matter episode caused by yellow sand (YS), which affected Taiwan on April 25, 2009. During this YS event, the hourly PM₁₀ concentrations higher than 1000 µg m⁻³, about 20 times higher than usual concentration, were observed at several sites over northern Taiwan. Moreover, 69 of all 76 air quality monitoring stations around Taiwan had recorded the daily mean PM₁₀ concentrations exceeding Taiwan's air quality standard of 125 µg m⁻³.

1. Introduction

- Dust storms in northern China and Mongolia triggered by the local strong winds with the dust spreading out through the southward cold air mass to Korea, Japan, and Taiwan during winter - spring seasons.
- On the yearly average, there are 4-5 YS events and 6.1 total dust days. Consequently, YS events were proved to cause the
 increasing of cardiopulmonary emergency visiting rate during the dust-affecting periods in Taipei when ambient PM₁₀
 concentrations were above 90 µg m³.

2. Sources of monitoring data and instruments

- Taiwan Environmental Protection Administration (TEPA) established the Taiwan Air Quality Monitoring Network in September 1993. And there are 76 monitoring stations around Taiwan as shown in Fig. 1.
- The hourly particulate matters (PM₁₀ and PM_{2.5}) data from the TEPA air-quality monitoring stations are used in this study.
- The sulfate data (hourly concentration) is from R&P 8400S
 Ambient Particulate Sulfate (SO4²-) Monitor at Taipei supersite (TAS) with a PM₁₀ inlet, followed by a PM_{2.5} sharp cut cyclone.

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Fig. 1 Locations of the monitoring stations adopted in this study; from Taiwan Air Quality Monitoring Network.

3. Unusual PM10 and PM2.5 episode over Taiwan on April 25, 2009

- An unusual episode of PM₁₀ was observed in Taiwan on April 25, 2009 with 69 stations discovered daily average concentrations of PM₁₀ exceeding³, the air quality standard for particulate matter in Taiwan.
- The time distribution of high PM concentration peaks in different areas could provide us a better idea about the path of YS transport.
- The PM₁₀ concentration Max values of the central and southern occurred in 4 hours and 14 hours later than that of the northern region.

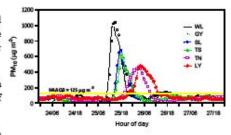


Fig. 2 Time series of PM₁₀ recorded from the sites located on western coast of Talwan April 25, 2009.

圖 2、論文海報(第一頁)

4. The YS event on April 25, 2006

 According to the surface weather maps, a cold front system passed over Taiwan on April 25 and the wind speed strengthened with the variation in wind direction from southwest into northeast.





Fig. 3 Photos in Taipel (a) before YS event dated on April 23, 2000 when PM₁₀ was 21 µg m³; (b) during YS event dated on April 25 , 2000 when PM₁₀ was 925 µg m³. (Photos was took from the top of TEPA building.)

- According to the NASA Ozone Monitoring Instrument aerosol index analysis result, a large-scale dust storm had taken place in northern China on April 25 (Fig. 4 (a)).
- Moreover, the result of NOAA 5 day backward trajectories (based on http://www.arl.noaa.gov) started from Taipei City at the time of PM₁₀ concentration Max (14:00 on April 25), the air masses moved southward with dust on April 22-23 affected Taipei on April 25. (Fig. 4 (b)).

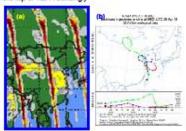
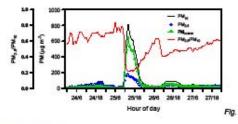
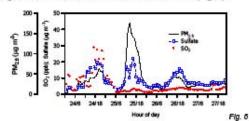


Fig 4

5. Observations at the Taipei aerosol supersite (TAS) during the YS period

- The maximum hourly values of PM_{2.5} and PM₁₀ particles at 175 and 814 μg m⁻³ respectively, were recorded at the TAS located in northern Taiwan (Fig. 5).
- The ratios of PM₁₀ to PM_{2.5} decreased from 0.6 during local accumulation period to 0.2 during YS period, indicating that
 the rise of the mass concentration of PM_{contre} (PM_{2.5-10}) is much higher than PM_{2.5} (Fig. 5).
- This YS event not only greatly increased the PM_{2.5} and PM₁₀ concentrations, but also enhanced the sulfate fraction of PM_{2.5}: 22 µg m⁻³ at TAS site during YS period was even higher than that derived from local accumulation. (Fig. 6).





6. Summary

- The YS impact on Taiwan air quality is characterized by a rapid increase of PM₁₀ level, high wind velocity, and low local
 gaseous pollutants. Notably, long-range transported sulfate concentration is even higher than that from local formation
 during poor dispersion condition.
- Several studies have indicated that the presence of yellow sand may be associated with increased health risks. The results
 of this study can provide the basis for further investigation into its effects on human health.

圖 2、論文海報(第二頁)

2.3 會議參加

據統計共有來自全球 78 個國家,超過 15000 人,參與此次會議,會議場地位於 Moscone Center West 如圖 3,研討會場大廳如圖 4。



圖 3、2009 年美國地球物理學會秋季年度會議會場(Moscone West)



圖 4、2009 年美國地球物理學會秋季年度會議大廳(Moscone West)

AGU 秋季年度會議綜合了太空、大氣、海洋、水文、地質等等不同學門的研究,發表方式包括演講、口頭報告及海報等,另有展示攤位。由於同一時段不同主題發表論文數量眾多,爲了節能減碳經省經費,會議並未印發論文集,而是以網路線上登載方式,與會者可以根據自己專長或有興趣之領域、題目選定參與之時段與會議場所。經上網選定參與場次如附件一。

12月14日上午前往會場報到,雖然研討會參與人數眾多,在主辦單位精心安排下,報到手續卻也井然有序,隨著研討會指引在很短時間內即順利完成報到手續,如圖 5。會場除提供無線網路作爲與會者聯絡上網之外,也提供電腦及印表機等服務,值得爲來辦理國際研討會參考。



圖 5、2009 年美國地球物理學會秋季年度會議報到(Moscone West)

海報發表場地位於 Moscone Center South 如圖 6,每日約有數百篇海報論文發表,如圖 7。論文以海報發表之作者,必需在當日自選一時段,於海報發表現場接受來賓詢問或討論。



圖 6、海報發表場地位於 Moscone Center South



圖 7、海報發表會場(Moscone Center South)

本次參加會議以「Unusual PM episode caused by yellow sand over Taiwan on April 25, 2009」發表 2009 年 4 月 25 日我國受大陸沙塵影響之論文,如附件一,海報現場如圖 8。

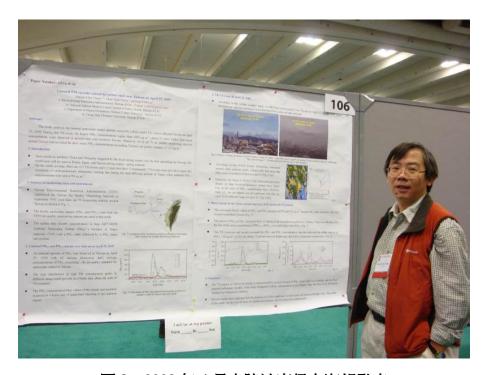


圖 8、2009 年 4 月大陸沙塵侵台海報發表

2.4 會議照片



圖 9、會前準備

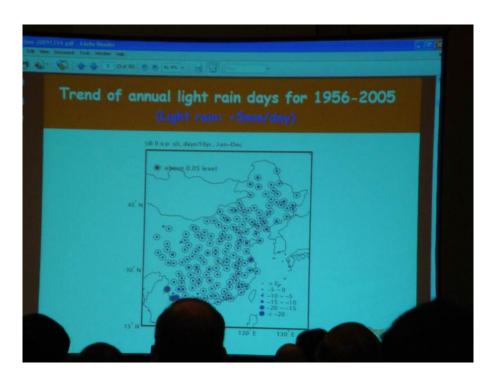


圖 10、參加研討會

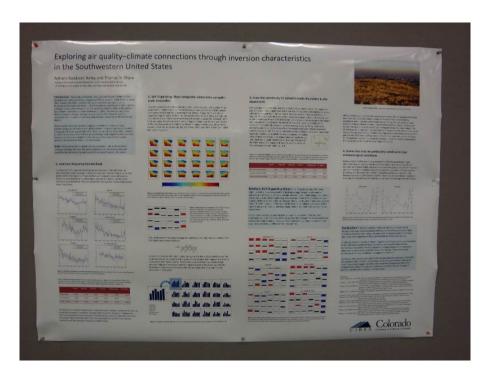


圖 11、參加海報發表研討



圖 12、參加海報發表研討



圖 13、展覽會場

叁、重要心得與建議

本次參與 2009 年美國地球物理學會秋季年度會議,除發表論文外,選擇業務相關之論文,例如衛星遙測技術於空氣污染物之監測應用、長程傳輸污染物之監測與模擬及空氣污染區域海陸氣流循環之監測與模擬等相關最新研究,瞭解最新技術研究及成果,作爲本署空氣品質及長程傳輸監測等相關業務推動參考。臚列重要心得與建議如下:

3.1 空氣污染物對於氣候變遷的影響日益重要,宜進行分析與追蹤

全球性空氣污染與溫室效應之關連性已漸漸受到重視空氣污染問題已不再 是地區性的影響,經由全球大氣循環,甚至可能影響到全球氣候。大氣微粒對於 環境影響除了導致能見度降低,對於地球的輻射平衡的干擾,也可能對氣候有重 大影響。微粒污染與微粒光學性質有關,對氣候的影響可分成直接與間接效應, 直接效應包含大氣微粒對太陽光輻射的吸收與散射;間接效應則是充當雲的凝結 核則改變雲的光學特性。西藏高原雪覆蓋面積減少、也懷疑與空氣污染有關。

不同氣膠成分對於太陽輻射的影響,如黑炭吸光造成增溫效應、硫酸鹽散光造成冷卻效應,在考慮全球溫室效應的同時,對於這些全球性空氣污染物的傳輸,尤須加以注意。中國大陸沙塵傳輸影響全球環境的現象,已是長久存在的問題,各種研究也日益深入,如傳輸機制、影響、預測等。而全球性空氣污染與氣候變遷之關連性則漸受重視,使得空氣污染已不再是地區性問題。經由全球大氣循環,空氣污染甚至可能影響到全球氣候。例如西藏高原雪覆蓋面積減少,懷疑與空氣污染有關,而中國大陸小雨天數減少,大雨天數增加,這與國內近年來的降雨型態變化頗爲類似,部分研究懷疑與空氣污染造成氣膠濃度增加有關,亦有研究認爲受全球溫室效應影響。類似氣候變遷導致之影響,官進行分析與追蹤。

3.2 衛星觀測應用於空氣污染物傳輸日趨成熟,值得投入研究

空氣污染物長程傳輸對於氣候的影響日漸受到關注,而對於長程傳輸的監測則有賴衛星觀測技術的應用。目前對於衛星觀測的氣膠光學厚度(Aerosol Optical Depth, AOD)應用於 PM_{2.5} 濃度監測已多有研究,值得繼續留意。尤其懸浮微粒一直是造成我國空氣品質問的汙染物,利用衛星觀測輔以地面傳統空氣品質監測

站,將更有助於建立我國空氣中懸浮微粒的污染特徵。

3.3 遙測技術應用於空氣污染物排放與空氣品質監測,值得國內參考

傳統空氣品質監測以單點方式進行監測,除了空間代表性易受氣象條件影響,監測項目也侷限在特定污染物種。以雷達遙測技術進行空氣污染排放與空氣品質監測,對於工業區與人口密集住宅區之影響或污染預防,應有所助益。

3.4 溫室氣體監測與推估技術,值得持續追蹤參考

隨著氣候變遷與全球暖化議題白熱化,溫室氣體監測研究方興未艾,包括監測技術的改進、模式模擬、區域排放量推估等等,都值得我國持續追蹤參考。目前我國以鹿林山與美國 NOAA 合作觀測溫室氣體,未來可以搭配國內溫室氣體減量之推動,持續進行有關溫室氣體之觀測與研究。

附件一、2009年美國地球物理學會秋季年度會議議程摘錄

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1:00 PM-5:00 PM, Grand Ballroom C (InterContinental San Francisco), **AGU Cultural Politics** of Climate Change Workshop

4:15 PM-5:30 PM, 2020-2024 (Moscone West), **AGU Member Open Forum on AGU Governance Changes**

Monday, December 14, 2009

8:00 AM-12:20 PM	A11D-0120. Formation of UV-vis Absorbing Organic Solutes, Films, and Suspended Precipitates in Sulfuric Acid Solutions at Stratospheric Aerosol Acidities A.L. Van Wyngarden; C.L. Belle; C.L. Dalle ore; M.J. Morrissey; J.M. Rodgers; L.T. Iraci
8:00-8:00 AM	A11D-0121. Toward Investigating Optically Trapped Organic Aerosols with CARS Microspectroscopy L.F. Voss
8:00-8:00 AM	A11D-0122. OH Oxidation Reactions of Unsaturated Self-assembled Monolayers on Germanium Surfaces: A System That Mimics Organics Adsorbed on Urban Surfaces S.G. Moussa; J. Raff; B.J. Finlayson-Pitts
8:00-8:00 AM	A11D-0123. Atmospheric Processing of Methylglyoxal and Glyoxal in Aqueous Environments J.L. Axson; V. Vaida
8:00-8:00 AM	A11D-0124. Role of Ozone in Particle Formation and Growth From the Nitrate Radical-Initiated Oxidation of α-Pinene V.M. PERRAUD; e.a. bruns; M.J. Ezell; S.N. Johnson; Y. Yu; M.L. Alexander; A. Zelenyuk; D.G. Imre; B.J. Finlayson-Pitts
8:00-8:00 AM	A11D-0125. Laboratory Investigation of Trace Gas Emissions from Biomass Burning on DoD Bases I.R. Burling; R.J. Yokelson; D.W. Griffith; J.M. Roberts; P.R. Veres; C. Warneke; T.J. Johnson
8:00-8:00 AM	A11D-0126. Effect of photosensitized chemistry on organic aerosol evolution A. Rouvière; P.F. DeCarlo; T. Bartels-Rausch; M. Ammann
8:00-8:00 AM	A11D-0127. Development of a Metastable Atom Bombardment (MAB) Source for Penning Ionization Time-of-flight Aerosol Mass Spectrometry C.B. Robinson; J.R. Kimmel; D. David; J.T. Jayne; A. Trimborn; D.R. Worsnop; J.L. Jimenez

8:00-8:00 AM	A11D-0128. Laboratory Studies of the Heterogeneous Oxidation of Levoglucosan in Biomass Burning Particles C.J. Hennigan; A. Sullivan; J.L. Collett; A.L. Robinson
8:00-8:00 AM	A11D-0129. Kinetics and Products of Heterogeneous Oxidation of Erythritol and Levoglucosan in Aerosol Particles S.H. Kessler; J.H. Kroll; K.R. Wilson; J.D. Smith
8:00-8:00 AM	Abstract Withdrawn
8:00-8:00 AM	A11D-0131. Gas-Phase and Particle-Phase Reaction and Kinetics of Epoxydiols from Photooxidation of Isoprene N.C. Eddingsaas; A.W. Chan; J.D. Surratt; J. Seinfeld; P.O. Wennberg
8:00-8:00 AM	A11D-0132. Improving and assessing vapour pressure estimation methods for organic compounds of atmospheric relevance using a Knudsen Effusion Mass Spectrometer (KEMS) A.M. Booth; D.O. Topping; G.B. McFiggans; A. Garforth; C.J. Percival
8:00-8:00 AM	A11D-0133. Heterogeneous Reactions of Surface-Adsorbed Catechol: A Comparison of Tropospheric Aerosol Surrogates. R.Z. Hinrichs; L.A. Woodill
8:00-8:00 AM	A11D-0134. Heterogeneous Reactions of NO3 and N2O5 with a Range of Organic Substrates R. lannone; S. Gross; S. Xiao; A.K. Bertram
8:00-8:00 AM	A11D-0135. Keto-Enol Tautomerizations Catalyzed by Water and Carboxylic Acids G. da Silva
8:00-8:00 AM	A11D-0136. Photoenhanced ozonation of polycyclic aromatic hydrocarbons on model urban film surfaces S.A. Styler; J.P. Wong; D.J. Donaldson
8:00-8:00 AM	A11D-0137. Reactions of Complex Phenols on Aerosols with Gaseous Ozone M.R. Hoffmann; S. Enami; A.J. Colussi
8:00-8:00 AM	A11D-0138. Temperature and Electrolyte Effects on the Thermochromism of Model Organic Aerosol Matter A.G. Rincon; M.I. Guzman; M.R. Hoffmann; A.J. Colussi
8:00-8:00 AM	A11D-0139. Formation of Secondary Organic Aerosol through Cloud Processing of Anthropogenic VOCs J.W. Hutchings; P. Herckes
8:00-8:00 AM	A11D-0140. Composition of Secondary Organic Aerosols Produced by Photo-Oxidation of Biomass Burning Emissions in a Smog Chamber Y. Desyaterik; A. Sullivan; C.J. Hennigan; A.L. Robinson; J.L. Collett
8:00-8:00 AM	A11D-0141. Can Secondary Organic Aerosol Formed in Atmospheric Simulation Chamber Be Continuously Aging? L. Qi; S. Nakao; Q. Malloy; B. Warren; D. Cocker

8:00-8:00 AM	A11D-0142. Secondary organic aerosol formation from m-xylene photooxidation: The role of the phenolic product S. Nakao; L. Qi; C. Clark; K. Sato; P. Tang; D. Cocker
8:00-8:00 AM	A11D-0143. Slow aging in Secondary Organic Aerosol observed by Liquid Chromatography coupled with High-Resolution Mass Spectrometry D.L. Bones; A.P. Bateman; T.B. Nguyen; J. Laskin; A. Laskin; S. Nizkorodov
8:00-8:00 AM	A11D-0144. Atmospheric Heterogeneous Stereochemistry G.Y. Stokes; A.M. Buchbinder; F.M. Geiger
8:00-8:00 AM	A11D-0145. Uptake of glyoxal on nitric acid/ice thin films under conditions relevant to the upper troposphere B.M. Connelly; D.O. De Haan; M.A. Tolbert
8:00-8:00 AM	A11D-0146. Detection of aerosol organic nitrate and ammonium nitrate by Thermal Dissociation – Laser Induced Fluorescence A.W. Rollins; J.F. Hunter; R.C. Cohen
8:00-8:00 AM	A11D-0147. Branching Ratios for the Reaction of Carbonyl-Containing Organic Peroxy Radicals with Hydroperoxy Radicals A. Hasson; G.S. Tyndall; S. Hernandez; S. Campbell; S. Singh; Y. Ibarra; J.J. Orlando
8:00-8:00 AM	A11D-0148. Particulate organic nitrate and organic sulfate contributions to SOA: Response of the AMS and constraints from field studies D. Farmer; A. Matsunaga; P.J. Ziemann; K. Docherty; A.C. Aiken; M. Cubison; K.R. Wilson; J.H. Kroll; J.D. Surratt; J.L. Jimenez
8:00-8:00 AM	A11D-0149. Changes in Concentrations and Stable Carbon Isotopic Compositions of Diacids, Ketoacids and α-Dicarbonyls in Atmospheric Aerosol Filter Samples with Photochemical Aging: A Laboratory Study C. Pavuluri; K. Kawamura
8:00-8:00 AM	A11D-0150. The Morphology of Internally Mixed SOA/DOP Particles and the Uptake of Gas-Phase DOP During SOA Formation T.D. Vaden; C. Song; R.A. Zaveri; D.G. Imre; A. Zelenyuk
8:00-8:00 AM	A11D-0151. Chemical aging of organic aerosols: Evidence of changing hygroscopicity and volatility F.D. Pope; P.J. Gallimore; P.T. Griffiths; S.L. Clegg; R. Cox; M. Kalberer
8:00-8:00 AM	A11D-0152. Substitution kinetics and energetics of aliphatic amines for ammonia in aerosols B.R. Bzdek; M.V. Johnston
8:00-8:00 AM	A11D-0153. Characterization of Potential Aerosol Mass using an Oxidation Chamber coupled to an Aerodyne HR-ToF-AMS during DAURE, SHARP, and FLAME-3 A.M. Ortega; W.H. Brune; M. Cubison; B.L. Lefer; S. Schallharter; A. Metzger; M. Mueller; A. Hansel; J.L. Jimenez

8:00-8:00 AM	A11D-0154. Changes in chemical composition observed during organic aerosol aging C.L. Heald; J.H. Kroll; J.L. Jimenez; K. Docherty; P.F. DeCarlo; A.C. Aiken
8:00-8:00 AM	A11D-0155. Determining aromatic reaction rates from the photo-oxidation of diesel exhaust using in-situ comprehensive multidimensional chromatography (GC × GC) D.R. Worton; A.H. Goldstein; N.M. Kreisberg; A.P. Teng; S.V. Hering; T. Gorecki; C.J. Hennigan; A.A. Presto; M. Ranjan; N.M. Donahue; A.L. Robinson
8:00-8:00 AM	A11D-0156. Effects of dilution on vehicle emissions of primary particles K.L. Hayden; S. Li; G. Liggio; M. McCurdy; T. Chan; J. Rostkowski
8:00-8:00 AM	A11D-0157. Field observations linking organic carbon content to optical properties in atmospheric aerosols B.A. Flowers; M.K. Dubey; C. Mazzoleni; A. Zelenyuk; J.J. Schauer
8:00-8:00 AM	A11D-0158. Summertime Anthropogenic and Biogenic Influences on Particle Composition at a Remote Forested Site in Western Canada J.G. Slowik; R.Y. Chang; S.J. Sjostedt; A. Vlasenko; K.L. Hayden; S. Li; G. Liggio; A. Macdonald; R. Leaitch; J. Abbatt
8:00-8:00 AM	A11D-0159. Can 3D Models Explain the Primary and Secondary, Fossil and non-Fossil Organic Aerosols during the 2006 MILAGRO Experiment? A. Hodzic; J.L. Jimenez; S. Madronich; J.D. Fast; A.S. Prevot; J.J. Schauer; E.M. Stone
8:00-8:00 AM	A11D-0160. Modeling of secondary organic aerosols formation during MILAGRO 2006 K. Dzepina; A. Hodzic; S. Madronich; R.A. Zaveri; R. Volkamer; C.D. Cappa; J.L. Jimenez
8:00-8:00 AM	A11D-0161. Investigation of Dairy Farm Silage Emissions C.S. McCluskey; D.R. Blake; M.M. Yang; J. Dehart
8:00-8:00 AM	A11D-0162. Apportionment of Size Distribution Data from the Aerodyne Aerosol Mass Spectrometer in Pittsburgh and Mexico City I.M. Ulbrich; Q. Zhang; A.C. Aiken; M. Canagaratna; N.L. Ng; D.R. Worsnop; J.L. Jimenez
8:00-8:00 AM	A11D-0163. Identification of Atmospheric Organic Matter in Fog Water: Exact Masses, Empirical Formulas, and Structural Insights L.R. Mazzoleni; J.L. Collett
8:00-8:00 AM	A11D-0164. Molecular distribution, seasonal variation, chemical transformation and sources of dicarboxylic acids and related compounds in atmospheric aerosols at remote marine Gosan site, Jeju Island S. Kundu; K. Kawamura; M. Lee

8:00-8:00 AM	A11D-0165. Measurements of gas-phase inorganic and organic acids in biomass fires by negative-ion proton-transfer chemical-ionization mass spectrometry (NI-PT-CIMS) P.R. Veres; J.M. Roberts; J.A. De Gouw; C. Warneke; I.R. Burling; R.J. Yokelson
8:00-8:00 AM	A11D-0166. Measurement of Organic Acids Produced By The Gas-Phase Ozonolysis of Simple Olefins Using Chemical Ionization Mass Spectrometry (CIMS) as a Function of Temperature And Humidity C.J. Percival; A. Bacak; K.E. Leather; M.R. McGillen
8:00-8:00 AM	A11D-0167. Measurements of Product-Specific VOC Reactivities during the PROPHET 2008 field intensive using proton transfer reaction linear ion trap (PTR-LIT) mass spectrometry L.H. Mielke; J.H. Slade; M. Alaghmand; S.B. Bertman; M. Carroll; S.M. Griffith; R.F. Hansen; S. Dusanter; P.S. Stevens; A. Hansel; P.B. Shepson
8:00-8:00 AM	A11D-0168. Formaldehyde emissions from industrial facilities in Houston, TX O. Pikelnaya; J. Stutz; D. Fu; J.H. Flynn; B.L. Lefer
8:00-8:00 AM	A11D-0169. Investigation of O Production Rates in the Mexico City Metropolitan Area during MILAGRO S. Dusanter; L.T. Molina; P.S. Stevens
8:00-8:00 AM	A11D-0170. Preliminary Results of Glyoxal Measurements at CABINEX 2009 M.E. Thurlow; A. O'Brien; M.M. Galloway; F. Keutsch
8:00-8:00 AM	A11D-0171. The Thermal Dissociation-Laser Induced Fluorescence (TD-LIF) technique for total Peroxy Nitrates (PNs) and comparisons to speciated PAN measurements P.J. Wooldridge; A.E. Perring; T.H. Bertram; F.M. Flocke; J.M. Roberts; H.B. Singh; G. Huey; J.A. Thornton; J.G. Murphy; J. Fry; A.W. Rollins; B.W. LaFranchi; R.C. Cohen
8:00-8:00 AM	A11D-0172. Measurements of Volatile Organic Compounds (VOCs) from Biomass Combustion – Emission Ratios, OH Reactivities and SOA Precursors W.C. Kuster; J.B. Gilman; P.D. Goldan; C. Warneke; J. deGouw; P.R. Veres; I.R. Burling; R.J. Yokelson
8:00-8:00 AM	A11D-0173. Isomeric Selective Studies of the Dominant Addition Channel in OH Initiated Oxidation of Isoprene B. Ghosh; A. Bugarin; B. Connell; S.W. North
8:00-8:00 AM	A11D-0174. Measurements of OH and HO ₂ Radical Chemistry in a Forest Environment R.F. Hansen; S. Dusanter; S.M. Griffith; P.S. Stevens
8:00-8:00 AM	A11D-0175. Measurement and model comparison of methacrolein and methyl vinyl ketone concentrations from the OH-initiated oxidation of isoprene under NOx free conditions M.A. Navarro; S. Dusanter; P.S. Stevens; R.A. Hites

8:00-8:00 AM	A11D-0176. OH and HO ₂ Concentrations during PROPHET 2008: Measurement and Theory S.M. Griffith; R.F. Hansen; S. Dusanter; P.S. Stevens; M.M. Galloway; J. Hottle; A. Kammrath; F. Keutsch; L.H. Mielke; M. Alaghmand; P.B. Shepson; N. Zhang; X. Zhou; S.B. Bertman; M. Carroll
8:00-8:00 AM	A11D-0177. Significant OH regeneration from the photooxidation of several atmospherically important alkenes J. Crounse; F. Paulot; J. Seinfeld; P.O. Wennberg
8:00-8:00 AM	A11D-0178. Measurements of OH reactivity in a South-East Asian tropical rainforest S. Vaughan; P. Edwards; K. Furneaux; L.K. Whalley; T. Ingham; A. Goddard; C. Seal; D.E. Heard; M.J. Evans; J. Lee; s. moller; J. Hopkins; C.E. Jones; J. Whitehead; C.N. Hewitt
10:20-10:35 AM	A12A-01. Impact of Climate Mitigation on Aerosol Concentrations and Health Effects in Asia N.E. Selin; C. Wang; J.M. Reilly; S. Paltsev; R.G. Prinn
10:35-10:55 AM	A12A-02. Containing Climate Change With Black Carbon Reductions: A Grand Challenge Field Experiment V. Ramanathan
10:55-11:15 AM	A12A-03. Aerosols in Asia and Global-to-Continental-scale Climate Change (<i>Invited</i>) V. Ramaswamy
11:15-11:35 AM	A12A-04. Changes in snow cover and water cycle over the Tibetan plateau induced by absorbing aerosols W.K. Lau; M. Kim; K. Kim; W. Lee
11:35-11:50 AM	A12A-05. Estimation of black carbon deposition from particulate data in the atmosphere at NCO-P site in Himalayas during pre-monsoon season and its implication to snow surface albedo reduction T.J. Yasunari; P. Bonasoni; P. Laj; K. Fujita; E. Vuillermoz; A. Marinoni; P. Cristofanelli; F. Calzolari; R. Duchi; G. Tartari; W.K. Lau
11:50-12:05 PM	A12A-06. Heavy pollution suppresses light rain in China: Observations and modeling Y. Qian; D. Gong; J. Fan; L. Leung; R. Bennartz; D. Chen; W. Wang
12:05-12:20 PM	A12A-07. Aerosols and Regional Climate Changes S.C. Liu
2:15-2:30 PM	A13K-03. Direct and Indirect Effects of Aerosols in China Z. Li; H. Chen; S. Tsay; J. Huang; W. Zhang

2:30-2:45 PM	A13K-04. An overview of dust aerosol effect on semi-arid climate during 2008 China-US joined field campaign J. Huang; J. Bi; W. ZHANG; J. Shi; S. Tsay; Z. Li; H. Chen; X. Wang; Z. Huang; B. Zhang; G. Wang; L. Zhang
2:45-3:00 PM	A13K-05. Air pollution and clouds in southern China: preliminary results from the observations in spring 2009 at Mt. Heng T. Wang; L. Xue; X. Gao; W. Nie; X. Wang; Y. Wang; M. Sun; A. Ding; S. Fan; Q. Zhang; W. Wang
3:00-3:15 PM	A13K-06. Aerosol Properties Observed on the Mesoscale and MODIS Aerosol Optical Depth Comparisons during the 2008 Pre-monsoon TIGERZ IOP in Kanpur, India D.M. Giles; B.N. Holben; S.N. Tripathi; T.F. Eck; W.W. Newcomb; I. Slutsker; R.R. Dickerson; A.M. Thompson; S. Wang; R. Singh
3:15-3:30 PM	A13K-07. Aerosol and Cloud Properties during the Cloud Cheju ABC Plume -Asian Monsoon Experiment (CAPMEX) 2008: Linking between Ground-based and UAV Measurements S. Kim; S. Yoon; M. Venkata Ramana; V. Ramanathan; H. Nguyen; S. Park; M. Kim
3:30-3:40 PM	A13K-08. Using unmanned aircraft to measure the impact of pollution plumes on atmospheric heating rates and cloud properties during the Cheju ABC Plume-Asian Monsoon Experiment (CAPMEX) M. Venkata Ramana; V. Ramanathan; H. Nguyen; Y. Xu; K. Pistone; C. Corrigan; Y. Feng; A. Zhu; S. Kim; S. Yoon; G.R. Carmichael; J.J. Schauer
5:18-5:32 PM	A14D-06. SOA Measurements vs. Models: a Status Report J.L. Jimenez; J.A. De Gouw
5:32-5:46 PM	A14D-07. Contributions of Selected Biogenic and Aromatic Compounds to the Formation of Tropospheric Secondary Organic Aerosol over Several Sites in the United States M. Jaoui; T.E. Kleindienst; M. Lewandowski; J.H. Offenberg; E.W. Corse; T. Gerald; E. Edney
5:46-6:00 PM	A14D-08. An AEROCOM Intercomparison Exercise in Organic Aerosol Modeling K. Tsigaridis; M. Kanakidou

Tuesday, December 15, 2009

8:00 AM-12:20 PM	A21A-0078. Global transport of Asian dust revealed by NASA/CALIPSO and a global aerosol transport model K. Eguchi; K. Yumimoto; I. Uno; T. Takemura
8:00-8:00 AM	A21A-0079. Variations of OC and EC concentrations in PM10, PM2.5 and PM1.0 in Jeju. S. Lim; M. Lee; G. Lee; K. Kang
8:00-8:00 AM	A21A-0080. Integrated Analysis of Bottom-up Emissions Information and Airborne Measurements in Support of NASA INTEX and ARCTAS Field Campaigns K. Choi; J. Woo; S.A. Vay; Y. Choi; H. Kim; S. Yoo; Y. Sunwoo
8:00-8:00 AM	A21A-0081. Particle Formation and Growth in North and East China: a Comparative Analysis of Measurements from Surface and Mountain -top Sites J. Gao; T. Wang; W.x. Wang; S. Poon; P.j. Xu; L.k. Xue; X.f. Wang; W. Nie; Z. Wang; W.s. Wu; F.h. Chai; X.z. Wang; A. Ding
8:00-8:00 AM	A21A-0082. Spatial and temporal variations of aerosols around Beijing in summer 2006: Model evaluation and source apportionment H. Matsui; M. Koike; Y. Kondo; N. Takegawa; K. Kita; Y. Miyazaki; M. Hu; S. Chang; D.R. Blake; J.D. Fast; R.A. Zaveri; D.G. Streets; Q. Zhang; T. Zhu
8:00-8:00 AM	A21A-0083. The Effects of Dust Emissions on the Summertime Climate of Southwest Asia: Incorporating Sub-grid Variability in a Regional Climate Model's Dust Scheme M.P. Marcella; E.A. Eltahir
8:00-8:00 AM	A21A-0084. Optical Properties of Fine/Coarse Mode Aerosol Mixtures T.F. Eck; B.N. Holben; A. Siniuk; R.T. Pinker; P. Goloub; H. Chen; B. Chatenet; Z. Li; R. Singh; S.N. Tripathi; O. Dubovik; D.M. Giles; J. Martins; J.S. Reid; N.T. O'Neill; A. Smirnov
8:00-8:00 AM	A21A-0085. Climate response to black carbon aerosols: Dependence on altitude G.A. Ban-Weiss; K.G. Caldeira; L. Cao; G. Bala
8:00-8:00 AM	A21A-0086. Effect of aerosol radiative forcing on the seasonal variation of snow over the northern hemisphere M. Kim; W.K. Lau; W. Lee; K. Kim
8:00-8:00 AM	A21A-0087. Long-term trends of aerosol carbon and nitrogen, their stable isotopic compositions, and water-soluble organic carbon in the western North Pacific K. Kawamura; E. Tachibana; N. Umomoto
8:00-8:00 AM	A21A-0088. Estimating bulk optical properties of aerosols over the western North Pacific by using MODIS and CERES measurements H. Oh; C. Ho; Y. Choi; R. Park; C. Song
8:00-8:00 AM	A21A-0089. Spectral aerosol light absorption measurement at urban, suburban, and remote sites in Korea Y.J. Kim; J. Jung; J. Kim; K. Lee; T. Batmunkh; M. Gonzaga-Cayetano; S. Kim

8:00-8:00 AM	A21A-0090. Evaluation of a newly developed below-cloud scavenging scheme of regional aerosol simulations: its implication for aerosol budget over East Asia S. Bae; R. Park; Y. Kim
8:00-8:00 AM	A21A-0091. Physical and hygroscopic properties and CCN characteristics of the aerosols measured at an island in the northwestern tip of South Korea S. Shim; J. Kim; W. Kim; S.S. Yum; I. Chang
8:00-8:00 AM	A21A-0092. Physicochemical evolution of dust particles observed in March 2007, Seoul, Korea I. Park; S. Lim; M. Lee; Y. Lee; G. Lee; J. Han
8:00-8:00 AM	A21A-0093. Comparison of PM _{2.5} carbonaceous aerosols in the northeast Asian regions M. Chang; M. Lee; S. Lim; J. Kim
8:00-8:00 AM	A21A-0094. The relationship between the Asian dust and Arctic Oscillation: An observational investigation Y. Lee; J. Kim; H. Cho
8:00-8:00 AM	A21A-0095. Solar Radiation and Cloud Change in China Calculated in a 20th Century Simulation with a Climate Model T. Nagashima; T. Nozawa; T. Hayasaka; K. Kawamoto
8:00-8:00 AM	A21A-0096. High Concentration of Surface Ozone Observed along the Khumbu Valley Nepal April 2007 J. Semple; K. Moore
8:00-8:00 AM	A21A-0097. Effect of the Presence of Crustal Elements (Ca2+, K+, and Mg2+) in Estimating Water Content in PM2.5 H. Lee; Y. Kim
8:00-8:00 AM	A21A-0098. The effects of aerosols on Thunderstorms and Lightning over the Pearl River Delta, China Y. Wang; R. Zhang; Q. Wan; W. Meng; F. Liao
8:00-8:00 AM	A21A-0099. Insights to the topographic and aerosol roles in the heavy rainfall at the Western Ghats and the scarce rainfall farther east at the rain shadow as observed in CAIPEEX R. Maheskumar; J.R. Kulkarni; M. Konwar; D. Rosenfeld; B. Goswami
8:00-8:00 AM	A21A-0100. The interplay between small and large CCN: Impacts on convective cloud microstructure as observed in CAIPEEX. M. Konwar; J. Kulkarni; R. Maheskumar; B. Goswami; D. Rosenfeld
8:00-8:00 AM	A21A-0101. Can Aerosol Offset Urban Heat Island Effect? M.s. Jin; J.M. Shepherd
8:00-8:00 AM	A21A-0102. Size distributions of secondary and primary aerosols in Asia: A 3-D modeling F. Yu; G. LUO; Z. Wang
8:00-8:00 AM	A21A-0103. Dust Aerosol Optical Properties Retrieval and Radiative Forcing over Northwestern China during 2008 China-US Joint Field Experiment J. Ge; J. Su; T.P. Ackerman; Q. Fu; J. Huang; J. Shi

8:00-8:00 AM	A21A-0104. Implementation of different dust emission schemes into WRF/Chem: An evaluation of dust emisson parameterizations J. Kang; S. Yoon; Y. Shao; E. Jung
8:00-8:00 AM	A21A-0105. Evidence of gravity waves in the PBL above New Delhi, during the total Eclipse of August 2009.
	B. Ayara; Y. Ahammed; I. sauvage
8:00-8:00 AM	A21A-0106. Unusual PM episode caused by yellow sand over Taiwan on April 25, 2009 S. Chang; C. Young; H. Lai
8:00-8:00 AM	A21A-0107. Recent Increase in Black Carbon Concentrations from a Mt. Everest Ice Core Spanning 1860-2000 AD S. Kaspari; M. Schwikowski; M. Gysel; P.A. Mayewski; S. Kang; S. Hou
8:00-8:00 AM	A21A-0108. How do the optical properties of Asian aerosols change when they cross the Pacific? E.V. Fischer; D.A. Jaffe
8:00-8:00 AM	A21A-0109. New Discoveries and Insights from Comparisons between Sahara and Asian Dust Using Satellites and a Coupled Climate/Microphysical Model L. Su; O.B. Toon
8:00-8:00 AM	A21A-0110. REMOTE SENSING MEASUREMENTS OF AEROSOL OPTICAL THICKNESS AND CORRELATION WITH IN-SITU AIR QUALITY PARAMETERS DURING A SMOKE HAZE EPISODE IN SOUTHEAST ASIA B. Chew; S.V. Salinas Cortijo; S. Liew
8:00-8:00 AM	A21A-0111. Emission projection and uncertainty analysis of exhaust emissions from global and Asian on-road vehicles F. Yan; E. Winijkul; T. Bond; D.G. Streets
8:00-8:00 AM	A21A-0112. Aerosol Direct and Semi-Direct Effects on Climate in Asia in the NASA GEOS-5 Model C.A. Randles; P.R. Colarco; A. da Silva; A.M. Colarco; R.C. Govindaraju
8:00-8:00 AM	A21A-0113. Dust Storm Reduction due to Precipitation and Temperature Enhancement in Northwestern China: A Direct Climatic Impact of Absorbing Aerosols Y. Gu; K. Liou; W. Chen; H. Liao
8:00-8:00 AM	A21A-0114. Constraining aerosol histories with modeled and observed clear-sky surface solar radiation fluxes in Asia and Europe C. Ruckstuhl; J.R. Norris
8:00-8:00 AM	A21A-0115. SULFATE PRODUCTION IN CLOUDS IN EASTERN CHINA: OBSERVATIONS FROM MT. TAI J.L. Collett; X. Shen; T. Lee; X. Wang; W. Wang; T. Wang
8:00-8:00 AM	A21A-0116. Relating Precipitation Phenomena with MODIS Detected Hot Spots in the Maritime Continent E.M. Ramirez; J.S. Reid; P. Xian; E. Hyer; J. Turk; M. Flatau; C. Zhang

8:00-8:00 AM	A21A-0117. Surface Measurements of dust/local aerosol properties over Northern China during 2008 China-US joined dust field campaign X. Wang; J. Huang
8:00-8:00 AM	A21A-0118. Retrieving optical properties of dusty clouds from MFRSR and Lidar measurements T. Wang; J. Huang
8:00-8:00 AM	A21A-0119. Development of an effective lidar retrieval algorithm using lidar measurements during 2008 China-US joined dust field campaign Z. Huang; J. Huang
8:00-8:00 AM	A21A-0120. Radiative Effect Difference between Multi-layered and Single-layer Cloud Derived from CERES ,CALIPSO and Cloudsat J. Li; Y. Yi; P. Minnis; J. Huang
8:00-8:00 AM	A21A-0121. Field observation analysis of atmosphere-land surface interaction over Loess Plateau of Northwest China G. Wang; J. Huang; W. Guo
8:00-8:00 AM	A21A-0122. Is Anti-Twomey effect real or an artifact? G. Pandithurai; S. Dipu; R.S. Maheskumar; J. Kulkarni; B. Goswami; D. Rosenfeld
8:00-8:00 AM	A21A-0123. Effects of aerosol and greenhouse gasses on the summertime Asian monsoon rainfall trend in the 20th century M. Arai; T. Miyasaka; T. Nozawa; T. Nagashima; M. Kimoto
8:00-8:00 AM	A21A-0124. Spatial and Interspecies Patterns of PM2.5 at Three Sites in the Pearl River Delta, China: One-Year Observations J. Yu; A.K. Lau; C. Wu; W. Ng; Z. Yuan; D. Wu
8:00-8:00 AM	A21A-0125. Seasonal variation of spherical aerosols distribution in East Asia based on ground and space Lidar observation and a Chemical transport model Y. Hara; K. Yumimoto; I. Uno; A. Shimizu; N. Sugimoto; T. Ohara
8:00-8:00 AM	A21A-0126. Spectral Discrimination of Fine and Coarse Mode Aerosol Optical Depth from AERONET Direct Sun Data of Singapore and South-East Asia S. Salinas Cortijo; B. Chew; S. Liew
8:00-8:00 AM	A21A-0127. Carbonaceous and ionic compositions of PM10, PM2.5 and PM1 in Seoul during 2007-2008 J. Han; M. Lee; S. Lim; J. Kim; I. Park; G. Lee; J. Han
8:00-8:00 AM	A21A-0128. A quantitative analysis for nitrogen source-receptor relationships in Northeast Asia based on numerical simulation Y. Kim; Y. Ma; J. Park; K. Choi; I. Chang; J. Kim; Y. Sunwoo
8:00-8:00 AM	A21A-0129. Secondary Organic Aerosol Formation in Urban Air: Temporal Variations and Possible Contributions from Unidentified Hydrocarbons M. Koike; H. Matsui; N. Takegawa; Y. Kondo; R.J. Griffin; Y. Miyazaki

8:00-8:00 AM	Abstract Withdrawn
1:40-2:40 PM	A23A-01. The biggest control knob: carbon dioxide in Earth's climate history (<i>Invited</i>) R.B. Alley
4:00-4:15 PM	A24C-01. Has the Age of Stratospheric Air Decreased over the Past Three Decades (1979-2008)? Q. Fu
4:15-4:30 PM	A24C-02. Sensitivity of the Strength of the Brewer-Dobson Circulation to SST Perturbations in an Aqua-planet Model G. Chen; R.A. Plumb; J. Lu
4:30-4:45 PM	A24C-03. Stratospheric Circulation Changes Inferred From Observations of Ozone and Other Trace Gases E.A. Ray; F.L. Moore; K.H. Rosenlof; J.W. Elkins; A. Engel
4:45-5:00 PM	A24C-04. What factors affect planetary wave driving in the Southern Hemisphere in spring? M. Hurwitz; L. Oman; P.A. Newman; E. Nielsen; J.T. Bacmeister; A.M. Molod
5:00-5:15 PM	A24C-05. Static Stability in the Global Upper Troposphere and Lower Stratosphere: Observations of Long-term Mean Structure and Variability using GPS Radio Occultation Data K.M. Grise; D.W. Thompson; T. Birner
5:15-5:30 PM	A24C-06. Residual circulation trajectories and transit times into the extratropical lowermost stratosphere T. Birner
5:30-5:45 PM	A24C-07. Impacts of Asian Summer Monsoon on Seasonal and Interannual Variations of tropical upper tropospheric pollution over South Asia L. Zhang; Q. Li; J. Jin; N.J. Livesey; J.H. Jiang; M. Luo
5:45-6:00 PM	A24C-08. Transport from the Asian monsoon to the stratosphere observed in satellite measurements of hydrogen cyanide W.J. Randel; M. Park; D.E. Kinnison; L.K. Emmons; P.F. Bernath; K.A. Walker; H.C. Pumphrey

Wednesday, December 16, 2009

8:00 AM-12:20 PM	A31A-0074. On the use of CloudSat and Calipso to Study Clouds Over Sea Ice in the Southern Beaufort Sea. L.M. Candlish; D.G. Barber
8:00-8:00 AM	A31A-0075. The impact of 21st Century sea ice decline on the hydrological budget of the Arctic J.J. Day; J.L. Bamber; P.J. Valdes; J. Kohler
8:00-8:00 AM	A31A-0076. Diurnal albedo of seasonally melting Arctic snow at wavelenghts of UV and visible O. Meinander
8:00-8:00 AM	A31A-0077. Impact of including fully interactive Greenland and Antarctic ice sheets on the climate sensitivity of an Earth system model of intermediate complexity H. Goelzer; M. Loutre; A. Mouchet; P. Huybrechts; T. Fichefet; H. Goosse
8:00-8:00 AM	A31A-0078. Simulations of Vegetation Impacts on Arctic Climate C. Bonfils; T.J. Phillips; W.J. Riley; W.M. Post; M.S. Torn
8:00-8:00 AM	A31A-0079. Investigating the effects of Eurasian snow cover on winter climate R.J. Allen; C.S. Zender
8:00-8:00 AM	A31A-0080. Exploring the Role of Sea-Ice for Seasonal Forecasts R. Senan; R.E. Benestad; M. Balmaseda; L. Ferranti; Y.J. Orsolini; A. Melsom
8:00-8:00 AM	A31A-0081. A comparison of polar vortex trend response to Pacific and Indian Ocean warming S. Li
8:00-8:00 AM	A31A-0082. Modulation in Interannual Sea-Ice Patterns in the Southern Ocean in Association with Large-Scale Atmospheric Mode Shift Y. Udagawa; K. Yamazaki; Y. Tachibana
8:00-8:00 AM	A31A-0083. Atmospheric forcing of Antarctic sea ice J.A. Renwick
8:00-8:00 AM	A31A-0084. The Feedback of Sea Ice Loss on Arctic Temperatures: 1970-2008 M. Hoerling; J. Perlwitz; J. Eischeid; A. Kumar
8:00-8:00 AM	A31A-0085. The Energetics of a Warming Arctic T. Graham; A. Ferraro; M. Vellinga; P. Wu
8:00-8:00 AM	A31A-0086. Very large warming in Antarctic mid-troposphere in winter: PSCs are implicated. T.A. Lachlan-Cope; H.K. Roscoe
8:00-8:00 AM	A31A-0087. On the Interactions of Arctic Sea Ice, Cloud Cover, and Surface Temperature from Satellite Observations
	Y. Liu; J.R. Key; X. Wang
8:00-8:00 AM	A31A-0088. Atmosphere-sea ice interactions over a wide range of climates I.L. Eisenman; T. Schneider; D. Battisti; C.M. Bitz

8:00-8:00 AM	A31A-0089. Parameterization of the Solar Fluxes over Mountain Surfaces for Application to Regional Climate Models W. Lee; K. Liou; A. Hall
8:00-8:00 AM	A31A-0090. Effects of cryospheric changes on Earth's solar energy budget M. Flanner
8:00-8:00 AM	A31A-0091. An Improved Method For Retrieving TOA Albedo Over Polar Regions Using MISR J. Corbett; R. Davies
8:00-8:00 AM	A31A-0092. Storm-induced polar warming and moistening under anthropogenic forcing J. KUG; D. Choi; F. Jin; W. Kwon; H. Ren
8:00-8:00 AM	A31A-0093. Modulation of Orbital Induced Climate Change by Ice, Cloud and Water Vapor Feedbacks D.F. Mantsis; A.C. Clement; A.J. Broccoli
8:00-8:00 AM	A31A-0094. The rapidly shrinking sea ice cover and ice-albedo feedback effects J.C. Comiso
8:00-8:00 AM	Abstract Withdrawn
8:00-8:15 AM	A31I-08. Overview Of The Development And Evaluation Of The NCEP Global Aerosol Modeling System: Particulates From Biomass Burning H. Huang; S. Lu; Y. Tang; D. Kim; M. Tsidulko; C. Tassone; J. Huang; J. McQueen; B. Lapenta; G. DiMego; S. Lord; A. Da Silva; M. Chin; T.L. Diehl
8:15-8:30 AM	A31I-02. Improve MISR's Capability of Predicting Ground Level PM2.5 Concentrations with Observed Aerosol Vertical Profiles (Invited) Y. Liu; Z. Wang
8:30-8:45 AM	A31I-03. Use of meteorological data to improve PM2.5 and AOD relationships in a western US valley M.C. Green; J.G. Watson; J.C. Chow
8:45-9:00 AM	A31I-04. Inferring the composition and concentration of aerosols by combining the AERONET, MPLNET and CALIOP data: comparison with in-situ measurements and utilization to evaluate and improve GCM results. D. Ganguly; P.A. Ginoux; V. Ramaswamy
9:00-9:15 AM	A31I-05. Experiments with data assimilation in comprehensive air quality models: Impacts on model predictions and observation requirements (<i>Invited</i>) R. Mathur
9:15-9:30 AM	A31I-06. Modeling evolution of aerosol mixing state and the associated optical and CCN activation properties (<i>Invited</i>) R.A. Zaveri; R.C. Easter; J. Barnard; N.S. Riemer; M. West; A.P. Ault; K.A. Prather

9:30-9:45 AM	A31I-07. The Aerosol Modeling Testbed: A Tool to Facilitate Improved Aerosol Process Modules J.D. Fast; W.I. Gustafson; E.G. Chapman; R.C. Easter; J. Rishel
19:45-10:00 AM	A31I-01. Satellite Remote Sensing of Particulate Matter Air Quality: Progress, Potential and Pitfalls (<i>Invited</i>) S.A. Christopher
1:40 PM-6:00 PM	A33C-0259. Effective Lidar Ratios of Dense Dust Aerosol Layers over North Africa Observed by the CALIPSO Lidar Z. Liu; D.M. Winker; A.H. Omar; M. Vaughan; C.R. Trepte; Y. Hu; C.A. Hostetler; W. Sun; B. Lin
1:40-1:40 PM	A33C-0260. Enhanced Aerosol Backscatter in the Vicinity of Marine Boundary Layer Clouds Revealed by Satellite-Based Lidar, CALIPSO J.L. Tackett; L. Di Girolamo
1:40-1:40 PM	A33C-0261. Development and Initial Testing of a Multi-Sensor Platform for Cloud-Aerosol Interactions in the Lower Troposphere A.R. Nehrir; D.S. Hoffman; K.S. Repasky; B. Todt; T. Sharpe; C. Half Red; J.L. Carlsten
1:40-1:40 PM	A33C-0262. Influence of humidified aerosol on lidar depolarization observed during SHEBA A.M. Fridlind; B. van Diedenhoven; A.S. Ackerman
11:40-1:40 PM	A33C-0263. Measuring Plume Meander in the Nighttime Stable Boundary Layer with Lidar A. Hiscox; D.R. Miller; C.J. Nappo
1:40-1:40 PM	A33C-0264. An automatic Planetary Boundary Layer height retrieval method with compact EZ backscattering Lidar in the frame of ICOS campaign S. Loaec; S. Lolli; I. sauvage; M. Boquet; I. Xueref-Remy
11·40-1·40 PM	A33C-0265. Active methods to measure multilayer Planetary Boundary Layer Dynamics Y. Wu; B. Gross
1:40-1:40 PM	A33C-0266. Lidar Approach in Estimating Particulate Mass Emissions from a Poultry Production Facility P.A. Lewandowski; W.E. Eichinger; J.H. Prueger; J. Hatfield
1:40-1:40 PM	A33C-0267. Automated Raman lidar for day and night operational observation of water vapor for meteorological applications T. Dinoev; Y. Arshinov; S. Bobrovnikov; I. Serikov; B. Calpini; P. Ristori; H. van den Bergh; V. Simeonov
1:40-1:40 PM	A33C-0268. Very high spatial and temporal lidar for moisture and temperature monitoring F. Martin; I. Serikov; P. Ristori; C.W. Higgins; M.B. Parlange; V. Simeonov
1:40-1:40 PM	Abstract Withdrawn

1:40-1:40 PM	A33C-0270. Aerosol remote sensing over land using AATSR G. De Leeuw; P. Kolmonen; L. Sogacheva; A. Sundstrom; E. Rodriguez
1:40-1:40 PM	A33C-0271. Diurnal variation of column NO ₂ observed from space L.C. Valin; A.R. Russell; R.C. Hudman; A.K. Mebust; R.C. Cohen
1:40-1:40 PM	A33C-0272. Validation of GOME-2 UV radiance Z. Cai; X. Liu; C.R. Nowlan; K. Chance; Y. Liu
1:40-1:55 PM	A33E-01. Stochastic Modeling of Water Vapor in the Climate System B. Chen; J. Duan; R. Pierrehumbert; X. Huang
1:55-2:10 PM	A33E-02. Water vapor and OLR feedbacks over the recent ENSO cycle A.L. Kursinski; E.R. Kursinski; C.O. Ao
2:10-2:25 PM	A33E-03. Investigating the Water Vapor Component of the Greenhouse Effect from the Atmospheric InfraRed Sounder (AIRS) A. Gambacorta; C. Barnet; F. Sun; M. Goldberg
2:25-2:40 PM	A33E-04. Consistent Differences in Climate Feedbacks between Coupled and Atmospheric GCMs K.M. Shell
2:40-2:55 PM	A33E-05. Observationally Based Estimates of Climate Feedbacks N.D. Gordon; P. Forster
2:55-3:10 PM	A33E-06. Mechanisms for the weakening of tropical circulation C. Chou; C. Chen
3:10-3:25 PM	A33E-07. An Observed Tropical Oceanic Radiative-Convective Cloud Feedback M.D. Lebsock; G.L. Stephens; C.D. Kummerow
3:25-3:40 PM	A33E-08. Interrelations of AIRS/AMSU-derived anomalies and trends of temperature, water vapor, clouds and OLR G.I. Molnar; J. Susskind
4:45-5:15 PM	A34C-03. Research activity of the greenhouse gas measurements using optical remote sensing in Japan (<i>Invited</i>) K. Asai
5:15-5:30 PM	A34C-04. Airborne Validation of Active CO ₂ LAS Measurements E.V. Browell; J. Dobler; S. Kooi; Y. Choi; F. Harrison; B. Moore; T. Zaccheo
5:30-5:45 PM	A34C-05. Pulsed Airborne Lidar measurements of Atmospheric CO2 Column Absorption and Line Shapes from 3-13 km altitudes J.B. Abshire; H. Riris; G.R. Allan; C.j. Weaver; W.E. Hasselbrack; X. Sun

4:00-4:20 PM	A34D-01. Isotopic Tracers to Identify Far-traveled Pollutant and Mineral Aerosols in Northern California (<i>Invited</i>) D.J. Depaolo; J.N. Christensen; S.A. Ewing; S.S. Cliff; S.T. Brown; R.A. VanCuren
5:00-5:15 PM	A34D-04. Summertime Ozone over California: A Model-Measurement Analysis across Scales G. Pfister; L.K. Emmons; C. Wiedinmyer; D.P. Edwards
5:15-5:30 PM	A34D-05. Oxygenated VOCs as indicators of regional-scale photochemistry: comparison of CMAQ model results with measurements from the UC Blodgett Forest Station, California A.J. Huisman; J.P. DiGangi; A. Kammrath; S.B. Henry; A.G. Carlton; F. Keutsch
5:30-5:45 PM	A34D-06. Measurements of Greenhouse Gases around the Sacramento Area: The Airborne Greenhouse Emissions Survey (AGES) Campaign A. Karion; M.L. Fischer; J.C. Turnbull; C. Sweeney; I.C. Faloona; N. Zagorac; T.P. Guilderson; S. Saripalli; T. Sherwood

Thursday, December 17, 2009

8:00 AM-12:20 PM	A41A-0073. Aerosol Precursor Emissions, Secondary Aerosol Production, and Climate-Forcing Gas Exchange in the Midwestern United States P.V. Doskey
8:00-8:00 AM	A41A-0074. Characterizing CH ₄ and N ₂ O Fluxes from a Soybean/Corn Ecosystem in Minnesota X. Zhang; X. Lee; T.J. Griffis; J.M. Baker; M. Erickson; N. Hu; W. Xiao
8:00-8:00 AM	A41A-0075. Quantification of Terpenes by 1DGC-MS and 2DGC-TOF -MS R.M. Flores; J.A. Perlinger; P.V. Doskey
8:00-8:00 AM	A41A-0076. Aerosol Production from the Great Lakes Surface J.H. Slade; G. Mwaniki; S.B. Bertman; T.M. VanReken; P.B. Shepson
4:00-4:20 PM	A44D-01. A Methylmercury Prediction Too For Surface Waters Across The Contiguous United States (<i>Invited</i>) D.P. Krabbenhoft; N. Booth; M. Lutz; M.N. Fienen; T. Saltman

4:20-4:40 PM	A44D-02. Mercury Isotopic Evidence for Contrasting Mercury Transport Pathways to Coastal versus Open Ocean Fisheries (Invited) J.D. Blum; D.B. Senn; E.J. Chesney; M.S. Bank; A. Maage; J.P. Shine
4:40-4:55 PM	A44D-03. Evidence for the free troposphere as a source of atmospheric mercury measured in Reno, Nevada, U.S.A. M.S. Gustin; S.N. Lyman
4:55-5:10 PM	A44D-04. Global source-receptor relationships for mercury under present and year 2050 anthropogenic emissions scenarios <u>E.S.</u> Corbitt; C. Holmes; D.J. Jacob; D.G. Streets; N.E. Selin; A. Sorensen; E.M. Sunderland
5:10-5:25 PM	A44D-05. Lake Recovery Following Mercury Deposition Changes L. Levin; K. Lohman
5:25-5:40 PM	A44D-06. Production and Cycling of Methylated Mercury Species in Arctic Marine Waters I. Lehnherr; V.L. St.Louis; H. Hintelmann
5:40-5:55 PM	A44D-07. Observations of iodine oxide and reactive gaseous mercury at a coastal site in Pensacola, FL S. Coburn; B.K. Dix; R. Sinreich; A.F. Terschure; E.S. Edgerton; R. Volkamer
5:55-6:00 PM	Poster Introductions Brief introduction of poster topics

Friday, December 18, 2009

1:40-1:55 PM	A53D-01. Enhanced clear sky reflectance near clouds: What can be learned from it about aerosol properties? A. Marshak; T. Varnai; G. Wen; J. Chiu
1:55-2:10 PM	A53D-02. Emissions of Black Carbon Particles from Biomass Burning and Their Physical and Chemical Properties Y. Kondo; L. Sahu; N. Moteki; N. Takegawa; Y. Zhao; S.A. Vay; G.S. Diskin; A. Wisthaler; L.G. Huey; J.L. Jimenez
2:10-2:25 PM	A53D-03. Ammonium Nitrate Formation near the Colorado Front Range A.M. Middlebrook; R. Bahreini; C.A. Brock; S.S. Brown; J. Cozic; G.J. Frost; A.O. Langford; B.M. Lerner; B. Matthew; S.A. McKeen; J. Neuman; J.B. Nowak; J.W. Peischl; P. Quinn; T.B. Ryerson; K. Schultz; H. Stark; M. Trainer; N. Wagner; E.J. Williams; A.G. Wollny

2:25-2:40 PM	A53D-04. Black carbon measurements in the Pearl River Delta region of China X. Huang; R. Gao; J.P. Schwarz; H. Ling-Yan; D.W. Fahey; W. Laurel A; L. Zeng
2:40-2:55 PM	A53D-05. The impact of the 1783-84 AD Laki Eruption on Aerosol Formation Processes and Cloud Condensation Nuclei A. Schmidt; K.S. Carslaw; G.W. Mann; M.B. Wilson; T.J. Breider; S.J. Pickering
2:55-3:10 PM	A53D-06. The Role of Ions in New Particle Formation in the Upper Troposphere and Lower Stratosphere Using WACCM/CARMA J.M. English; O.B. Toon; M.J. Mills; F. Yu
3:10-3:25 PM	A53D-07. Multisite reconciliation of sub- and supersaturated particle water uptake. M. Irwin; N.A. Good; J. Crosier; D.O. Topping; J.D. Allan; H. Coe; T.W. Choularton; G.B. McFiggans
3:25-3:40 PM	A53D-08. In situ vertical profiles of black carbon aerosol over the Pacific, Arctic, and Antarctic Regions (80°N to 67°S Latitudes) J.P. Schwarz; J.R. Spackman; R. Gao; W. Laurel A; P. Stier; S.M. Davis; D.W. Fahey

Formation of UV-vis Absorbing Organic Solutes, Films, and Suspended Precipitates in Sulfuric Acid Solutions at Stratospheric Aerosol Acidities

A. L. Van Wyngarden; ¹; C. L. Belle; ^{5, 6}; C. L. Dalle ore; ⁴; M. J. Morrissey; ⁷; J. M. Rodgers; ³; L. T. Iraci; ²;

- 1. Department of Chemistry, San Jose State University, San Jose, CA, United States.
- 2. Atmospheric Science Branch, NASA Ames Research Center, Moffett Field, CA, United States.
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- 4. Dartmouth College, Hanover, NH, United States.
- 5. NASA Undergraduate Student Research Program, NASA Ames Research Center, Moffett Field, CA, United States.
- 6. Colorado College, Colorado Springs, CO, United States.
- 7. University of California, Berkeley, Berkeley, CA, United States.

Body: The stratospheric aerosol layer has traditionally been thought of as being composed of pure sulfuric acid/water aerosols, but recent airborne measurements of single particle composition (e.g. Murphy et al. 2007) have shown that 10-20% of sampled aerosols in the lower stratosphere have significant organic content. Since there is very little water vapor in the stratosphere, these sulfuric acid/organic particles will be extremely acidic (40-80 wt% sulfuric acid). Here we show that small amounts (approx. 0.1wt%) of carbonyl compounds allowed to react with such concentrated sulfuric acid solutions produce highly-colored solutions that continue to darken for months after mixing. We examined propanal, methylglyoxal, and glyoxal singly as well as in various combinations. In the case of propanal, colored organic films form on the surfaces of the solutions on a timescale of days to weeks depending on acidity. Solutions of various compositions also formed precipitates suspended near the liquid surface. UV-vis absorption spectra of the colored solutions will be presented along with kinetics of film formation as a function of acidity, temperature, and organic content. These results will be used to assess whether it is possible that such films could exist in the lower stratosphere (or upper troposphere) in quantities that would be significant enough to impact the direct and/or indirect climate forcing of these aerosols and/or satellite retrievals of atmospheric species that employ UV-vis wavelengths.

Contact Information: Contact Information

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Toward Investigating Optically Trapped Organic Aerosols with CARS Microspectroscopy

L. F. Voss; 1;

1. Bowdoin College, Brunswick, ME, United States.

Body: The Intergovernmental Panel on Climate Change notes the huge uncertainty in the effect that atmospheric aerosols play in determining overall global temperature, specifically in their ability to nucleate clouds. To better understand aerosol chemistry, the novel coupling of gradient force optical trapping with broad bandwidth coherent anti-Stokes Raman scattering (CARS) spectroscopy is being developed to study single particles suspended in air. Building on successful designs employed separately for the techniques, this hybrid technology will be used to explain how the oxidation of organic compounds changes the chemical and physical properties of aerosols. By trapping the particles, an individual aerosol can be studied for up to several days. Using a broad bandwidth pulse for one of the incident beams will result in a Raman vibrational spectrum from every laser pulse. Combined with signal enhancement due to resonance and coherence of nonlinear CARS spectroscopy, this technique will allow for acquisition of data on the millisecond time scale, facilitating the study of dynamic processes. This will provide insights on how aerosols react with and absorb species from the gas phase. These experiments will increase understanding of aerosol oxidation and growth mechanisms and the effects that aerosols have on our atmosphere and climate. Progress in efforts developing this novel technique to study model systems is presented.

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OH Oxidation Reactions of Unsaturated Self-assembled Monolayers on Germanium Surfaces: A System That Mimics Organics Adsorbed on Urban Surfaces

S. G. Moussa; 1; J. Raff; 1; B. J. Finlayson-Pitts; 1;

1. Chemistry, University of California, Irvine, Irvine, CA, United States.

Body: OH radical is a major oxidant of organics in the atmosphere, including unsaturated hydrocarbons. Alkenes are ubiquitous in the troposphere, both in the gas phase and adsorbed on surfaces such as buildings and in and on aerosol particles. While a great deal is known about the kinetics and mechanisms of OH-alkene reactions in the gas phase, much less is known about the chemistry on surfaces and in and on particles. In this study, terminal alkene self assembled monolayers (SAMs) were used as proxies for organics adsorbed on surfaces of buildings and airborne dust particles. The SAMs were covalently bound to the surface of an infrared-transmitting Ge crystal. OH was generated in air above the SAM by photolyzing different precursors in a newly designed and constructed cell. The reaction of the SAM with OH was then followed in real time using Fourier transform infrared spectroscopy at 1 atm pressure in air and at 298 K. The OH radicals were generated using photolysis of three different precursors: 1) isopropyl nitrite, 2) ozone in the presence of water vapor and 3) hydrogen peroxide. The first source produces OH radicals in the presence of nitrogen oxides. In the second case, there is competition between O3 and OH for reaction with the alkene SAM. The third source constitutes the cleanest way of generating OH radicals, and is most relevant to low NOx and remote conditions. Results from the experiments using these three OH sources will be compared and the implications for reactions of surface-adsorbed alkenes in air are discussed.

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Atmospheric Processing of Methylglyoxal and Glyoxal in Aqueous Environments

J. L. Axson; ¹; V. Vaida; ¹;

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Body: Methylglyoxal is a known oxidation product of biogenic and anthropogenic VOCs, being observed by field studies and incorporated into atmospheric models. While the gas-phase chemistry of this compound is fairly well understood, its modeled concentration and role in SOA formation remains controversial. Methylglyoxal, like other aldehydes and ketones, when in the presence of water is hydrated to form alcohols. When the alcohol forms, it is likely to partition into clouds and aerosols because of its tendency to form intermolecular hydrogen bonds. This study evaluates the water-mediated equilibrium between methylglyoxal and its hydrates in the gas phase. The results can be used to understand the atmospheric fate and processing of this and similar organics. My experimental approach employs Fourier-Transform spectroscopy to characterize gas phase reagents and products as a function of relative humidity and obtain an equilibrium constant between methylglyoxal and its hydrate.

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Role of Ozone in Particle Formation and Growth From the Nitrate Radical-Initiated Oxidation of α-Pinene

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Body: The three major atmospheric oxidants involved in SOA formation from biogenic volatile organic compounds are O3, OH and nitrate radical (NO3). While O3 and OH-initiated oxidation occur during the day, NO3 radical-initiated oxidation is recognized to be a major contributor to the night-time chemistry of volatile organic compounds in the troposphere. Specifically, the reaction of biogenic hydrocarbons with NO3 is relatively fast (for example, lifetime of α -pinene is only ~11 min at 2.5 x 10^8 NO3 molecules cm-3), so this reaction is expected to be a major sink of the organics at night, and also to contribute to the removal of NOx from the atmosphere. Previous studies by other groups have shown that SOA yield from the NO3 reaction is relatively small due to the high volatility of the reaction products, in contrast to the O3 reaction which is an important SOA source in the atmosphere.

We report the results of experiments designed to probe the influence of ozone on particle formation and composition in the NO3 oxidation of α -pinene. The experiments were performed in a large diameter flow tube using the reaction of NO2 with O3 as the source of NO3 radicals. Particle size distributions were measured using a scanning mobility particle sizer (SMPS) and an aerodynamic particle sizer (APS). Real-time aerosol mass spectrometers (SPLAT-II and AMS) as well as integrated collection of particles on ZnSe impaction disks and quartz fiber filters were used to characterize the chemical composition of the particles. Organic nitrates were major components of the particles at high ratios of NO2/O3 but decreased as this ratio decreased. The particle size distribution also shifted. In addition, the formation of other species with low vapor pressures such as carboxylic acids was observed due to the increasing contribution from the O3-initiated oxidation of α -pinene. The role of ozone as a source of nucleation in this system will be discussed.

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Laboratory Investigation of Trace Gas Emissions from Biomass Burning on DoD Bases I. R. Burling; 1; R. J. Yokelson; 1; D. W. Griffith; 2; J. M. Roberts; 3; P. R. Veres; 3, 4; C. Warneke; 3, 4; T. J. Johnson: 5:

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Body: Vegetation representing fuels commonly managed with prescribed fires was collected from five DoD bases and burned under controlled conditions at the USFS Firelab in Missoula, MT. The smoke emissions were measured with a large suite of state-of-the-art instrumentation. Seventy-seven fires were conducted and the smoke composition data will improve DoD land managers' ability to assess the impact of prescribed fires on local air quality. A key instrument used in the measurement of the gas phase species in smoke was an open-path FTIR (OP-FTIR) spectrometer, built and operated by the Universities of Montana and Wollongong. The OP-FTIR has to date detected and quantified 20 gas phase species – CO_2 , CO, H_2O , N_2O , NO_2 , NO, HONO, NH_3 , HCI, SO_2 , CH_4 , CH_3OH , and CH_3OH , CH_3OH , HCHO, HCOOH, C₂H₂, C₂H₄, CH₃COOH, HCN, propylene and furan. The spectra were analyzed using a nonlinear least squares fitting routine that included reference spectra recently acquired at the Pacific Northwest National Laboratories. Preliminary results from the OP-FTIR analysis are reported here. Of particular interest, gas-phase nitrous acid (HONO) was detected simultaneously by the OP-FTIR and negative-ion proton-transfer chemical ionization spectrometer (NI-PT-CIMS), with preliminary fire-integrated molar emission ratios (relative to NOx) ranging from approximately 0.03 to 0.20, depending on the vegetation type. HONO is an important precursor in the production of OH, the primary oxidizing species in the atmosphere. There existed little previous data documenting HONO emissions from either wild or prescribed fires. The non-methane organic emissions were dominated by oxygenated species, which can be further oxidized and thus involved in secondary aerosol formation. Elevated amounts of gasphase HCl were also detected in the smoke, with the amounts varying depending on location and vegetation type.

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Effect of photosensitized chemistry on organic aerosol evolution

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Body: Photochemistry in aerosol particles is an emerging new field of atmospheric science. Up to now, photochemical processes in the condensed phase of atmospheric aerosol particles are not well understood. Primary and secondary compounds in the gas and aerosol phase continuously interact and change phase during their or their descendants life time in the atmosphere. Partially oxised aromatic compounds such as aromatic ketones may act as photosensitizer to promote charge and energy transfer to other organic compounds under conditions, where direct photolysis processes of the latter are not possible. The resulting radicals undergo numerous secondary chemical reactions, some of which may lead to polymerization.

In this study we show that the presence of a photosensitizer in the aerosol phase leads to significant processing of simple organic compounds such as small organic acids due to exposure to simulated sunlight. The aerosol flow tube experiments were performed in a photoreactor, which was coupled to a chemical ionization mass spectrometer and a scanning mobility particle sizer. For some experiments an aerosol mass spectrometer was also used. We used ammonium sulfate and organic acids as matrix and Benzoyl Benzoic Acid (BBA) as sensitizer. BBA is a well known photosensitizer absorbing in the UV. The results will be shown and discussed.

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Development of a Metastable Atom Bombardment (MAB) Source for Penning Ionization Time-of-flight Aerosol Mass Spectrometry

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Body: The Aerodyne time-of-flight aerosol mass spectrometer (ToF-AMS) utilizes thermal vaporization followed by electron ionization (EI) to convert aerosol components to gas-phase ions. The method enables quantification of chemical classes, but the extensive fragmentation caused by EI limits the specificity of both chemical analysis and source identification by factor analysis. To better identify the molecular components of aerosols, we have constructed a metastable atom bombardment (MAB) ionization source that can be interfaced to standard ToF-AMS hardware. A beam of metastable rare gas atoms is produced by a low-voltage DC discharge and focused toward the vaporization plume, yielding Penning Ionization of the analyte molecules. By changing gases, the excited energies of the metastables can be adjusted between 20.61 eV (He) and 9.92 eV (Kr). Source parameters, including pressures, current, geometry, and materials, were optimized for He, Ar, and Kr. Instrument sensitivity and induced fragmentation was characterized for each using lab-generated oleic acid particles. The demonstrated sensitivities are 0.1% of EI (3% of the SNR of EI in the V-mode, comparable to the Q-AMS SNR), which is sufficient for ambient monitoring. A metastable flux of 2.6e14 sr-1sec-1 has been achieved. The MAB-AMS has been deployed to the FLAME-3 campaign at the USDA Fire Sciences Laboratory in Missoula, MT, and used to sample smoke from open burning of different biomass samples. Preliminary results from FLAME-3 will be presented.

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Laboratory Studies of the Heterogeneous Oxidation of Levoglucosan in Biomass Burning Particles C. J. Hennigan; ¹; A. Sullivan; ²; J. L. Collett; ²; A. L. Robinson; ¹;

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Body: Biomass burning is thought to be an important source of atmospheric aerosol. Levoglucosan (1,6-anhydro-β-Dglucopyranose) is a major product of cellulose combustion and is used extensively as a tracer to infer the contribution of biomass burning to measured aerosol concentrations. Chemical mass balance models used to apportion measured aerosol concentrations to different sources currently assume that compounds such as levoglucosan are inert and nonvolatile. However, recent laboratory and field-based studies have found that primary organic compounds undergo significant reaction when exposed to oxidants at typical atmospheric levels. We have conducted smog chamber experiments to investigate the heterogeneous oxidation of levoglucosan in particles emitted from wood combustion. Wood smoke from hard wood fires and particles generated from wood smoke extract were injected into a smog chamber at initial aerosol concentrations (~20-200 µg/m3) representative of conditions within biomass burning plumes. The primary particles and vapors were then exposed to UV lights, which initiated photo-chemistry. Significant SOA formation was observed in the wood smoke experiments while a net decrease in particle mass occurred in the wood smoke extract experiments. Hydroxyl radical (OH) levels were typically between 3-6 x 10⁶ molecules/cm3, which are representative of summertime and biomass plume conditions. Levoglucosan concentrations were determined from filter samples analyzed offline using high performance anion exchange chromatography with pulsed amperometric detection. In every experiment, levoglucosan decay between 20 and 85% was observed. This decay could have resulted from gas-phase reactions and the re-partitioning of levoglucosan to the gas phase, however, heterogeneous oxidation by reaction with OH was the most likely cause. The variability in levoglucosan decay was due to differences in OH levels and exposure times; oxidation rates across experiments were similar. The range of oxidant exposures represented aging in the atmosphere of about 0.5-3 days, demonstrating that the reaction of levoglucosan is likely an important removal process on time scales relevant to particle lifetimes. Finally, the kinetic data were used to estimate potential biases in receptor model estimates of biomass burning aerosol concentrations.

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Kinetics and Products of Heterogeneous Oxidation of Erythritol and Levoglucosan in Aerosol Particles S. H. Kessler; ¹; J. H. Kroll; ^{2, 3}; K. R. Wilson; ⁴; J. D. Smith; ⁴;

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Body: Although organic aerosols in the atmosphere have been implicated in concerns related to both human health and global radiative forcing, they remain collectively a significant source of uncertainty in long-term predictions, in part because of the inherent chemical complexity of possible oxidation products formed from a given compound during its atmospheric lifetime. Here we study the heterogeneous oxidation of model compounds used as surrogates for biomass burning aerosol and secondary organic aerosol (SOA): levoglucosan, a frequently used tracer for biomass burning, and erythritol ((2R,3S)-butane-1,2,3,4-tetraol) an analog of the methyltetrols found in isoprene oxidation SOA. The present experiments are aimed at examining the kinetics and products of further oxidation of both compounds, in order both to explore how each compound contributes to atmospheric aerosol formation and to examine model singlecomponent systems to determine how structural and compositional differences between compounds affect the relative paths of oxidative degradation. Particles are sent through a flow tube reactor where they are exposed to high concentrations (~10¹³ molecule ¹ s ¹ cm⁻³) of hydroxyl radicals (OH), after which the aerosols are sized and their composition analyzed using a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) with both electron impact (EI) and vacuum-ultraviolet (VUV) ionization techniques. Although erythritol and levoglucosan have similar second-order degradation rate constants $(2.03 \pm 0.20 \times 10^{-13})$ and $4.7 \pm 0.5 \times 10^{-13}$ cm³ molecule $^{-1}$ s⁻¹, respectively), the differences between the loss of particle mass upon an equivalent amount of oxidation (80% vs 30% respectively) are much more pronounced.

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Gas-Phase and Particle-Phase Reaction and Kinetics of Epoxydiols from Photooxidation of Isoprene

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Body: The emission of isoprene from deciduous plants is estimated to be upwards of 500 Tg a year and plays an important role in tropospheric chemistry over large regions of the globe. Both laboratory and field measurements have found evidence of large amounts of isoprene derivatives in secondary organic aerosols (SOAs). SOA formation has been shown to be favored under low-NOx conditions, producing aerosols with high concentrations of tetrols, sulfate esters, and C5 repeating polymeric species. The exact gas phase chemical precursors of the organics found in these SOAs are still debated. Recently, it has been shown that under low-NOx conditions the photooxidation of isoprene will result in the formation of isoprene epoxydiols. We have studied the gas-phase, aerosol-phase, and liquid-phase reactivity of a number of alkane epoxydiols. Experiments on the production of epoxydiols from the photooxidation of isoprene and butadiene as well as the uptake of these species by aerosol seed particles were performed in the Caltech dual 28-m3 environmental chamber. The gas-phase loss of the epoxydiols by aerosol uptake was monitored by chemical ionization mass spectrometry along with the products of the photooxidation of the epoxydiols. The organic composition of aerosols from the uptake of epoxydiols as well as from the photooxidation of isoprene was determined by a number of mass spectrometry techniques from chamber filter samples. Finally, the products and kinetics of the liquid-phase acid hydrolysis of the epoxides was determined by NMR. The epoxydiols were found to be readily incorporated into acidic aerosols by reactive uptake as well as partitioning. The organic composition of the SOAs from uptake of epoxydiols or the photooxidation of isoprene by acidic seeds were found to be very similar to the products of the acidic hydrolysis of the epoxydiols consisting of tetrols, sulfate esters, and polymeric species.

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Improving and assessing vapour pressure estimation methods for organic compounds of atmospheric relevance using a Knudsen Effusion Mass Spectrometer (KEMS)

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Body: Aerosol particles influence climate directly through the scattering and absorbing radiation and indirectly through their role as cloud condensation nuclei (CCN). Traditionally, models aiming to capture the behaviour of aerosols in the atmosphere have concentrated on the role of inorganic compounds. However, organic components, covering a huge range of chemical and physical properties (Jacobson et.al., 2000), may constitute a significant fraction depending on location (Houghton et.al., 2001).

Knowledge of pure component vapour pressures is essential for calculations of gas/particle partitioning. There are many methods of estimating vapour pressures but most of the experimental data collected to date has been for intermediate or high pressure compounds (and often measured at temperatures considerably above ambient) and the proportion of experimental data for low (less than 100Pa) vapour pressure compounds has been very small. Hence the datasets used for developing the estimation methods have reflected this bias in addition to the fact that components studied tend to have one or two functional groups at the most. Thus it is unsurprising that some of the estimation methods can give errors in vapour pressure of several orders of magnitude for multifunctional compounds at ambient temperatures.

Knudsen Effusion Mass Spectrometer (KEMS) has been used to measure solid state vapour pressures for multifunctional organic compounds based on dicarboxylic acids (Booth et al 2009). In the atmosphere these compounds are likely to exist in the sub-cooled state so Differential Scanning Calorimetry (DSC) was used to obtain thermochemical data to effect a correction between solid and sub-cooled vapour pressures. The group contribution method of Nanoolal and co-workers (Nanoolal et al., 2008) is one of the best predictive methods in terms of reproducing available low volatility vapour pressure data (barley et al., 2009). The Nanoolal method relies on the use of primary and secondary functional groups and interaction parameters, derived from experimental data, to reliably predict boiling points and vapour pressures. A sensitivity study was undertaken to establish the impact of the new experimentally determined vapour pressures on partitioning models.

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Heterogeneous Reactions of Surface-Adsorbed Catechol: A Comparison of Tropospheric Aerosol Surrogates.

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Body: Surface-adsorbed organics can alter the chemistry of tropospheric solid-air interfaces, such as aerosol and ground level surfaces, thereby impacting photochemical cycles and altering aerosol properties. The nature of the surface can also influence the chemistry of the surface-adsorbed organic. We employed diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) to monitor the adsorption of gaseous catechol on several tropospheric aerosol surrogates and to investigate the subsequent reactivity of adsorbed-catechol with nitrogen dioxide and, in separate preliminary experiments, ozone. Graphite, kaolinite, and sodium halide (NaF, NaCl, NaBr) powders served as carbonaceous, mineral and sea salt aerosol surrogates, respectively. Broad OH stretching bands for adsorbed catechol shifted to lower wavenumber with peak frequencies following the trend NaBr > NaCl > NaF ≈ kaolinite, consistent with the increasing basicity of the halide anions and basic Brønsted sites on kaolinite. The dark heterogeneous reaction of NO₂ with NaCl-adsorbed catechol at relative humidity (RH) <2% promoted nitration forming 4-nitrocatechol and oxidation forming 1,2-benzoquinone and the ring cleavage product muconic acid, with product yields of 88%, 8%, and 4%, respectively. 4-Nitrocatechol was the dominant product for catechol adsorbed on NaF and kaolinite, while NaBr-adsorbed catechol produced less 4-nitrocatechol and more 1,2-benzoquinone and muconic acid. For all three sodium halides, the reactions of NO₂ with adsorbed catechol were orders of magnitude faster than between NO_2 and each NaX substrate. 4-Nitrocatechol rates and product yields were consistent with the relative ability of each substrate to enhance the deprotonated nature of adsorbed-catechol. Increasing the relative humidity caused the rate of each product channel to decrease and also altered the product branching ratios. Most notably, 1,2benzoquinone formation decreased significantly even at 13% RH. The dramatic reactivity of surface-adsorbed catechol contrasts prior observations which found thin films of pure catechol unreactive with NO2, indicating that thin films do not always serve as reliable models for surface-adsorbed species.

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Heterogeneous Reactions of NO3 and N2O5 with a Range of Organic Substrates

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Body: Although NO3 and N2O5 are important nighttime species in the troposphere, reactions between these gasphase species and organic particles remain unexplored with few exceptions. These heterogeneous reactions may be
important for several reasons. For example, these reactions may be a loss pathway for condensed-phase organic
compounds in the atmosphere, which has implications for source apportionment. We have investigated
heterogeneous reactions between NO3 and N2O5 and several organic surfaces and films such as organic selfassembled monolayers (SAMs), solid polycyclic aromatic hydrocarbon films (PAHs), liquid organic films, frozen
organic films, and soot. These substrates and films are used as proxies for organic particles in the atmosphere.

Determinations of the reactive uptake coefficients for NO3 and N2O5 on these different substrates will be presented.
Since the studied substrates and films are used as proxies for organic particles in the atmosphere, we demonstrate
how the uptake data for these reactions may be significant under atmospheric conditions.

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Keto-Enol Tautomerizations Catalyzed by Water and Carboxylic Acids

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Body: The ability of weakly-bound complexes to influence the kinetics of gas phase reactions, particularly in atmospheric chemistry, has long been speculated. This study uses quantum chemistry and statistical reaction rate theory to identify that bound water molecules can significantly reduce barriers to intramolecular hydrogen shift reactions, via a double-hydrogen-shift mechanism. The bound water molecule directly participates in the hydrogen shift reaction, exchanging a H atom with its counterpart. For the vinyl alcohol to acetaldehyde keto-enol tautomerization this mechanism cuts the reaction barrier approximately in half, reducing it by over 30 kcal mol-1. In contrast, while a non-participatory 'bystander' water molecule also reduces the hydrogen shift barrier, it is only by around 3 kcal/mol. When a carboxylic acid replaces water in the double-hydrogen-shift mechanism the barrier to keto-enol tautomerization is decimated, reduced to less than 6 kcal/mol (around 15 kcal/mol in the reverse direction). This results from reduced strain in the hydrogen shift transition state, and achieves enol lifetimes in the troposphere that become short on relevant timescales. Rapid enol to ketone isomerizations are currently required to explain the oxidation products of isoprene. The wider significance of rapid hydrogen shift reactions in atmospherically relevant molecules and radicals is also explored.

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Photoenhanced ozonation of polycyclic aromatic hydrocarbons on model urban film surfaces

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Body: In polluted urban environments, impervious surfaces rapidly become coated with a complex mixture of semivolatile chemicals, including a wide variety of PAH species. This coating, commonly referred to as "urban film", has been shown to be a significant substrate for heterogeneous atmospheric chemistry. Despite the film's high surface-tovolume ratio and its direct exposure to sunlight, however, comparatively little attention has been paid to photochemical processes occurring within it. Given that the products of PAH ozonation typically display enhanced toxicity, the photooxidation of PAH in urban films may have important implications for human health. Indeed, our group's recent observation that the heterogeneous ozonation of solid pyrene films is enhanced upon illumination underscores the need for further study in this area. In the present experiments, the photoenhancement of the heterogeneous reaction of ozone with a variety of solid PAH films was examined by monitoring PAH loss as a function of illumination using laser-induced fluorescence detection. In order to explore the substrate dependence of the photoozonation reaction, some experiments were also performed using PAH contained in octanol films, where octanol was used as a proxy for the organic component of urban surface films. Interestingly, the loss of solid-film PAH proceeded more quickly than that of PAH contained in octanol films, even in the dark. Furthermore, while the ozonation of some solid PAH films displayed a light enhancement, the magnitude of which depended upon the PAH identity, no such enhancement was observed for PAH incorporated in octanol films. Taken together, these results suggest that the observed photoenhanced heterogeneous ozonation of PAH arises via the formation of charge-transfer complexes between excited-state PAH species and ozone and/or oxygen at the surface of the solid film.

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Reactions of Complex Phenols on Aerosols with Gaseous Ozone

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Body: We report that α-tocopherol (α-TOH/α-TO¯), as a model of substituted phenols in atmosphere, reacts with closed shell $O_3(g)$ on the surface of inert solvent microdroplets within 1 ms to produce persistent (n = 1 – 4) adducts detectable by online electrospray ionization mass spectrometry. The prototype phenolate PhO- undergoes electron transfer under identical conditions. These reactions occur at the gas/liquid interface since their rates: (1) depend on pH, (2) are several orders of magnitude faster than those in the bulk of O_3 3-saturated microdroplets, and (3) approach $O_3(g)$ mass accommodation rates. Furthermore, they fail to incorporate solvent into the products: the same species are formed on acetonitrile or nucleophilic methanol microdroplets. Signals initially evolve with the concentration of ozone as expected from first-generation species. However, α-TO¯ reacts further with ozone via a collision-induced dissociation into a $C_{19}H_{40}$ fragment (vs. $C_{19}H_{38}$ from α-TO¯, carrying the phytyl side-chain, whereas the higher homologues (α-TO- $O_{10} \ge 2$) are not reactive with $O_3(g)$. On this basis, α-TO¯ is assigned to a chroman-6-ol (4a, 8a)-ene oxide (an epoxide), α-TO- O_{2} to an endoperoxide, and α-TO- O_{3} to a secondary ozonide. These products are previous unreported. The atmospheric degradation of the substituted phenols detected in forest fires and combustion emissions is therefore expected to produce related oxidants on aerosol particles.

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Temperature and Electrolyte Effects on the Thermochromism of Model Organic Aerosol Matter

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Body: The absorptivity of tropospheric aerosol particles in the visible, which is determined by the properties of their complex water-soluble organic carbon (WSOC) components under ambient conditions, remains a major unknown factor in the Earth's radiation balance. Herein we report experiments on model WSOC showing that its visible absorptivity is not an intrinsic property but a function of time that responds to changes in insolation, ambient temperature and relative humidity. The aliphatic polyfunctional (alcohol, carboxyl, carbonyl and ether) oligomer mixtures produced by photolysis of aqueous pyruvic acid (a proxy for α-dicarbonyls absorbing at $\lambda > 300$ nm) exhibit thermochromism, developing visible absorptions that are significantly enhanced, red-shifted and accelerated in the presence of 'inert' electrolytes, such as NH₄HSO₄, Na₂SO₄ or NH₄ClO₄. Spectral absorbances, A(λ), evolve following Kohlrausch kinetics at rates that depend inversely on λ and markedly increase with temperature, can be bleached upon irradiation and repeatedly recycled in the dark in a matter of hours. Thermochromism is ascribed to the generation of C=C unsaturations (detectable by ¹H- and ¹³C-NMR spectrometries) via β-dehydration of alcohol functionalities with an activation energy E_a ~ 30 kcal mol⁻¹. This process is catalyzed by higher acidity and ionic strength, and can be reversed photochemically.

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Formation of Secondary Organic Aerosol through Cloud Processing of Anthropogenic VOCs

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Body: Cloud and fog processing of volatile organic carbon species (VOCs) can lead to the formation of secondary organic aerosol (SOA) mass. Biogenic species and their reaction products have received some attention recently but the present study addresses the potential of SOA formation in clouds from predominately anthropogenic VOCs, specifically benzene, toluene, ethylbenzenes, and xylenes (BTEX). Aqueous phase BTEX concentrations have been established in cloud water samples collected in Northern Arizona. While BTEX species contribute little (less than 1 percent) to the total organic carbon (TOC) in clouds, the aqueous concentrations are orders of magnitude higher than predicted by Henry's law from ambient BTEX concentrations.

Aqueous phase reactivity of BTEX has been investigated using a solar simulator set-up. BTEX reactivity studies with hydroxyl radical show that BTEX species degrade readily in deionized (18Mohm) water and simulated cloud water. The products of the reactions have been identified and quantified and include ring-retaining as well as ring-opening products. Most of the identified products have lower volatility than the parent VOC and will remain in the particle phase upon droplet evaporation. Therefore the laboratory studies show that cloud processing of anthropogenic VOCs can also lead to SOA mass formation by aqueous phase pathways.

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Composition of Secondary Organic Aerosols Produced by Photo-Oxidation of Biomass Burning Emissions in a Smog Chamber

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Body: Knowledge of the chemical composition of atmospheric organic aerosols (OA) is essential for accurate representation of OA in air quality and climate models. Both the sources of OA and their properties and effects remain poorly understood. In particular, we still know relatively little about the atmospheric formation of secondary organic aerosols (SOA). There is growing interest in the impact of biomass burning emissions on air quality, human health, and radiative forcing. Through a series of experiments, we are working to quantify changes in the chemical composition of wood smoke particles as a result of photochemical aging under well-controlled laboratory conditions. One specific objective of this study is to identify markers for biomass burning SOA and test whether these markers can be used in atmospheric samples to quantify SOA formation from aging of biomass burning emissions. We analyzed SOA generated in a smog chamber by photooxidation of smoke produced by burning oak wood. In order to initiate photochemistry, the chamber was irradiated with UV light. Aqueous extracts of collected aerosol samples were analyzed with Electrospray Ionization Time-of-Flight Mass Spectrometry. The high mass accuracy of these measurements reduces ambiguity in the assignment of elemental compositions for observed ions. Analysis has shown that primary oak smoke aerosol includes products of the thermal decomposition of cellulose (levoglucosan, cyclotene etc.) and lignin (guaiacol and syringol derivatives, mostly aldehydes and alcohols). After 2 hours of aging at typical summertime hydroxyl radical concentrations, the aerosol mass increased 2.5 fold due to the production of secondary organic aerosol. Mass spectra of the secondary organic aerosol formed are dominated by organic nitrates (nitrophenol, nitrocresol, nitrocatechol, and nitroguaiacol) and aromatic acids (benzoic acid, mono and dihydroxybenzoic acid). Both nitrates and acids most likely are formed due to oxidation of the lignin decomposition products (guaiacol and syringol derivatives) by reaction with OH and NO2. This research highlights the dynamic nature of fire emissions and atmospheric organic aerosols in general.

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Can Secondary Organic Aerosol Formed in Atmospheric Simulation Chamber Be Continuously Aging?

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Body: Recent smog chamber studies have found that the oxidative processing (i.e. aging) of organic aerosol affects the chemical and physical properties for both aromatic and terpene aerosol precursors. Evidence from laboratory experiments suggests that organic aerosol can be converted from a hydrophobic to a hydrophilic state with aging. Several possible chemical mechanisms have been proposed based on chamber studies from other research groups e.g. heterogeneous reaction at the particle surface. Previous experiments conducted in the UC Riverside/CE-CERT environment chamber have shown little evidence of particle aging in terms of changes in hygroscopic properties from α-pinene dark ozonolysis systems. In this study, we simulate chemical aging of carbonaceous aerosol generated from α-pinene ozonolysis, α-pinene photooxidation and m-xylene photooxidation with an emphasis on the further uptake of oxidants, the evolution of aerosol hygroscopicity, particle density and elemental chemical composition (C:O:H) estimated from aerosol mass spectra to further investigate chamber secondary organic aerosol (SOA) aging behavior. Experimental results indicate that the SOA formed from photooxidation systems do get more functionalized as the oxidative age process go while dark ozonolysis SOA do not show aging phenomena within the normal chamber experiment duration.

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Secondary organic aerosol formation from m-xylene photooxidation: The role of the phenolic product S. Nakao; ^{1, 2}; L. Qi; ^{1, 2}; C. Clark; ^{1, 2}; K. Sato; ²; P. Tang; ^{1, 2}; D. Cocker; ^{1, 2};

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Body: Aromatic hydrocarbons comprise a significant fraction of volatile organic compounds in the urban atmosphere and their importance as precursors to secondary organic aerosols (SOA) has been widely recognized. However, SOA formation from aromatics is one of the least understood processes among all the classes of volatile organic compounds (VOCs) due to its complex multi-generation reactions. Phenolic compounds have been identified as one of the significant products from OH-initiated reaction of aromatic hydrocarbons and are suggested to have a very high potential of SOA formation (e.g., cresol isomers having SOA yield 9~42%, Henry et al., Atmos. Environ., 2008). We examined the effect of extent of oxidation of m-xylene on chemical composition and physical properties using m-xylene and xylenol as reactants in environmental chamber experiments. Chemical composition of SOA was investigated by Liquid Chromatography / Time of Flight Mass Spectrometer (LC/ToF-MS), and Aerodyne High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS). Physical properties of SOA such as density, volatility, and hygroscopicity were investigated by Aerosol Particle Mass Analyzer - Scanning Mobility Particle Sizer (APM-SMPS), Hygroscopicity/Volatility - Tandem Differential Mobility Analyzer (H/V-TDMA), respectively. Also SOA yields were obtained to evaluate the importance of xylenol as an intermediate product.

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Slow aging in Secondary Organic Aerosol observed by Liquid Chromatography coupled with High-Resolution Mass

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Body: This study investigated long term changes in the chemical composition of model biogenic secondary organic aerosol (SOA) prepared via ozonolysis of the terpene limonene. This SOA has been observed to turn brown when exposed to NH4+. Our hypothesis is that the chromophoric compounds responsible for this color change are suspected to be imidazole-like or pyridinium-like compounds. These compounds are only present in small relative amounts, hence standard mass spectrometry is insufficient to unambiguously detect these compounds. However, a combination of HPLC and high resolution electrospray ionization mass spectrometry allows assignments of chemical formulae to individual peaks. These and other experiments confirm the presence of N-containing compounds in treated SOA. We are in the process of determining the exact identity of these species by MS/MS methods. LC-MS can also provide information about the polarity of the compounds in SOA. Most compounds in limonene-O3 SOA are polar and are detected at short retention times; peaks suggesting trimeric species appear at longer retention times in the case of fresh SOA, but at shorter times with the bulk of the components for aged SOA. Limonene SOA has been shown to be composed of monomers, dimers, trimers and larger oligomers. The appearance of trimers in specific regions of the chromatogram suggests these species are genuine SOA components and not an artifact of electrospray ionization. Changes in biogenic SOA over time are important because of the propensity of SOA to affect direct and indirect radiative forcing.

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Atmospheric Heterogeneous Stereochemistry

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Body: This paper addresses the timescale and mechanism of heterogeneous interactions of laboratory models of organic-coated mineral dust and ozone. We are particularly interested in investigating the role of stereochemistry in heterogeneous oxidation reactions involving chiral biogenic VOCs. Using the surface-specific nonlinear optical spectroscopy, sum frequency generation, we tracked terpene diastereomers during exposure to 10^11 to 10^13 molecules of ozone per cm^3 in 1 atm helium to model ozone-limited and ozone-rich tropospheric conditions. Our kinetic data indicate that the diastereomers which orient their reactive C=C double bonds towards the gas phase exhibit heterogeneous ozonolysis rate constants that are two times faster than diastereomers that orient their C=C double bonds away from the gas phase. Insofar as our laboratory model studies are representative of real world environments, our studies suggest that the propensity of aerosol particles coated with chiral semivolatile organic compounds to react with ozone may depend on stereochemistry. Implications of these results for chiral markers that would allow for source appointment of anthropogenic versus biogenic carbon emissions will be discussed.

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Uptake of glyoxal on nitric acid/ice thin films under conditions relevant to the upper troposphere B. M. Connelly; ¹; D. O. De Haan; ²; M. A. Tolbert; ¹;

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Body: Several laboratory and field studies have suggested that the simple aldehyde glyoxal could be a significant source of secondary organic aerosol (SOA) found in the lower troposphere. Recent studies have found that particles in the free troposphere also contain significant amounts of organic material, some greater than 80% organic by mass. We have examined whether glyoxal could be a source of SOA in the upper troposphere. One type of particle that could potentially uptake glyoxal is cirrus ice, and several studies indicate cirrus ice may be coated with nitrate. Thus we have utilized a high vacuum Knudsen Cell to measure the uptake of glyoxal on ice exposed to nitric acid at temperatures and pressures relevant to the upper troposphere. Here we present kinetic and spectroscopic data that indicates uptake of glyoxal is efficient on these films and isothermally irreversible. Additionally, when the products of the uptake are dried and then exposed to water vapor, uptake of glyoxal continues on the products even at water pressures below the saturation pressure of ice.

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Detection of aerosol organic nitrate and ammonium nitrate by Thermal Dissociation – Laser Induced Fluorescence A. W. Rollins; ¹; J. F. Hunter; ¹; R. C. Cohen; ^{1, 2};

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Body: We demonstrate the ability to specifically detect both organic nitrates (R-ONO2) and ammonium nitrate in submicron particles. The technique is based on the well established method of Thermal Dissociation followed by Laser Induced Fluorescence of NO2 (TD-LIF) which has been previously used for detection of total (gas + particle) organic nitrates and nitric acid. In our application an activated carbon denuder is used at the inlet to remove the gas phase nitrates prior to detection yielding a particle only signal. The limit of detection is determined by the mixing ratio of NO2 produced by the thermal dissociation of RONO2 or HNO3, and for the instrument implemented here is 80 ppt NO2 for 10 seconds of signal averaging. This is equivalent to 0.20 μg/m3 of NO3- from ammonium nitrate at standard conditions. Results of chamber experiments are presented which demonstrate the unambiguous quantification of organic nitrates in the aerosol formed from oxidation of biogenic VOC.

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Branching Ratios for the Reaction of Carbonyl-Containing Organic Peroxy Radicals with Hydroperoxy Radicals A. Hasson; ¹; G. S. Tyndall; ²; S. Hernandez; ¹; S. Campbell; ¹; S. Singh; ¹; Y. Ibarra; ¹; J. J. Orlando; ²;

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- 2. Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, CO, United States.

Body: An important chemical sink for organic peroxy radicals (RO2) in the troposphere is reaction with hydroperoxy radicals (HO2). While this reaction is typically assumed to form hydroperoxides as the major products (R1a), a small number of carbonyl-containing peroxy radicals have been shown to undergo other reactions (R1b and R1c) with substantial branching ratios

RO2 + HO2 → ROOH + O2 R1a

RO2 + HO2 → ROH + O3 R1b

RO2 + HO2 → RO + OH + O2 R1c.

In this work, branching ratios for R1a-c were measured for six organic peroxy radicals: C2H5C(O)O2, C3H7C(O)O2, CH3C(O)CH2O2, CH3C(O)CH(O2)CH3, CH2CICH(O2)C(O)CH3 and CH2CIC(CH3)(O2)CHO. The temperature (248 – 298 K) and pressure (100 – 800 Torr) dependence of the branching ratios for acetyl peroxy and acetonyl peroxy radicals was also investigated. Product yields were determined using a combination of long path Fourier Transform infrared spectroscopy, High Performance Liquid Chromatography with fluorescence detection, Gas Chromatography with Flame Ionization Detection and Gas Chromatography-Mass Spectrometry. All of the carbonyl-containing peroxy radicals investigated have significant branching ratios for Reaction R1c. The branching ratios for both acetyl peroxy and acetonyl peroxy radicals are temperature dependent but pressure independent over the ranges investigated. The mechanistic and atmospheric implications of these results will be discussed.

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Particulate organic nitrate and organic sulfate contributions to SOA: Response of the AMS and constraints from field

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- 4. MIT, Cambridge, MA, United States.
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Body: Organic nitrates (ON, RONO2) are produced in the atmosphere through gas-phase reactions of anthropogenic and biogenic VOCs in the presence of NOx, and by VOC oxidation by NO3 radicals. These species serve as indicators of ozone production and can alter the NOx/VOC reactivity dependence of O3 production. Large, multifunctional organic nitrates should have lower vapor pressures, allowing them to condense in the particle-phase and form secondary organic aerosol (SOA). Despite their potential importance, few studies have been able to quantify particulate organic nitrates in ambient data. Organic sulfates (OS, ROSO3H) have been recently been reported as components of biogenic SOA from laboratory and field studies. The OS moiety will increase water uptake from SOA and can act as an acid by donating a proton from the sulfate group. Aerodyne Aerosol Mass Spectrometers (AMSs) are very commonly used to quantify and characterize organic aerosol (OA) during field studies. The AMS uses electron impact ionization, and the response of these instruments to ON and OS is unclear. Here we report the response of a High Resolution Time-of-Flight AMS to several ON and OS standards and mixtures. We also report data acquired using soft-ionization with the Advanced Light Source AMS. Most of the ON nitrogen appears in the NO+ and NO2+ ions in the AMS; these ions are generally assumed to be dominated by inorganic nitrate during field studies. Similarly, most of the OS sulfur appears in the HxSOy+ ions which are also characteristic of inorganic sulfate. Minor ON (OS) ions are observed that contain C and N (C and S), although their identity and signal fraction varies between standards. A new technique is demonstrated for the challenging problem of quantifying the weak CHN ions that are generally sandwiched between large CH and CHO ions in the high-resolution AMS spectra. However several of these ions are also produced from other species, so care should be used before using their full intensity to estimate ON and OS concentrations. The NO+/NO2+ ratio is higher for ON than NH4NO3, providing one possible route for evaluation of the presence of ON. In contrast, the ion ratios for OS are similar to those for inorganic sulfate. Interestingly, OS are found to be partially associated with NH4+ counterions, suggesting that these species are at least partially in ionic form in ambient aerosols. The characterizations emerging from the laboratory standards are used to constrain the importance of ON and OS in recent HR-ToF-AMS field studies in Mexico City, Riverside, CA, and Blodgett Forest, CA. We evaluate the potential of using NOx+ fragment ratios, CN and CS-containing ions, and the ammonium balance of the nominally inorganic species for constraining the concentrations of ON and OS for these ambient datasets. A clear conclusion is that the NOx+ and HxSOy+ signals in the AMS are dominated by the inorganic species in these studies, but that ON and OS can still play a role for OA oxidation state.

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Changes in Concentrations and Stable Carbon Isotopic Compositions of Diacids, Ketoacids and α-Dicarbonyls in Atmospheric Aerosol Filter Samples with Photochemical Aging: A Laboratory Study

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Body: Low molecular weight dicarboxylic acids, ketoacids and dicarbonyls are ubiquitous in atmospheric aerosols and are considered to act as cloud condensation nuclei (CCN) and play an important role in cloud process and climate change. They be directly emitted into the atmosphere from primary sources, but they are mainly produced by secondary photochemical processes in the atmosphere. Although observation studies for these compounds have been conducted in different regions, their formation and degradation processes are still not fully understood. Compound-specific stable carbon isotope analysis (CSCIA) of organic compounds has successfully been used to understand the photochemical aging of atmospheric aerosols. In this study, we conducted a laboratory experiment to investigate the photochemical processes of water-soluble organics using atmospheric aerosol samples. About 10 cm^2 of two atmospheric aerosol filter samples collected using high-volume air sampler at Chennai, India in winter and summer (one in each season) were exposed to UV light (254 nm) for 0.5, 1.5, 3.0, 6.0, and 12 hours in the quartz tube reactor under humid conditions. Samples were analyzed for water-soluble organic compounds using a GC and GC/isotope ratios mass spectrometer. Concentration of total diacids (1030 ng m⁻³) in winter sample before UV exposure gradually decreased to 408 ng m⁻³ with photochemical processing (6 hrs) and then slightly increased to 594 ng m⁻³ in 12 hrs. In contrast, their concentrations of summer aerosol filter sample (initially 640 ng m⁻³) decreased to 480 ng m⁻³ in 0.5 hrs, but gradually increased with UV irradiation up to 836 ng m⁻³ in 12 hrs. On the other hand, concentrations of ketoacids and dicarbonyls generally increased with photochemical aging (12 hrs) in both winter and summer samples, although there are few exceptions. Measurements of stable carbon isotopic composition (δ¹³C) showed the 13 C enrichment of oxalic (C₂), malonic (C₃) and glyoxylic (ω C₂) acids in both winter and summer samples during an early stage of UV exposure. However, other species did not show a clear trend of δ^{13} C, although their concentrations increased with photochemical processing. Here, we present the results and discuss the plausible pathways for the production and degradation of low molecular weight dicarboxylic acids and ketoacids during photochemical aging based on the changes in their concentrations and molecular composition as well as stable carbon isotopic ratios.

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The Morphology of Internally Mixed SOA/DOP Particles and the Uptake of Gas-Phase DOP During SOA Formation *T. D. Vaden;* ¹; *C. Song;* ¹; *R. A. Zaveri;* ¹; *D. G. Imre;* ²; *A. Zelenyuk;* ¹;

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Body: Traditional semi-empirical secondary organic aerosol (SOA) models include primary organic aerosols (POA) as additional organic mass to absorb greater amounts of oxidized organic gases significantly enhancing the modeled SOA yields. An important assumption in the models is that the SOA/POA particles form a well-mixed organic aerosol phase. The very same models also assume that the organics substances in POA are nonvolatile. We present a detailed experimental investigation of the morphologies of particles formed during ozonolysis of α-pinene in the presence of dioctyl phthalate (DOP) particles using our single particle mass spectrometer, SPLAT II. Two different types of mixed SOA/DOP particles were generated and characterized: those formed by condensation of the oxidized α-pinene products on size-selected DOP particles (Figure 1c) and by condensation of DOP on size-selected α-pinene SOA particles (Figures 1a and 1b). We find that the hydrophilic SOA material and hydrophobic DOP do not mix, but form instead distinct and separated phases. An examination of SOA particles formed by homogeneous nucleation in the presence of DOP (vapor pressure of 1.3 10-7 Torr) particles shows them to be encapsulated by a thin layer of DOP (Figure 1d). These findings could have significant impact on SOA formation in the atmosphere, where various volatile and non-volatile organic compounds may be present.

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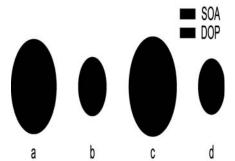


Figure 1. A schematic illustrating the types of particles characterized in this study: a) 160 nm SOA particles coated with 65 nm layer of DOP; b) 140 nm SOA particles coated with 20 nm layer of DOP; c) 195 nm DOP particles coated with a 50 nm layer of SOA that is coated by ~2 nm of DOP; d) 151 nm SOA particles formed by homogeneous nucleation in the presence of DOP vapor. These particles acquire a ~4 nm thick layer of DOP coating.

Chemical aging of organic aerosols: Evidence of changing hygroscopicity and volatility

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Body: Organic aerosols constitute a large fraction of the tropospheric aerosol budget. The composition and size of aerosol is important with respect to its effect on radiative forcing and impacts upon human health. During its lifecycle, an aerosol will encounter changing physical parameters (temperature, pressure, relative humidity) and different chemical fields (O3, OH, NO3, halogens, etc.). The composition of the aerosol can evolve via condensational aging, heterogeneous chemistry aging and homogenous chemistry aging. Condensational aging occurs via the evaporation and condensation of semi-volatile species from the aerosol to gas phase. The hygroscopicity of the aerosol dictates the water content and to a large extent the size of the aerosol.

Our group have recently investigated the role an evolving chemical environment has upon the size and mass of aerosol. These laboratory studies have used a mixture of electrodynamic balance, mass spectrometric, and spectroscopic methodologies to probe aerosol size and composition. In particular, the ozonolysis of organic aerosol, containing alkenyl moieties, will be discussed. It is found that short exposure times of aerosols to small ozone concentrations result in dramatic changes in the volatility and hygroscopicity of various types of organic aerosol. In some cases, previously non-volatile aerosols become significantly volatilized upon reaction with ozone. The phase of the ozone-attacked aerosol is important to the reaction mechanism. Furthermore the chemical aging of the organic fraction within mixed organic/inorganic aerosols is investigated. Mechanistic pathways and their atmospheric implications will be presented.

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Substitution kinetics and energetics of aliphatic amines for ammonia in aerosols

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Body: While aliphatic amines are present in the atmosphere at concentrations estimated to be two orders of magnitude lower than that of ammonia, they have been detected at elevated concentrations during particle nucleation events. Proton affinity data suggest that proton transfer should be more facile for an amine-acid cluster than for an ammonia-acid cluster. A recent theoretical study indicated that amines may enhance sulfuric acid-water nucleation; however, little experimental work has been performed to study the formation of amine sulfate clusters. This work examines the exchange of monomethylamine, dimethylamine, and trimethylamine into ammonium sulfate and ammonium nitrate clusters using Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS). Ammonium salt clusters were introduced to the FT-ICR-MS by electrospray and were reacted with gaseous amine at constant pressure. Reaction time was varied until the reaction reached completion. A reaction profile was created and data were statistically fit to obtain rate constants for the exchange, from which reaction probabilities (uptake coefficients), equilibrium constants, and Gibbs free energy values were determined. In general, reaction occurred very quickly, with amine fully displacing ammonia after a few seconds. Amine salt clusters were also introduced to the spectrometer by electrospray and were reacted with gaseous ammonia to obtain rate constants for the reverse reaction. Mixed clusters containing both ammonia and amine were isolated and reacted with amine and ammonia gases in order to more directly determine rate constants for exchange. Additionally, amine salt clusters were introduced to the FT-ICR-MS and were reacted with other gaseous amines in order to examine the exchange kinetics of one amine versus another. The results suggest that organic amine concentrations will be greatly enhanced relative to ammonia concentrations in atmospheric particles. Implications for ambient particle nucleation and growth will be discussed.

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Characterization of Potential Aerosol Mass using an Oxidation Chamber coupled to an Aerodyne HR-ToF-AMS during DAURE, SHARP, and FLAME-3

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Body: The Potential Aerosol Mass (PAM) oxidation chamber (Kang et al., ACP 2007) used in front of an aerosol instrument provides an indication of the secondary inorganic and organic aerosol formation potential in an airmass. The chamber, a flow tube with small residence time, rapidly oxidizes ambient air through exposure to high concentrations of ozone (O3) and hydroxy (OH) and hydroperoxy (HO2) radicals. Here we use a recently-modified PAM chamber in conjunction with an Aerodyne High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS; DeCarlo et al., Anal. Chem. 2006) during two ambient studies and one source study: the Determination of the Sources of the Atmospheric Aerosol in Urban and Rural Environments in Spain (DAURE) in February 2009, the Study of Houston Atmospheric Radical Precursors (SHARP) in April 2009, and the Fire Lab at Missoula Experiment phase 3 (FLAME-3) in Sep. 2009. The AMS samples alternatively between ambient air and chamber-processed air every 2.5 min., and a cycling of the UV light intensity (OH exposure) is also used with a maximum equivalent exposure of about 7 days. Large variations in the organic PAM were observed in the different studies. In particular very large PAM values were observed in several evenings in Houston when the site was impacted by emissions from petrochemical facilities. Optimum OH exposure for maximum PAM was observed at intermediate OH levels. For all experiments we compare the amount of SOA formed in the PAM chamber to the yield predicted by the measured precursors (from PTRMS instruments).

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Changes in chemical composition observed during organic aerosol aging

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Body: Organic aerosol in the atmosphere consists of a multitude of organic species which are the products of a variety of chemical reactions. This complexity challenges our ability to explicitly characterize the chemical composition of these particles. In recent years methods have been developed to characterize the bulk organic aerosol composition (eg. AMS high resolution elemental analysis). We will explore how this bulk composition changes with aerosol aging. We show these changes in the space of a Van Krevelen diagram (H:C vs. O:C) to highlight the common compositional changes observed for both field and lab organic aerosol under a variety of conditions and environments. We find that carboxylic acid addition is a proxy for the observed compositional changes associated with organic aerosol aging. This finding has important implications for our understanding of aerosol processing and representation of these processes in models.

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Determining aromatic reaction rates from the photo-oxidation of diesel exhaust using in-situ comprehensive multidimensional chromatography (GC × GC)

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Body: Aromatic hydrocarbons are an important class of volatile organic carbon compounds present in the atmosphere. The major loss process in the atmosphere is through reaction with the hydroxyl radical (OH) that leads to the formation of both tropospheric ozone and particulate matter, with implications for air quality and climate. Since comparisons between laboratory chamber studies of single compounds or simplified mixtures and ambient observations are not always straightforward, there is a need to investigate the physical and chemical transformation processes taking place within real mixtures under atmospherically relevant conditions. Diesel exhaust is a complex mixture that contains a large size and volatility range of aromatic and polycyclic aromatic hydrocarbons (PAH). The complexity of this type of mixture provides a significant measurement challenge. Recently, we have successfully modified our thermal desorption aerosol gas chromatography instrument (TAG) to incorporate comprehensive multidimensional chromatography (GC × GC), which provides dramatically improved compound separation. The GC × GC technique also offers structured chromatograms where related compounds tend to appear in distinct bands in a two dimensional plane indicative of the volatility versus polarity of the observed molecules. The reduction in co-eluting peaks produces 'cleaner' mass spectra, which along with the polarity versus volatility separation greatly facilitates peak identification. In this work, we present data from the photo-oxidation of diesel exhaust conducted at the Carnegie Mellon University smog chamber and determine oxidation rates for the n-alkyl benzenes, alkylated naphthalenes, and phenanthrene. Substantial secondary organic aerosol (SOA) was produced in the experiments and the reaction rate of aromatics was compared to the SOA production rate.

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Effects of dilution on vehicle emissions of primary particles

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Body: Dilution of primary aerosols from vehicles into the ambient atmosphere can change their physical and chemical characteristics. In order to study these processes, experiments were conducted in an engine testing facility at Environment Canada in Ottawa, Ontario. Exhaust from a light duty diesel engine was vented into a constant volume sampling (CVS) system where it underwent primary dilution at an ambient temperature of 25oC, leading to a primary dilution ratio of 10-15. From the CVS, the exhaust was further diluted using a combination of a Dekati ejection diluter and mixing with zero air in a flow tube, achieving secondary dilution ratios of up to 3000. Particle and gas measurements were made through multi-ports in the CVS and the flow tube using an SMPS, FMPS, AMS, and SP2, and instruments to measure CO, CO2, NOx, and total hydrocarbons (THC). Preliminary results indicate that regardless of dilution ratios, primary particles contain significant amounts of organic material that appear to reside on small black carbon cores. With increasing dilution ratios, the primary particle sizes become progressively smaller, suggesting volatilization of the adsorbed organic material. Results from various engine operating modes (simulating different driving conditions) will be presented.

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Field observations linking organic carbon content to optical properties in atmospheric aerosols

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Body: Ground and airborne measurements of aerosol optical properties and chemical composition are reported from the fall 2008 Cheju Atmospheric Brown Cloud Plume-Asian Monsoon Experiment (CAPMEX; www-ramanathan.ucsd.edu/capmex.html) and spring 2007 Indirect and Semi-direct Aerosol Campaign (ISDAC; acrf-campaign.arm.gov/isdac/) field campaigns. Correlation between increased short wavelength absorption, measured by a 3-laser photoacoustic soot spectrometer (LANL), and increased brown carbon content, measured by single particle laser ablation aerosol mass spectrometry (PNNL) and filter based thermo-optical methods (UW), are observed in both Asian continental outflow and Arctic Haze aerosols. In both campaigns, we observe significant darkening in single scatter albedo at 405 nm (down to ~0.7) relative to 532 and 781 nm for aerosols with larger brown carbon (soot + organic) mass fractions. We investigate the nature of optical property/composition correlations and their implications for radiative forcing; determination of the wavelength dependence of mass absorption cross sections for brown carbon aerosols; and the utility of diagnosing aerosol sources using the wavelength dependence of their optical properties.

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Summertime Anthropogenic and Biogenic Influences on Particle Composition at a Remote Forested Site in Western Canada

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Body: As part of the Whistler 2008 field campaign, Aerodyne time-of-flight aerosol mass spectrometers (AMS) were deployed at Whistler, BC, Canada, from May 15 to June 18, 2008. Whistler is located in a remote, forested region approximately 100 km north of Vancouver. The two AMS instruments were located at separate sites on Whistler Mountain: (1) Olympic Station (1019 m ASL, below treeline), and (2) Whistler Peak (2182 m ASL, above treeline). The sampling period was chosen to maximize measurements of periods dominated by biogenic emissions, while minimizing the influence of seasonal biomass burning events. The particulate organic fraction at both sites is analyzed using Positive Matrix Factorization, yielding factors related to hydrocarbon-like organic aerosol from primary emissions and oxygenated organic aerosol (OOA). Variation in the extent of oxidation of the OOA is represented by more oxygenated (OOA-1) and less oxygenated (OOA-2) factors. Comparison of the organic mass spectra and PMF factors at the two sites with CO, volatile organic compounds measured by proton transfer reaction mass spectrometers (PTR-MS), and particulate inorganics enable evaluation of the relative contributions of biogenic and anthropogenic emissions to the organic aerosol. Observed influences at the site include biogenic emissions from the surrounding coniferous forest, transported organics from Vancouver, long-range transport from Asia, and emissions from local construction activity.

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Can 3D Models Explain the Primary and Secondary, Fossil and non-Fossil Organic Aerosols during the 2006 MILAGRO Experiment?

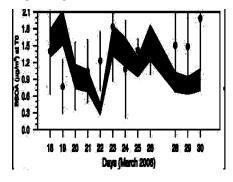
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Body: Organic aerosols (OA) remain one of the less well characterized components of the atmosphere despite their effects on climate and human health. In the framework of MILAGRO, the predictions of a meso-scale chemistrytransport model have been compared to AMS/PMF and 14C data to identify major sources and formation processes leading to a large amount of primary (POA) and secondary (SOA) organic aerosols in Mexico City during March 2006. SOA formation was modeled by both (i) the traditional one-step oxidation of anthropogenic (eg aromatics), biogenic (monoterpenes and isoprene), and biomass-burning SOA precursors followed by their partitioning into organic and aqueous phases; and (ii) the volatility basis set-based oxidation and partitioning of semi-volatile and intermediate volatility primary organic vapors (S/IVOCs; Robinson Sci07; Grieshop ACP09). Based on a month long comparison performed at the surface within the city and aloft in the outflow region it was found that anthropogenic and biomass burning emissions of primary OA (and SOA precursors) are reasonably captured within the city, although model overestimation of BBOA can be seen downwind of large fires. In contrary, large discrepancies are encountered for SOA, with a factor of 2-10 model underestimate when only traditional anthropogenic SOA precursors and yields are considered. A large increase in the partitioning coefficients (simulating e.g. oligomerization) reduces the modelmeasurement discrepancy but does not close the whole gap. Accounting for S/IVOC leads to a substantial increase of the predicted SOA. Although S/IVOC runs help explain the missing SOA mass, inconsistencies in the spatial SOA distributions and O/C ratios are found for both the original (Robinson 07) and the updated (Grieshop 09) approach. Model results also indicate that SOA found in the Mexico City basin is substantially influenced by regional SOA (~1.5 µg/m3) of biogenic origin which is formed over the coastal mountain ranges and advected around Central Mexico. The presence of biogenic SOA was confirmed by tracer-derived estimates of 1.14-1.35 µg/m3 of biogenic SOA within the city, in good agreement with model predictions, and in the first such comparison to our knowledge. The presence of this biogenic SOA helps explain the significant amount of modern carbon in the OA inside the city during low biomass burning periods (35%). For the first time, the model predictions were also evaluated in terms of the concentrations of modern and fossil carbon using combined 14C and AMS measurements.

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Measurement-derived (line) and model-predicted (dots) daily-averaged concentrations of biogenic SOA at T0. The variability is also indicated (+/- 1 sigma).

Modeling of secondary organic aerosols formation during MILAGRO 2006

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Body: The MILAGRO field campaign took place in March 2006 with the aim of gaining a better understanding of the evolution of trace gases and particulate matter emitted in Mexico City. During MILAGRO, a wide range of meteorological, chemical and particulate measurements were performed over local, regional and large-scale domains, all of which makes MILAGRO the most complete study of a tropical megacity's atmosphere to date. Measurements of secondary organic aerosols (SOA) performed during MILAGRO cannot be explained by traditional SOA models. For example, Kleinman et al. (ACP 2008) reported model vs. measurement discrepancy of about an order of magnitude higher measured SOA than expected from aromatic oxidation using MILAGRO aircraft data. Likewise, Hodzic et al. (ACPD 2009) and Tsimpidi et al. (ACPD 2009) reported model vs. measurement discrepancies of the same order when using traditional SOA models inside two different regional models over Mexico City. Dzepina et al. (ACP 2009) recently showed that the inclusion of several newly proposed SOA formation mechanisms can close the gap in SOA mass between measurements and models for the case study during MCMA-2003 field campaign. Here we apply the same model as in Dzepina et al. (ACP 2009) to model the evolution of organic gases and particles advected from downtown Mexico City at an altitude of ~3.5 km during three days of aging and dilution. Traditional SOA model products, mainly from the oxidation of aromatics, cannot explain the observed $OA/\Delta CO$ ratios in aged pollution. However, over the regional scale there is a significant contribution of low-NOx aromatic pathway (Ng et al., ACP 2009) that has a very high SOA yield and forms non-volatile SOA which in the model always remains in the particle phase as dilution proceeds and semi-volatiles evaporate. The non-traditional SOA model (Robinson et al., Science 2007) still produces the most SOA model mass, although its mass fraction of total SOA decreases during aging. We also characterize the volatility and O/C ratio of model SOA mass after one, two and three days of aging, and compare it to the MILAGRO measurements. Finally, we explore the behavior of the SOA model mass upon rapid lifting to the upper troposphere during convection.

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Investigation of Dairy Farm Silage Emissions

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Body: California's Central Valley is one of the most ozone polluted areas in the United States. For better understanding of the sources of this increasing tropospheric ozone concentration, an experiment was conducted on a dairy farm located in the central valley area. Dairy farm silage is a suspected source of tropospheric ozone due to recent findings of ethanol emissions resulting from the fermentation process that occurs during the preparation of silage. However, a silage pile consists of three main layers and each layer has different physical and chemical properties. During the distribution period, the inner layer is most exposed. This experiment was focused on wheat silage, and different layers of the individual silage pile were tested to investigate their emissions. Samples were collected using air canisters and analyzed via FID gas chromatography in the University of California Irvine Rowland/Blake Lab. The samples collected did reveal ethanol concentrations, and a difference was observed between the layers of the silage pile. The dry outer layer of the pile had a smaller amount of gaseous emissions than the inner "moist" section of the pile. Additionally, an unexpected peak in the inner layer's chromatogram showed a propyl alcohol concentration of 28,000 ppbv in comparison to an ethanol concentration of 15,000 ppbv. Propyl alcohol has a higher Maximum Incremental Reactivity (MIR) value, than that of ethanol. MIR is a numerical value assigned to compounds based on their ozone forming potential. Therefore, a high concentration of propyl alcohol in silage is probable to be a contributor to the tropospheric ozone concentration in the atmosphere. The information provided by this research experiment can induce further research on dairy farm emissions. Continuing this research could potentially provide scientific information required to create regulations.

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Apportionment of Size Distribution Data from the Aerodyne Aerosol Mass Spectrometer in Pittsburgh and Mexico City I. M. Ulbrich; ¹; Q. Zhang; ²; A. C. Aiken; ¹; M. Canagaratna; ³; N. L. Ng; ³; D. R. Worsnop; ³; J. L. Jimenez; ¹;

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Body: Aerodyne aerosol mass spectrometers (AMS) collect spectra of ambient aerosols that are a mix of various primary and secondary sources. Many recent studies have reported the identification of sources / components based on factor analysis of total mass spectrum mode (bulk submicron) data (e.g. Zhang et al., ES&T 2005; ACP 2005; GRL 2007; Lanz et al., ACP 2007; Ulbrich et al., ACP 2009; Aiken et al., ACP 2009; Huffman et al., ACP 2009; Slowik et al., ACPD 2009). Data collected in the PToF (particle time-of-flight) mode of the AMS represent chemical composition as a function of particle size (vacuum aerodynamic diameter). We conceptualize these data as a 3-dimensional matrix and discuss the various approaches to their factorization in order to investigate the information content about aerosol sources and processing. Factoring the size distribution data utilizes an additional dimension of information that may allow greater factor separation than using the mass spectral mode data alone. We briefly review previous applications of factor analysis to size distribution data (chemically resolved and not). We use the Multilinear Engine (Paatero, J. Comput. Graph. Stat., 1999) to determine the changing size distribution and concentration of chemical components (e.g., sulphate, nitrate, and types of organic aerosol (OA) including oxidized (OOA), hydrocarbon-like (HOA), and biomass-burning (BBOA)). We demonstrate the method with AMS datasets acquired in Pittsburgh in 2002 (Q-AMS; Zhang et al., ES&T 2005; Ulbrich et al., ACP 2009) and Mexico City in 2006 (HR-ToF-AMS; Aiken et al., ACP 2009). To our knowledge this is the first attempt to factorize highly time- and size-resolved composition data.

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Identification of Atmospheric Organic Matter in Fog Water: Exact Masses, Empirical Formulas, and Structural Insights L. R. Mazzoleni; ¹; J. L. Collett; ²;

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Body: Aqueous processing of organic matter by clouds and fogs may significantly alter aerosol-climate properties. Heterogeneous chemical reactions that serve to promote oxidation of apolar primary emission components may result in an increase of hydrophilic organic compounds, while reactions that serve to promote oligomerization and/or formation of larger components, such as HULIS, may result in a decrease. Since aerosol organic matter is very complex and its identity is not well understood, we chose to study the detailed molecular composition of atmospheric organic matter (AOM) of polluted fogs by ultra-high resolution FT-Ion Cyclotron Resonance Mass Spectrometry (FT-ICR MS). In all of our analyses, we found a high degree of complexity across the mass range of 100 to 400 u and in some of our analyses we observed our mass range to extend up to 1000 u. The detected negative organic ions were multifunctional compounds which include C. H. N. O. and S elements. We observed organic nitrogen (CHNO), organic sulfur (CHOS), and organic nitrooxy-sulfate compounds (CHNOS) as well as many masses with only CHO elemental composition. Analysis of the atomic valances by double bond equivalents (DBE) calculations suggests that these compound structures range from highly aliphatic to aromatic with DBE values of 1-11, suggesting a wide variety of precursor compounds with variable oxidation states. This resulted in a high degree of complexity in the low mass range which was greatly reduced by data filtering strategies that group assigned formulas into homologous series and oligomeric series with increasing chain lengths. The AOM oligomeric series with formula differences of C3H4O2 are very likely due to an aqueous esterification reaction, originally suggested by Altieri et al., 2008. We found over 400 oligomer series in our dataset, representing approximately 80% of the CHO and CHNO compounds combined. A very high number of homologous series of compounds and polyfunctional oligomers were found in this dataset, suggesting that these compounds are amphiphilic, meaning that they contain both hydrophilic and hydrophobic structural aspects. Thus, the role of the AOM identified organic compounds with respect to aerosol-climate properties is quite complex.

Altieri, K. E., S. P. Seitzinger, A. G. Carlton, B. J. Turpin, G. C. Klein and A. G. Marshall (2008). "Oligomers formed through in-cloud methylglyoxal reactions: Chemical composition, properties, and mechanisms investigated by ultrahigh resolution FT-ICR mass spectrometry." Atmospheric Environment 42(7): 1476-1490.

URL: http://www.chemistry.mtu.edu/~lrmazzol/

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Molecular distribution, seasonal variation, chemical transformation and sources of dicarboxylic acids and related compounds in atmospheric aerosols at remote marine Gosan site, Jeju Island

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- 3. Korea University, Department of Earth and Environmental Sciences, Seoul 136-701, Korea, Republic of.

Body: Abstract:

A homologous series of C_2 - C_{12} α , ω -dicarboxylic acids, ω -oxocarboxylic acids (C_2 - C_9), pyruvic acid and α dicarbonyls (C2-C3) were detected in atmospheric aerosols collected between April 2003 and April 2004 from remote marine Gosan site (33°29' N, 126°16' E) located in Jeju Island, South Korea. They were determined using a GC-FID and GC/MS. Total diacid concentration ranged from 130 to 1911 ng m⁻³ (av. 642 ng m⁻³), whereas total oxoacid concentration ranged from 7 to 155 ng m $^{-3}$ (av. 43 ng m $^{-3}$), and pyruvic acid and α -dicarbonyls ranged from 0.5 to 15 $^{-3}$ (av. 5 ng m⁻³) and 2-108 ng m⁻³ (av. 17.3 ng m⁻³), respectively. Oxalic ($^{\circ}$ C₂) acid was the most abundant in all seasons followed by malonic (C_3) or succinic (C_4) acid, and phthalic (Ph) acid. The concentration of diacids decreased with an increase in carbon number except for azelaic (C_9) acid, which was more abundant than suberic (C_9) acid, which was more abundant than suberic (C_9) $_8$) acid. Glyoxylic acid was predominant ω -oxoacid contributing to 92% of total ω -oxoacid. Total diacids, oxoacids and dicarbonyls showed maximum concentrations in spring and occasionally in winter, while minimum concentrations were observed in summer. Air mass trajectory analysis suggests that either spring or winter maxima can be explained by strong continental outflow associated with cold front passages, while summer minima are associated with warm southerly flows, which transport clean marine air from low latitudes to Jeju Island. The comparison between total diacid concentration level of this study and other study results of urban and remote sites of East Asia reveals that Gosan site is more heavily influenced by the continental outflow from China. The seasonal variation of $malonic/succinic \ (C_3/C_4), \ malic/succinic \ (hC_4/C_4), \ fumaric/maleic \ (F/M), \ oxalic/pyruvic \ (C_2/Py) \ and \ oxalic/Glyoxal \$ /Gly) ratios showed maxima in summer due to an enhanced photo-production and degradation of diacids and related compounds. Throughout all seasons C_3/C_4 ratio at Gosan site, located between Chinese cities and Chichi-jima Island in Japan was observed higher than those in Chinese cities and lower than that of the Chichi-jima Island, pointing to the formation of diacid during long range transport. The lowest values of adipic/azelaic (C_g/C_q) and phthalic/azelaic (Ph/C_9) were observed as a result of the overwhelming biogenic emission of the precursors (e.g., unsaturated fatty acids) of azelaic acid in summer. In this study, we will also discuss the sources and transport pathways of diacids and related compounds resolved using a hybrid receptor model, potential source contribution function (PSCF) and model results will be compared with available in-situ observations in East Asia.

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Measurements of gas-phase inorganic and organic acids in biomass fires by negative-ion proton-transfer chemical-ionization mass spectrometry (NI-PT-CIMS)

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- 3. Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, United States.
- 4. Chemistry, University of Montana, Missoula, MT, United States.

Body: We investigated emissions from 40 controlled laboratory biomass fires at the combustion facility at the U.S. Department of Agriculture (USDA) Forest Sciences Laboratory (FSL) in Missoula, MT. Gas-phase organic and inorganic acids were quantified using Negative-Ion Proton-Transfer Chemical-Ionization Mass Spectrometry (NI-PT-CIMS), Fourier Transform Infrared Spectroscopy (FTIR), and Proton-Transfer-Reaction Mass Spectrometry (PTRMS). Measurements by NI-PT-CIMS, a novel technique which measures mass to charge ratio of ions generated from reactions of CH3COO- ions with inorganic and organic acids, was validated by inter-comparing in situ measurements of HONO with FTIR and HCOOH with both FTIR and PTR-MS. Emission ratios for various, important, reactive acids with respect to CO are determined. Emission ratios for HNCO, benzenediol, acrylic, methacrylic, propionic, and glycolic acid were measured for the first time. Our measurements show a significant contribution of HONO to initial biomass emissions. The presence of HONO, a source of OH, in fresh emissions speeds up initial plume chemistry. Emission ratios for these important reactive acids, with respect to CO, are presented.

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Measurement of Organic Acids Produced By The Gas-Phase Ozonolysis of Simple Olefins Using Chemical Ionization Mass Spectrometry (CIMS) as a Function of Temperature And Humidity

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Body: Non-methane hydrocarbons (NMHCs) form an important trace component of the atmosphere and are of particular environmental interest because of their deleterious effects on air quality, their numerous (and potentially counteractive) effects on Earth's climate system and their sophisticated semiochemical roles in the world's ecosystems. NMHCs are also important precursors to the formation of secondary organic aerosol (SOA) (e.g. Pandis et al., 1991; Kavouras et al., 1999). The ozonolysis reactions of olefins result in complex menageries of products, of which the acids are ubiquitous. Although the gas phase acid concentrations are small, they are thought to be key species in SOA formation as a result of their low volatility (e.g., Ma et al., 2009). Despite this, the factors that control acid formation are not well understood, especially with regards to humidity and temperature.

Acid yields will be measured using the newly commissioned EXTreme RAnge (EXTRA) chamber (Leather et al., 2009). EXTRA is a 125 L stainless steel chamber, which can be temperature controlled using a commercial chest freezer unit (for T ≤ -20 °C) or a purpose built oven for T > 25 °C. The EXTRA chamber can be operated at pressures from 10–3800 Torr and at temperatures from 180–473 K. The stainless steel chamber walls have been coated with PFA to minimize wall loss of radicals. Fans, located at both ends of the cylinder, promote rapid mixing of reactants. Six sample ports are located at either end of the chamber for connection to ADS-GC-ECD, CIMS and commercial sensors such as a Thermo Electron Corporation 49i Ozone Analyzer, an Edinburgh Instruments Gascard CO2 sensor and a Trace Analytical inc. RGA3 CO analyzer.

Experiments will be performed as a function of atmospherically relevant temperatures (T= 180–300 K). The field CIMS has sub ppt(v) L.O.D.s with a sub 1 Hz time response so will enable products to be quantified at very low concentrations in real time. Acid products will be detected using both the acetate ion (Verez et al., 2008) and silicon pentafluoride ion (Huey et al., 1998) reaction schemes, both of which have been used previously in atmospheric measurements, with little interference from water vapour.

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Measurements of Product-Specific VOC Reactivities during the PROPHET 2008 field intensive using proton transfer reaction linear ion trap (PTR-LIT) mass spectrometry

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Body: A major aim of the PROPHET 2008 field intensive conducted at the University of Michigan Biological Station was to more completely understand the local formation of secondary organic aerosol from oxidation of biogenic volatile organic compounds (BVOCs). This oxidation was monitored at every step including gas phase reactant VOCs, oxidants, reaction products, and aerosol number and size distribution. A proton transfer reaction - linear ion trap (PTR-LIT) mass spectrometer was developed and utilized to quantify and distinguish isomeric VOCs as well as test for interferents by allowing for MSn experiments while retaining a LOD in the 100 ppt range for most compounds. This field deployment marks the first time MVK and MACR have been speciated at ambient concentrations with an ion trap mass spectrometer. The extended capabilities of the PTR-LIT were used to measure a variety of VOCs and BVOC species and their oxidation products. We then use these and supporting data to calculate what we refer to as product specific reactivities. For example, the organic nitrate reactivity is defined as the first order rate of production of organic nitrate from VOC oxidation by OH. Similarly, we calculate the aerosol reactivities from published aerosol yields, as well as the NO₃ reactivities, to compare the importance of different VOCs to ozone, nitrogen, and aerosol chemistry.

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Formaldehyde emissions from industrial facilities in Houston, TX

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- 1. Atmospheric and Oceanic Sciences, University of California Los Angeles, Los Angeles, CA, United States.
- 2. Earth and Atmospheric Sciences, University of Houston, Houston, TX, United States.

Body: Formation of particulates and ozone in the urban environments is driven by the OH/HO₂ radical chemistry. Consequently, it is critical to understand the OH/HO₂ budget and to quantify radical precursors, such as HCHO. Recent modeling studies of the Houston area indicate that direct formaldehyde emissions are underestimated; and flares from petrochemical facilities were proposed as potential sources of HCHO. However, little is known about the possible HCHO emissions from different types of industrial facilities since accurate measurements are sparse, in part due to a lack of techniques that allow the fence-line detection of such emissions.

Here we will present direct measurements of HCHO by an Imaging Differential Optical Absorption Spectroscopy instrument (I-DOAS) during the 2009 FLAIR/SHARP project in Houston, TX. I-DOAS, which will be described in detail, measures HCHO absorptions along 50 elevation viewing angles. By scanning in the azimuth direction, 2-D "images" of HCHO plume column densities can be derived.

During the FLAIR project, the I-DOAS was deployed at different industrial facilities in the Houston area to visualize and quantify the emissions from industrial flares and smoke stacks. We will present first observations of HCHO plumes from burning flares and other industrial sources.

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Investigation of O Production Rates in the Mexico City Metropolitan Area during MILAGRO S. Dusanter; 1; L. T. Molina; 2, 3; P. S. Stevens; 1;

- 1. Center for Research in Environmental Science, School of Public and Environmental Affairs, and Department of Chemistry, Indiana University, Bloomington, IN, United States.
- 2. Molina Center for Energy and the Environment, La Jolla, CA, United States.
- 3. Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA, United States.

Body: Understanding the oxidative capacity of the atmosphere and the formation of secondary pollutants are important issues in atmospheric chemistry. For instance, the photochemical production of tropospheric ozone (O_3) is of particular interest due to its detrimental effects on both human health and agricultural ecosystems. A detailed characterization of tropospheric O₃ production rates will help in the development of effective control strategies.

The 2006 Mexico City Metropolitan Area field campaign (MCMA-2006) was one of four components of MILAGRO (Megacity Initiative: Local And Global Research Observations) intended to collect information on the impact of megacity emissions on local, regional and global scales. In this presentation, rates of production of O_{v} ($O_{v} = O_{3} + O_{3} + O_{4}$) NO₂) species during MCMA-2006 at the supersite T0 (Instituto Mexicano del Petroleo) will be presented using different approaches based on measured and modeled concentrations of RO_x (OH + HO_2 + RO_2) radicals. In addition, we will examine both the reactivity of OH and the contribution of specific peroxy radicals to the oxidation rate of NO to estimate the contribution of groups of VOCs (alkanes, alkenes, aromatics, oxygenated and biogenic VOCs) to the total production rate of $\boldsymbol{\mathsf{O}_{_{\boldsymbol{\mathsf{Y}}}}}$ species.

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Preliminary Results of Glyoxal Measurements at CABINEX 2009

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Body: Glyoxal is of interest in tropospheric chemistry as it can serve as a tracer of both the oxidation of volatile organic compounds (VOCs) as well as the formation of secondary organic aerosol (SOA), to which it also has been proposed to contribute directly. Modeling results show that the majority of global glyoxal production results from oxidation of biogenic VOCs; however, experimental data that measure glyoxal in environments dominated by biogenic emissions are limited. Measurements of gas-phase glyoxal, the smallest α-dicarbonyl, were made at the CABINEX campaign during July and August 2009 at the PROPHET tower, which is located at the University of Michigan Biological Station, Northern Michigan, in a rural forest environment.

Glyoxal was measured using a laser-induced phosphorescence technique, which is highly sensitive due to the isolation of the long-lived phosphorescence signal from all instantaneous and fast interferences. Glyoxal was measured above the detection threshold (precision ca. 2-3 pptv/min) of the instrument throughout the campaign. Continuous sampling alternated between an inlet located 13 meters above the canopy and an inlet located in the understory on the timescale of 30 minutes. Larger nighttime concentrations of glyoxal were observed above the canopy relative to the understory throughout the campaign. During the daytime a smaller, reversed gradient was often observed. Preliminary results are presented including: vertical gradients of glyoxal; diurnal profiles; comparison with organic precursors and other volatile organic compounds; deposition processes.

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The Thermal Dissociation-Laser Induced Fluorescence (TD-LIF) technique for total Peroxy Nitrates (PNs) and comparisons to speciated PAN measurements

P. J. Wooldridge; ¹; A. E. Perring; ¹; T. H. Bertram; ²; F. M. Flocke; ³; J. M. Roberts; ⁴; H. B. Singh; ⁵; G. Huey; ⁶; J. A. Thornton; ⁷; J. G. Murphy; ⁸; J. Fry; ⁹; A. W. Rollins; ¹; B. W. LaFranchi; ¹; R. C. Cohen; ¹;

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- 2. Chemistry and Biochemistry, University of San Diego, La Jolla, CA, United States.
- 3. Atmospheric Chemistry Division, NCAR, Cambridge, MA, United States.
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- 5. Ames Research Center, NASA, Moffitt Field, CA, United States.
- 6. Earth & Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA, United States.
- 7. Atmospheric Sciences, University of Washington, Seattle, WA, United States.
- 8. Chemistry, University of Toronto, Toronto, ON, Canada.
- 9. Chemistry, Reed College, Portland, OR, United States.

Body: The UC Berkeley TD-LIF technique detects the sum of all peroxynitrates (PNs) via laser-induced fluorescence (LIF) of NO2 produced by the by thermal dissociation (TD) of peroxyacetyl nitrate (PAN) and its chemical analogues. We review the various deployments and compare the Berkeley PNs measurements with the sums of PAN and PAN-type species detected individually by other instruments. The observed PNs usually agree to within 10% with the summed individual species, thus arguing against the presence of significant concentrations of unmeasured PAN-type compounds in the atmosphere as suggested by some photochemical mechanisms. Interferences to the TD-LIF measurements are described along with strategies to minimize their effects.

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Measurements of Volatile Organic Compounds (VOCs) from Biomass Combustion – Emission Ratios, OH Reactivities and SOA Precursors

W. C. Kuster; ¹; J. B. Gilman; ^{1, 2}; P. D. Goldan; ^{1, 2}; C. Warneke; ^{1, 2}; J. deGouw; ^{1, 2}; P. R. Veres; ^{1, 2}; I. R. Burling; ³; R. J. Yokelson; ³;

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- 2. Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, United States.
- 3. Department of Chemistry, University of Montana, Missoula, MT, United States.

Body:

Multiple instruments including a gas chromatograph/mass spectrometer (GC/MS), a proton transfer reaction mass spectrometer (PTR-MS), a proton ion trap mass spectrometer (PIT-MS), a negative-ion proton-transfer chemical-ionization mass spectrometer (NI-PT-CIMS) and a Fourier-transform infrared spectrometer (FTIR) acquired data from 77 burns of various biomass fuels conducted at the U.S. Department of Agriculture (USDA) Forest Sciences Laboratory (FSL) in Missoula, MT in February 2009. We compare VOC measurements of oxygenates, aromatics and biogenics from the various instruments as well as from the various fuel types collected in southeastern and southwestern regions of the United States. The relative contribution of the combustion species to the total reactivity with the OH radical is calculated and compared to ambient air reactivity in various locations. Total reactivity for measured species in these fires occasionally exceeded 1000 sec-1 with the majority of this reactivity due to alkenes. In addition, we investigate the relative contribution of the combustion species to the potential for secondary organic aerosol (SOA) formation. Measurements of several compounds not previously reported in various urban ambient air measurement campaigns such as pyrazole (C3H4N2), pyrrole (C4H5N), benzofuran (C8H6O) and 2-furanaldehyde (C5H4O2), which are highly reactive with OH, will be presented.

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Isomeric Selective Studies of the Dominant Addition Channel in OH Initiated Oxidation of Isoprene

B. Ghosh; ¹; A. Bugarin; ¹; B. Connell; ¹; S. W. North; ¹;

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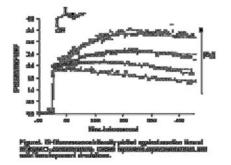
Body: We report the first isomeric selective study of the dominant isomeric pathway in the OH initiated oxidation of isoprene in the presence of O2 and NO using the Laser Photolysis-Laser Induced Fluorescence (LP-LIF) technique. The photolysis of monodeuterated/non deuterated 2-iodo-2-methyl-but-3-en-1-ol results exclusively in the dominant OH-isoprene addition product, providing important insight into the oxidation mechanism. Based on kinetic analysis of OH cycling experiments we have determined the rate constant for O2 addition to the hydroxy alkyl radical to be $(1.0\pm0.5) \times 10^{\circ}(-12) \text{ cm}^{\circ}(3) \text{ s}^{\circ}(-1)$ and we find a value of $(8.05\pm2.3) \times 10^{\circ}(-12) \text{ cm}^{\circ}(3) \text{ s}^{\circ}(-1)$ for the overall reaction rate constant of the hydroxy peroxy radical with NO. We also report the first clear experimental evidence of the (E-) form of the δ-hydroxyalkoxy channel through isotopic labeling experiments and quantify its branching ratio to be 0.1 ± 0.025 . Since it corresponds to missing carbon balance in isoprene oxidation, we have been able to identify some of the missing carbon balance. Since our measured isomeric selective rate constants for the dominant outer channel in OH initiated isoprene chemistry are similar to the overall rate constants derived from non isomeric kinetics, we predict that the remaining outer addition channel will have similar reactivity.

We have extended this study to the OH initiated oxidation of 1,3-butadiene. We have obtained isomeric selective rate constants on the dominant channel of the butadiene oxidation chemistry and measured the branching ratio for the δ -hydroxyalkoxy channel. These results on butadiene studies will be discussed.

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Measurements of OH and HO $_2$ Radical Chemistry in a Forest Environment R. F. Hansen; $^{1, 3}$; S. Dusanter; $^{2, 3}$; S. M. Griffith; $^{2, 3}$; P. S. Stevens; $^{1, 2}$;

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- 2. School of Public and Environmental Affairs, Indiana University, Bloomington, IN, United States.
- 3. Center for Research in Environmental Science, Indiana University, Bloomington, IN, United States.

Body: Approximately 30 % of the earth's surface is covered by forested areas, which make large contributions of biogenic volatile organic compounds (BVOCs) to the atmosphere. BVOCs have a significant impact on atmospheric chemistry at the local and regional scales, and, as a result, a complete understanding of the fast photochemistry involving BVOCs is important for air quality studies. Because of their high reactivity, measurements of hydroxyl (OH) and hydroperoxyl (HO₂) radical concentrations can provide a rigorous test of our understanding of atmospheric chemistry. Previous measurements of OH in forest environments have shown that models of atmospheric chemistry usually underpredict daytime and nighttime concentrations of OH, suggesting that there may be additional sources of radicals or recycling mechanisms of OH that are not included in current models of atmospheric photochemistry. During the summers of 2008 and 2009, total OH reactivity and OH and HO₂ radicals were measured above and below the forest canopy in a northern Michigan forest. Preliminary measurements are presented together with estimated production rates of OH from O₃ photolysis to ascertain whether additional primary and secondary sources of OH are required to sustain the observed concentrations. In addition, measurements from above and below the forest canopy are grouped together in several temperature bins to investigate a potential temperature dependence of the radical sources.

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Measurement and model comparison of methacrolein and methyl vinyl ketone concentrations from the OH-initiated oxidation of isoprene under NOx free conditions

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1. Center for Research in Environmental Science, School of Public and Environmental Affairs, and Department of Chemistry, Indiana University, Bloomington, IN, United States.

Body: Isoprene, the dominant natural hydrocarbon emitted into the atmosphere by deciduous trees, contributes significantly to the production of tropospheric ozone, organic nitrates, and secondary VOCs due to its high reactivity with hydroxyl radicals (OH). As a result, the chemical mechanism for the oxidation of isoprene is a subject of considerable interest in atmospheric chemistry. Recent measurements of OH and HO₂ radical concentrations in forest environments do not agree with modeled concentrations, bringing into question our understanding of the atmospheric chemistry of isoprene and other reactive biogenic emissions.

In this study, a small UV-irradiated reaction chamber was coupled to an on-line mass spectrometer to investigate the formation of isoprene oxidation products under NOx free conditions using UV-photolysis of hydrogen peroxide (H₂O₂) as the OH source. Time-resolved concentration profiles were measured for the two main products of isoprene oxidation: methacrolein (MAC) and methyl vinyl ketone (MVK). These experimental concentration profiles were compared to the values predicted by both the explicit Master Chemical Mechanism (MCM) and the condensed Regional Atmospheric Chemistry Mechanism (RACM). Several changes to the oxidation mechanism were incorporated, including the addition of recently proposed OH recycling reactions, in order to determine their impact on the predicted yields of MAC and MVK.

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OH and HO₂ Concentrations during PROPHET 2008: Measurement and Theory

S. M. Griffith; 1, 2; R. F. Hansen; 2, 3; S. Dusanter; 1, 2; P. S. Stevens; 1, 2; M. M. Galloway; 4; J. Hottle; 4, 11; A. Kammrath; 4; F. Keutsch; 4; L. H. Mielke; 5; M. Alaghmand; 5; P. B. Shepson; 5, 6; N. Zhang; 7; X. Zhou; 7; S. B. Bertman; 8; M. Carroll; 9, 10;

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- 8. Department of Chemistry, Western Michigan University, Kalamazoo, MI, United States.
- 9. Department of Chemistry, University of Michigan, Ann Arbor, MI, United States.
- 10. Department of Atmospheric, Oceanic, and Space Sciences, University of Michigan, Ann Arbor, MI, United States.
- 11. Air Force Office of Scientific Research, Arlington, VA, United States.

Body: Hydroxyl (OH) and hydroperoxy (HO₂) radicals are key species driving the gas-phase oxidation of organic trace gases that lead to the formation of ozone and secondary organic aerosols in the troposphere. Previous measurements of OH and HO_2 radicals in forest environments have shown serious discrepancies with modeled concentrations of these radicals. These discrepancies bring into question our understanding of the atmospheric chemistry of isoprene and other reactive biogenic emissions.

During the summer of 2008, OH and HO₂ (HO_x) radical measurements were made at the forested PROPHET (Program for Research on Oxidants: PHotochemistry, Emissions, and Transport) site in northern Michigan using a laser-induced fluorescence instrument developed at Indiana University. A suite of additional measurements including isoprene, methyl vinyl ketone, methacrolein, total monoterpenes, glyoxal, formaldehyde, nitrogen oxides (NO,), ozone, and UV actinic flux were measured simultaneously. These measurements were used to constrain a zerodimensional model based on the Regional Atmospheric Chemistry Mechanism to predict the expected HO_v radical concentrations. This analysis investigates individual day and night measured and modeled $\mathrm{HO}_{\mathbf{x}}$ concentrations, the $\mathrm{HO}_{2}/\mathrm{OH}$ ratio, and the production and loss processes governing HO_{χ} in this remote forested environment. Included in this analysis are 1) efforts to characterize the ozonolysis of unquantified highly reactive terpenes, which may be a potentially significant unaccounted for source of radicals in forested environments, and 2) the impact of recently proposed OH recycling in the isoprene oxidation mechanism that could lead to higher sustained OH concentrations.

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Significant OH regeneration from the photooxidation of several atmospherically important alkenes

J. Crounse; ¹; F. Paulot; ¹; J. Seinfeld; ¹; P. O. Wennberg; ¹;

1. Caltech, Pasadena, CA, United States.

Body: The reaction of RO2 with HO2 has long been thought to yield almost exclusively hydroperoxides. Recent experimental studies have provided evidence that for certain systems a significant amount of OH can also be produced. Here we present results collected during the low-NOx photooxidation of methacrolein and methylvinylketone, two main products of the atmospheric oxidation of isoprene, and 2-methyl-3-buten-2-ol, a volatile organic compound emitted by coniferous trees. All show significant OH recycling. Oxidation products were monitored using chemical ionization mass spectrometry (CIMS) methods with the CF3O- reagent ion. The sensitivity of this technique to organic hydroperoxides and the products of the alkoxy channel allows for the estimation of the OH recycling. Additional experiments were conducted using labeled 18-OH. As the recycled OH is not labeled, we obtain a second independent estimate of the OH recycling efficiency using the ratio of unlabeled to labeled products.

Currently, most chemical models (eg. MCM 3.1) do not include OH recycling for any of these systems. Inclusion of this recycling in the models will certainly improve the modeled-to-measured OH discrepancies at high isoprene levels which have been pointed out in several recent studies. Experimental details as well as the best estimates for the OH recycling efficiencies for each system will be presented.

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Measurements of OH reactivity in a South-East Asian tropical rainforest

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- 2. School of Earth and Environment, University of leeds, Leeds, United Kingdom.
- 3. Department of Chemistry, University of York, York, United Kingdom.
- 4. University of Manchester, Manchester, United Kingdom.
- 5. University of Lancaster, Lancaster, United Kingdom.

Body: The Oxidant and Particle Photochemical Processes (OP3) project took place within and above a South-East Asian tropical rainforest on the island of Borneo. Measurements of OH reactivity were made using a sliding injector flow-tube reactor with OH detection by LIF. Mean OH reactivities of 15.5 s⁻¹ were observed with a mean daily maximum of 24.7 ± 11.1 s⁻¹ shortly after local solar noon, coinciding with a peak in isoprene concentrations; minimum values of 7.2 ± 2.2 s⁻¹ were observed just before sun rise. These data are used with the simultaneous direct measurements of OH in a constrained box model to investigate the complex oxidation processes within the forest canopy. We find that the sinks of OH that were measured are unable to account for the high measured OH reactivity, so that a range of unmeasured sinks must be invoked to simulate the observations. Thus, our simultaneous measurements of OH reactivity and OH concentration enabled the separation of OH sources and sinks, allowing a more comprehensive test of our understanding of the radical chemistry occurring in this chemically complex environment.

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Impact of Climate Mitigation on Aerosol Concentrations and Health Effects in Asia

N. E. Selin; ¹; C. Wang; ¹; J. M. Reilly; ¹; S. Paltsev; ¹; R. G. Prinn; ¹;

1. Massachusetts Institute of Technology, Cambridge, MA, United States.

Body: We quantify the potential future changes in atmospheric aerosol (PM2.5) due to climate mitigation activities in Asia, and assess the impacts of these changes on human health and economic welfare. We combine approaches from atmospheric modeling using the MIT/NCAR CAM aerosol simulation, economic modeling of emissions trajectories using the MIT Emissions Prediction and Policy Analysis (EPPA) model, and health and economic impact analysis using the MIT EPPA Health Effects (EPPA-HE) model. Emissions trajectories for aerosol precursors are projected globally and for Asia with EPPA under climate mitigation scenarios to 2050 and 2100. Atmospheric simulations are conducted with MIT/NCAR CAM to project PM2.5 concentration changes, and expected health impacts and related economic costs are calculated using EPPA-HE. We find that economic co-benefits due to aerosol reductions could partially offset the costs of climate mitigation in Asia. We additionally assess the relative uncertainties in combining atmospheric modeling with economic and health impact analysis.

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Containing Climate Change With Black Carbon Reductions:

A Grand Challenge Field Experiment

V. Ramanathan; 1;

1. Scripps Institution of Oceanography, UCSD, La Jolla, CA, United States.

Body: The manmade greenhouse gases that are now blanketing the planet is thick enough to push the system beyond the tipping point for several elements of the climate system such as the arctic sea ice and the Himalayan-Tibetan glaciers, to name a few. Even with a targeted reduction in CO2 emission of 50% by 2050, we would still be adding more than 50 ppm of CO2 and thicken the manmade blanket by another 30%. Fortunately there are scientific ways to contain the warming and these will be outlined. But these need a truly transformational and interdisciplinary approach that brings together social scientists, natural scientists, energy experts and engineers to develop effective mitigation pathways. Towards this goal an interdisciplinary team of academics, NGOs and intergovernmental organizations from US, Europe and India have developed Project Surya to drastically decrease emissions of the major non-CO2 climate warmers (soot, methane, ozone precursor gases) from rural areas in India and China. Surya will undertake the most comprehensive data collection, to-date, on the impact of reducing biomass burning on climate forcing, health and the wellbeing of rural inhabitants most of whom live under a dollar a day. The experiment thus offers the opportunity to field test our ideas and hypotheses about the impact of black carbon and brown clouds on dimming, the Asian monsoon and the melting of the Himalayan-Tibetan glaciers. The data from this soft 'geo-engineering' experiment is also anticipated to lead to a sustainable way of energy consumption for the roughly 4 billion who are forced to use solid bio-fuels for all of their energy needs.

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Aerosols in Asia and Global-to-Continental-scale Climate Change (Invited)

V. Ramaswamy; 1;

1. NOAA/ GFDL, Princeton, NJ, United States.

Body: Aerosols arising as a result of human-induced emissions and originating in the continental areas in the Northern Hemisphere have perturbed the Earth's radiative energy balance and become a key 'driver' of global and continental climate change. The Asian climate regime is a particularly critical element of the Earth System and represents a singularly important science challenge. The NOAA/ Geophysical Fluid Dynamics Laboratory's climate model is employed to compare the effects of the anthropogenic aerosols with that of the long-lived greenhouse gases on the global-mean to continental spatial scales. On the basis of model simulations and observations, it is evident that aerosols have provided an "offset" to the warming tendency due to increases in the greenhouse gases in the 20th century. In the Asian region which has experienced substantial changes in aerosols over the past fifty years, simulations reveal a marked sensitivity of the climate variables (e.g., surface temperature, precipitation) to the evolution of sulfate and carbonaceous aerosol concentrations. However, significant uncertainties remain concerning the changes in the various aerosol species and the degree of aerosol absorption that has occurred in Asia. We address the uncertainties associated with aerosols which, in turn, affect the inferences about the net anthropogenic radiative forcing, climate response, detection-attribution of climate change, and climate projections. Of central importance are: the roles of the different aerosol species; aerosol microphysics; aerosol-cloud interactions; and the global impact of localized aerosol changes on the 21st century climate projections. The differing roles of long-lived greenhouse gases and various aerosol species on climate have significant implications for decision processes seeking policy options to adapt to and mitigate climate change.

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Changes in snow cover and water cycle over the Tibetan plateau induced by absorbing aerosols

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- 2. U. Maryland, Baltimore County, Baltimore, MD, United States.
- 3. Kongju University, Gongju, Korea, Republic of.
- 4. Kongju University, Gongju, Korea, Republic of.

Body: The warming of the land surface and retreating glacier and snowpack in Hindu-Kush-Himalayas-Tibet (HKHT) are well known observations often attributed to effect of greenhouse warming. In this study, based on numerical experiments with the NASA fvGCM, we find that the elevated -heat-pump (EHP) effect by absorbing aerosols (dust and black carbon) over the Indo-Gangetic Plain and Himalayas foothills can lead to substantial warming of the atmosphere and land surface, and reduction in snow cover over the HKHT region. Atmosphere and surface energy analyses show that beginning in April, the middle atmosphere, near the high-altitude surface over the Tibetan Plateau, heats up due to absorption of solar radiation by black carbon and dust. The near surface heating increases convection, which spreads the warming to the upper troposphere over the Plateau. As the monsoon season approaches in May, the increased convection draws in warmer and moister monsoon air into the HKHT, which further enhances the convection, cloudiness and precipitation over the region in late May and early June. The moister and warmer atmosphere over the HKHT region suppresses evaporation and sensible heat fluxes from the surface during April-May. The excess heat received by the surface goes into melting of the snowpack, exposing more of the bare land. The exposed land surface leads to further increase in land surface temperature over the HKHT.

Results show that most of the snow melt occurs in the western region (west of 90E) in April and May, when black carbon aerosol loading in the atmosphere is highest, and dust loading is on the rise. The equivalent surface albedo change due to snow melt is reduced by about 8-10%, and 4-6% over the western and eastern TP respectively. The proposed mechanism appear to be relevant on interannnual time scales and beyond. The consequence of these changes in HKHT on the water cycle of Asia will be discussed.

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Estimation of black carbon deposition from particulate data in the atmosphere at NCO-P site in Himalayas during premonsoon season and its implication to snow surface albedo reduction

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- 4. Laboratoire de Glaciologie, CNRS, Université J. Fourier, Grenoble, France.
- 5. Graduate School of Environmental Studies, Nagoya University, Nagoya, Japan.
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Body: The black carbon (BC) impact on snow surface may contribute to snow melting and acceleration of glacier retreat. The BC deposition amount onto snow surface in 2006 during pre-monsoon season (March-May) was estimated from the observed equivalent BC (eqBC) concentration (MAAP) and aerosol size distribution observation (SMPS and OPC) in the atmosphere at Nepal Climate Observatory at Pyramid (NCO-P) site in Himalayan region. We, first, carried out correlation analyses in time series data between the eqBC and aerosol size distribution and then determined main eqBC size range here as higher correlations coefficient of more than 0.8. The corresponding eqBC size at NCO-P site was determined predominantly in the 103.1-669.8 nm size range. Simply terminal velocity for each particle size bin was used for calculating deposition flux of BC onto surface. Our estimation of the deposition is considered to be minimal estimation because deposition velocity is in general faster if we include aerodynamic and other terms; moreover we didn't take into account deposition processes other than gravitational deposition. We estimated the BC deposition of 209 µg m-2 for March-May. If we use snow density variations in surface snow of 192-512 kg m-3, as measured at Yala glacier in Himalayas, the BC concentrations in 2-cm surface snow of 20.4-53.6 µg kg-1 is estimated. This leads to a snow albedo reduction of 1.6-4.1% by using regression relationship between BC concentration in snow and snow albedo reductions by previous studies. If we used the values of the albedo reductions as continuous forcing for a sensitivity test of glacier melting by using a mass-balance model with the same initial settings in a previous study (pointed out for Dongkemadi Glaciers in Tibetan region), increase of total melt water runoff of 54-149 mm w.e. is expected. We are aware of the limitation of this preliminary estimate but it is important to consider that it clearly indicates that BC deposition during March-May may significantly reduce snow surface albedo leading to potential impacts on Himalayan glacier dynamics.

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Heavy pollution suppresses light rain in China: Observations and modeling

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Body: Long-term observational data reveal that both the frequency and amount of light rain have decreased in eastern China (EC) for 1956–2005 with high spatial coherency. This is different from the trend of total rainfall observed in EC, which decreases in northern EC and increases in southern EC. To examine the cause of the light rain trends, we analyzed the long-term variability of atmospheric water vapor and its correlation with light rain events. Results show very weak relationships between large-scale moisture transport

and light rain in EC. Because of human activities, pollutant emission has increased dramatically in China for the last few decades, leading to a significant reduction in

visibility between 1960 and 2000. Cloud-resolving model simulations over EC show that aerosols corresponding to polluted conditions can significantly increase the cloud

droplet number concentration (CDNC) and reduce droplet sizes compared to pristine conditions. This can lead to a significant decline in raindrop concentration and delay

raindrop formation because smaller cloud droplets are less efficient in the collision and coalescence processes.

Together with weaker convection, the precipitation frequency and amount are significantly reduced in the polluted case in EC. Satellite data also reveal

higher CDNC and smaller droplet size over polluted land in EC relative to pristine regions, which is consistent with the model results. Observational evidences and simulations

results suggest that the significantly increased aerosol concentrations produced by air pollution are responsible for the decreased light rain events observed in China over the past 50 years.

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Aerosols and Regional Climate Changes

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Body: In this work, 45 years (1961-2005) of hourly meteorological data in Taiwan, including temperature, humidity, and precipitation have been analyzed with emphasis on their diurnal asymmetries. We find a significant increase of nighttime temperature throughout Taiwan, but little trend in daytime temperature. Effects of anthropogenic aerosols, direct and/or indirect effects, are the major contributor to the lack of trend in the daytime. We find a long term decreasing trend for relative humidity (RH) and the trend is significantly greater in the nighttime than in daytime, apparently due to a greater warming at night. The warming at night in three large urban centers is large enough to impact significantly on the average temperature trend in Taiwan between 1910 and 2005. There is a decrease in the diurnal temperature range (DTR) that is largest in major urban areas, and becomes smaller but doesn't disappear in smaller cities and offshore islands. The nighttime reduction in RH is likely the main cause of a significant reduction of fog events over Taiwan. The smaller but consistent reductions in DTR and RH in the three off-coast islands suggests that, in addition to local land-use changes, a regional scale process such as the indirect effect of anthropogenic aerosols may also contribute to these trends.

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Direct and Indirect Effects of Aerosols in China

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- 3. NASA/GSFC, Greenbelt, MD, United States.
- 4. Lanzhou University, Lanzhou, China.

Body: By modulating atmospheric heating profile, surface energy balance and cloud microphysics, the heavy loading of aerosols in China have been hypothesized to interact with the Asian monsoon system and play a significant role in observed changes in precipitation, temperature and atmospheric circulation. Testing the hypotheses requires extensive and reliable measurements concerning their properties, radiative fluxes, cloud microphysics, precipitation, and other atmospheric variables, which is the primary goal of two major ongoing field campaigns conducted before, during and after the Beijing Olympic Games under the East Asian Study of Tropospheric Aerosols: an International Regional Experiment (EAST-AIRE) and the ARM Mobile Facility deployment in China (AMF-China). In my talk, I will review the status of the two observation campaigns; present some preliminary findings; elaborate the potential usage of the data in dealing with the aforementioned issues.

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An overview of dust aerosol effect on semi-arid climate during 2008 China-US joined field campaign

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Body: To improve understanding and capture the direct evident of the impact of dust aerosol on semi-arid climate, the 2008 China-US joined field campaign are conducted. Three sites involved this campaign, including one permanent site (Semi-Arid Climate & Environment Observatory of Lanzhou University (SACOL)) (located in Yuzhong, 35.95°N/104.1°E), one SACOL's Mobile Facility (SMF) (deployed in Jintai, 37.57°N/104.23°E) and the U.S. Department of Energy Atmospheric Radiation Measurements(ARM) Ancillary Facility (AAF mobile laboratories, SMART-COMMIT) (deployed in Zhangye, 39.08°N/100.27°E). This study presents a description the objectives, measurements, and sampling strategies for this joined campaign. Major dust episodes captured during the campaign were investigated. Preliminary observation results show that the semi-direct effect may be dominated by the interaction between dust aerosols and clouds over arid and semi-arid areas and partly contribute to reduced precipitation. These results suggest that the local anthropogenic and nature absorbing aerosols make significant contribution to the regional interaction among aerosol-cloud-radiation-precipitation processes and need to be future investigation.

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Air pollution and clouds in southern China: preliminary results from the observations in spring 2009 at Mt. Heng

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Body: Aerosols and clouds play a key role in climate change, and the interaction between them also affect the chemical transformation and removal of air pollutants. As a part of China's National Basic Research Project (National '973 Project) on acid deposition, trace gases, aerosols, and cloud water composition were measured in March-May 2009 at the summit of Mount Heng in southern China (Long: 112° 42' E, Lat: 27° 18' N, 1269 m a.s.l.). The preliminary results from this study are presented. Frequent clouds/fogs occurred at this site. Despite its southern location, the impact of a dust storm from northern China was observed in late April during which the PM10 concentration exceeded 800 ug/m3. Moderate levels of pollution were observed. Formation and growth of new particles frequently occurred on clear days as indicated from the measurement of particle number and size distributions. Cloud water was mostly acidic with a mean acidity of 3.75 (range: 2.51-6.91). The chemical data from this study are compared with those from Mt Tai to show the difference in southern and northern China. The sources of air pollution at this mountain site and the implications of the results will be discussed.

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Aerosol Properties Observed on the Mesoscale and MODIS Aerosol Optical Depth Comparisons during the 2008 Premonsoon TIGERZ IOP in Kanpur, India

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Body: The Indo-Gangetic Plain (IGP) in northern India produces anthropogenic pollution from urban, industrial, and rural combustion sources nearly continuously with convection-induced winds driving desert and alluvial dust into the atmosphere during the pre-monsoon period. The NASA Aerosol Robotic Network (AERONET) project initiated the TIGERZ measurement campaign in May 2008 with an intensive operational period (IOP) from May 1 to June 23, 2008. One component of the TIGERZ IOP assessed the mesoscale spatial variability of aerosols in the IGP around the industrial city of Kanpur, India. Data collected during TIGERZ spatial variability study (SVS) deployments indicated higher average aerosol optical depth (AOD, τ) at 500nm downwind of Kanpur (Δτ500=0.03-0.09). In addition, these SVS AOD averages were compared to the long-term Kanpur AERONET monitoring site data. Four SVS AOD averages were within 1σ of the climatological mean at the AERONET Kanpur site (τ500=0.75±0.26 for May; τ500=0.78±0.43 for June), while one SVS average (τ500=0.31±0.02) was within 2σ of the May monthly climatological mean. Aerosol absorption properties derived from AERONET inversion retrievals for the TIGERZ IOP were evaluated with respect to in situ measurements from other regional aerosol campaigns and compared to typical values from the Kanpur AERONET site. For one case study, the measured 440-870nm Angstrom exponent of ~0.38, the retrieved 440-870nm absorption Angstrom exponent (AAE) of 1.15-1.53, and the retrieved sphericity parameter near zero suggest the occurrence of large, strongly absorbing, non-spherical aerosols over Kanpur, such as mixed black carbon and dust; and the variation among the SVS sites indicated lower AAE values (i.e., more absorbing aerosols) downwind of the city. Furthermore, the 10km standard Terra (MOD04_L2) and Aqua (MYD04_L2) MODIS C005 algorithm and Aqua (MYD04 L2) MODIS Deep Blue C051 algorithm retrievals at τ550 were compared to the interpolated TIGERZ IOP dataset. For MODIS/AERONET Kanpur site matchups during the May and June 2008 premonsoon period, the correlation coefficients (R) for the C005 MOD04_L2, C005 MYD04_L2 and C051 MYD04 L2/Deep Blue algorithms were 0.72, 0.83 and 0.83, respectively, for the lowest quality level (QA≥0), while Deep Blue showed a high τ550 bias with a slope near one. Although MODIS retrievals at higher quality levels were near or within the stated retrieval uncertainty [±(0.05 + 0.15_T)], the total number of MODIS matchups (N) was significantly reduced with quality level over Kanpur during the pre-monsoon TIGERZ IOP (N=25, QA≥0; N=9, QA≥1; N=6, QA≥2; N=1, QA=3) primarily due to the complex surface terrain (urban and barren cropland), complex aerosol mixture (black carbon and dust) and cloud-contaminated pixels.

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Aerosol and Cloud Properties during the Cloud Cheju ABC Plume -Asian Monsoon Experiment (CAPMEX) 2008:

Linking between Ground-based and UAV Measurements

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Body: Cheju Atmospheric Brown Cloud (ABC) Plume-Monsoon Experiment (CAPMEX), comprehsensive groundbased measurements and a series of data-gathering flights by specially equipped autonomous unmanned aerial vehicles (AUAVs) for aerosol and cloud, had conducted at Jeju (formerly, Cheju), South Korea during August-September 2008, to improve our understanding of how the reduction of anthropogenic emissions in China (so-called "great shutdown") during and after the Summer Beijing Olympic Games 2008 effcts on the air quliaty and radiation budgets and how atmospheric brown clouds (ABCs) influences solar radiation budget off Asian continent. Large numbers of in-situ and remote sensing instruments at the Gosan ABC observatory and miniaturized instruments on the aircraft measure a range of properties such as the quantity of soot, size-segregated aerosol particle numbers, total particle numbers, size-segregated cloud droplet numbers (only AUAV), aerosol scattering properties (only ground), aerosol vertical distribution, column-integrated aerosol properties, and meteorological variables. By integrating ground-level and high-elevation AUAV measurements with NASA-satellite observations (e.g., MODIS, CALIPSO), we investigate the long range transport of aerosols, the impact of ABCs on clouds, and the role of biogenic and anthropogenic aerosols on cloud condensation nuclei (CCN). In this talk, we will present the results from CAPMEX focusing on: (1) the characteristics of aerosol optical, physical and chemical properties at Gosan observatory, (2) aerosol solar heating calculated from the ground-based micro-pulse lidar and AERONET sun/sky radiometer synergy, and comparison with direct measurements from UAV, and (3) aerosol-cloud interactions in conjunction with measurements by satellites and Gosan observatory.

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Using unmanned aircraft to measure the impact of pollution plumes on atmospheric heating rates and cloud properties during the Cheju ABC Plume-Asian Monsoon Experiment (CAPMEX)

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- 4. Civil and Environmental Engineering Department, University of Wisconsin-Madison, Madison, WI, United States.

Body: The CAPMEX (Cheju ABC Plume-Asian Monsoon Experiment) campaign took place off the Coast of Cheju Island in South Korea to take advantage of the unique event associated with the shutdown of anthropogenic emissions surrounding Beijing during the Olympics in summer 2008. CAPMEX studied pollution plumes before, during, and after the Beijing reductions using ground-level and high-elevation measurements, i.e., from unmanned aircrafts. Additionally, the campaign documented the effect on solar heating and clouds due to aerosols carried by the long range transport of pollution plumes.

The unmanned aerial vehicle (UAV) measurement component of this campaign took place during Aug 9 to Sept 30, 2008. The AUAV payload was mission-specific and was outfitted to perform a particular set of measurements. These measurements include aerosol concentration, aerosol size distribution, aerosol absorption, cloud drop size distribution, solar radiation fluxes (visible and broadband), and spectral radiative fluxes. Throughout the CAPMEX experiment, long-range transport of aerosols from Beijing, Shanghai and Marine plumes were sampled in aerosol layers up to 3-4 km above sea level. During this period, we captured both heavy and light pollution events and witnessed air masses from both pristine oceanic sources and from major cities including Beijing and Shanghai. Analysis of specific plumes allowed us to quantify the impact of anthropogenic pollution on heating rates and cloud properties.

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Final ID: A14D-06

SOA Measurements vs. Models: a Status Report

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Body: The advent of fast and more detailed organic aerosol (OA) and VOC measurements in the last decade has allowed clearer model-measurement comparisons for OA and secondary OA (SOA). Here we summarize the patterns emerging from studies to date.

At least 8 studies have reported a large (x5-10) underestimation of SOA for polluted regions when using traditional models (those developed until ~2006) (Heald GRL05, Volkamer GRL06, Johnson ACP06, Kleinman ACP08, Matsui JGR09, Dzepina ACP09, Hodzic ACP09, Tsimpidi ACP09). This is especially obvious when models are evaluated with the Δ OA/ Δ CO ratio.

Close to pollution sources, discrepancies of an order-of-magnitude in SOA lead to smaller discrepancies (often x2-3) for total OA due to the presence of primary OA (de Gouw EST09). Such OA discrepancies have been repeatedly observed (e.g. Vutukuru JGR06, McKeen JGR07&09, Heald JGR07, Fast ACP09, Hodzic ACP09).

The discrepancy is reduced when recently-updated yields for aromatics (Ng ACP07) and SOA from glyoxal (Volkamer GRL07) are used, and is eliminated when using SOA formation from S/IVOC (Robinson Sci07) although with an overprediction of SOA at long aging times (Dzepina ACP09; Hodzic ACP09b). It is not clear whether the urban discrepancy is removed for the right reasons.

4 evaluations of biogenic SOA formed in unpolluted regions find reasonable agreement between SOA from traditional models and field measurements (Tunved Sci06; Hodzic ACP09; Chen GRL09; Slowik ACPD09). One evaluation reports a significant underprediction (Capes ACP09), although the amount of precursor reacted was difficult to ascertain for that case. The difference with the systematic underprediction observed for anthropogenic SOA may be due to the lack of primary S/IVOC in biogenic emissions, or to other reasons (NOx, SO2, POA, etc.).

Comparisons for biogenic SOA formed in polluted regions are more complex. Several studies have reported a lack of clear influence of biogenic VOCs in SOA formation in polluted regions (de Gouw JGR05, GRL09; Weber JGR07; Bahreini JGR09), but 14C studies suggest a large fraction of modern C (Weber JGR07). Synergistic effects of pollution and BVOCs appear likely (e.g. Goldstein PNAS09).

SOA from biomass burning emissions appears variable in the field (Capes JGR09; Yokelson ACP09), likely due flaming vs. smoldering fraction and biomass identity, and perhaps also to prompt SOA formation triggered by HONO photolysis. Several studies report significant SOA formation, given enough photochemical processing. Models based on traditional precursors appear to underpredict SOA from BB sources (Grieshop ACP09; Hodzic ACP09).
The very large SOA source in the free troposphere postulated by Heald (GRL05) has not been reported in later

studies. Dunlea (ACP09) did not find evidence of this source across the Pacific near North America, though precipitation removal precludes any strong conclusions. However Carlton (EST09) reported better comparisons after implementing in-cloud SOA formation.

Future model evaluations should compare POA, SOA precursors, OVOCs, oxidants, and boundary conditions. Multiple OA measurements (WSOC, OC, AMS, molecular tracers, 14C, etc.) are necessary to overcome the limitations of any one method. Measurements of semivolatile species are critically needed to constrain models.

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FM09: Question regarding A14D-06'>click here to send an email

Final ID: A14D-07

Contributions of Selected Biogenic and Aromatic Compounds to the Formation of Tropospheric Secondary Organic Aerosol over Several Sites in the United States

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- 2. U.S. Environmental Protection Agency, Research Triangle Park, NC, United States.

Body: The National Exposure Research Laboratory of the U.S. Environmental Protection Agency recently undertook an integrated laboratory and field research effort to better understand the contribution of biogenic and aromatic hydrocarbons to the formation of submicron ambient secondary organic aerosol (SOA). In the laboratory, isoprene, α-pinene, β-caryophyllene, 1,3-butadiene, 2-methyl-3-buten-2-ol, benzene, and toluene were individually irradiated under a wide range of conditions in a photochemical reaction chamber in the presence of nitrogen oxide (NOx). These hydrocarbons are thought to contribute to ambient SOA formation. In field studies conducted in Research Triangle Park, NC; Duke Forest in Chapel Hill, NC; Atlanta, GA; Pensacola, FL; Birmingham and Centerville, AL; Riverside, CA; Detroit, MI; Northbrook, East St. Louis and Bondville, IL; and Cincinnati, OH, ambient PM2.5 samples were collected for various periods between 2003 and 2006. The SOA collected from these laboratory experiments and the ambient PM2.5 samples were analyzed for organic carbon (OC) concentration and for organic tracer compounds by GC-MS using BSTFA derivatization for their identification and quantification. An organic tracer-based method was developed for estimating ambient SOA concentrations from individual SOA precursors to allow an assessment of SOA model predictions with ambient data.

The results show that several major reaction products detected in SOA formed in the laboratory photooxidations were among the major compounds detected in field samples, effectively connecting laboratory and field results. Using the tracer-based method, the contributions of isoprene and monoterpenes to SOA formation show strong seasonal dependencies. However, no clear seasonal variations were observed for sesquiterpenes and aromatic hydrocarbons. The contribution of 2-methyl-3-buten-2-ol to ambient SOA was found to be not only season dependent but also higher in locations dominated by conifers, which are believed to be the major emitters of 2-methyl-3-buten-2-ol into the atmosphere. The ratios of secondary organic carbon (SOC) to OC for these locations ranged from 20% during the winter to 50% and occasionally higher during the summer. In addition, during the winter season, levoglucosan (a biomass burning tracer) appeared to be a dominant contributor to ambient PM2.5 OC concentrations, while the SOA contributions remained low. However, during the summer, the SOA contributions of isoprene, monoterpenes, sesquiterpenes, and aromatic hydrocarbons along with OC concentrations increased, while the biomass burning contribution decreased. The limitations of these methods will also be addressed.

Disclaimer: This work has been funded wholly or in part by the United States Environmental Protection Agency under Contract EP-D-05-065 to Alion Science and Technology. It has been subjected to Agency review and approved for publication.

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Final ID: A14D-08

An AEROCOM Intercomparison Exercise in Organic Aerosol Modeling

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Body: Comparisons of individual models with organic aerosol (OA) measurements have shown a large underestimate of the OA component by the models. These discrepancies are very high in the free troposphere. Trying to bridge the gap between models and observations, several recent model developments account for secondary OA (SOA), intermediate volatility organics, multiphase chemistry and semi-volatile primary OA (POA).

OA simulations have many degrees of freedom due to the missing knowledge on the behavior and fate of both POA and SOA in the troposphere. Thus, several assumptions need to be made and are translated to model tuning parameters that vary greatly from one model to the other. The organic aerosol intercomparison AEROCOM exercise aims to evaluate the actual status of global modeling of the OA occurrence in the global troposphere and analyze discrepancies between models as well as between models and observations. It will quantify the uncertainties and attribute them to major contributors. It will also try to identify and analyze potential model systematic biases. The ensemble of the simulations will be used to build an integrated and robust view of organic aerosol sources and sinks in the troposphere.

The year 2006 was selected as the base year for the study. The first results of the intercomparison will be presented together with a compilation of field data that will be used for the validation of the models results.

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Global transport of Asian dust revealed by NASA/CALIPSO and a global aerosol transport model

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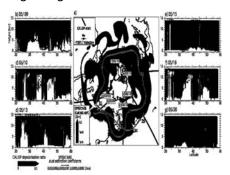
Body: Trans-Pacific transport of mineral dust and air pollutants originating from Asia to North America is well known. Eguchi et al. (2009, ACP) pointed out that the Taklimakan Desert supplies mineral dust for upper troposphere and can play an important role in intercontinental-scale dust transport. Asian dust is also detected from ice cores on Greenland and French Alps. The effects of Asian dust on cloud systems and the associated radiative forcing can extend over the Northern Hemisphere. In this study, we report the detailed structure of Asian dust during the global transport using integrated analysis of observations by CALIOP on-boarded NASA/CALIPSO satellite and a glocal aerosol transport model.

We used the CALIOP Level 1B data products (ver. 2.01), containing the total attenuated backscatter coefficients at 532/1064 nm and the volume depolarization ratio at 532 nm. Dust extinction coefficients are then derived from the Fernald's inversion method by setting the lidar ratio to S1=50 sr. As for a global aerosol transport model, we used the Spectral Radiation Transport Model for the Aerosol Species (SPRINTARS; Takemura et al., 2005, JGR). We performed a sensitivity experiment that aims at an analysis specified for a single dust event originating from the Taklimakan. The simulation was performed over May 2007.

A sever dust storm occurred on 8–9 May 2007 in Taklimakan Desert. Dust cloud emitted during this dust storm is uplifted to altitude of 8–10 km and starts the travel of full circuit around the globe. It has a meridional width of 100–200 km. About one tenth of the original uplifted dust mass (8.1 Gg) is encircling the globe taking about 2 weeks. Because of its high transport height, the dust cloud almost unaffected by wet removal so that the decay of its concentration level is small. Over the western North Pacific of 2nd circuit, the dust cloud pulls down to the lower troposphere by anticyclonic down draft, and finally it settles on North Pacific because of wet removal process. We found that Asian dust can influence the global radiation budget by stimulating cirrus cloud formation and marine ecosystems by supplying nutrients to the open ocean.

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a) CALIPSO orbit path, HYSPLIT dust trajectory and SPRINTARS simulated dust extinction coefficient alone the HYSPLIT trajectory. Number indicates the date of May 2007; (b)–(g) the latitude-height cross-section of CALIOP observed depolarization and SPRINTARS dust extinction coefficient. X denotes the position of HYSPLIT trajectory crossing at each section.

Variations of OC and EC concentrations in PM10, PM2.5 and PM1.0 in Jeju.

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Body: PM10, PM2.5 and PM1.0 were measured at Gosan-super site in Jeju Island during from August 2007 to June 2008. Mean concentrations of PM10, PM2.5 and PM1.0 were 29.28 μg/m\$^{3}\$, 17.83 μg/m\$^{3}\$ and 14.30 μg/m\$^{3}\$, respectively. PM2.5/ PM10 was 60.9% and PM1.0/ PM10 was 48.8% with 80.2% of PM1.0/ PM2.5. While nitrate showed different size-distribution in PM1.0-10 and PM1.0 with higher concentration in PM1.0-10, sulfate was subsequently dominant in PM1.0. When high concentration episodes were occurred, nitrate was accompanied by mass in PM1.0-10, however both mass and nitrate and sulfate increased in PM1.0, especially nitrate. Nitrate and sulfate showed larger increase with northwest wind, while relatively higher sulfate appeared with southeast wind, too. PM2.5/ PM10 was 84.8(99.7)% for OC(EC), PM1.0 / PM10 was 67.8(84.2)% and PM1.0 / PM2.5 was 80.0(42.6)%. OC and EC were much higher in PM1.0 and EC in PM1.0-2.5 was distinguishable. High concentration episodes considered, nitrate and sulfate were higher during episodes periods of EC than that of OC, because it's likely that EC was emitted from anthropogenic source and had a large surface area. Most of all, nitrate increase during episodes periods of EC and OC was marked feature, which was 2 times larger than mean concentrations. The further work about high concentration episodes of OC and EC will be presented in the meeting.

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Integrated Analysis of Bottom-up Emissions Information and Airborne Measurements in Support of NASA INTEX and ARCTAS Field Campaigns

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Body: Intercontinental Chemical Transport Experiment (INTEX)-B was an airborne campaign conducted during a 10-week period from March 1 to May 15 in 2006 to understand the transport and transformation of gases and aerosols on transcontinental/intercontinental scales and assess their impact on air quality and climate. In April and July of 2008, NASA also sponsored the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission that coincided with other International Polar Year (IPY) activities seeking to better understand the behavior of Polar Regions and their roles in the broader Earth System. High-resolution chemical measurements made onboard the NASA DC-8 aircraft during INTEX-B and ARCTAS offer the opportunity to broaden our understanding of the atmospheric behavior of chemicals for Arctic and mid-latitude regions. An emissions inventory and processing methodology have been developed to elucidate the influence of Asian emissions outflow. Remote Sensing-based land cover and atmosphere information has also been included to analyze the effect of plants uptake and emissions. The results of our combined data analysis will be presented at the conference.

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Particle Formation and Growth in North and East China: a Comparative Analysis of Measurements from Surface and

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- 4. Nanjing University, Nanjing, China.

Body: Particle formation and growth is a key process that determines the size distribution of aerosols in the atmosphere. In this report we present the measurement result of number and size distribution of aerosol particles (10–10000nm in diameter) obtained in spring and summer of 2005-2007 in North and East China which experience serious regional air pollution problems. The measurements were conducted at an urban area (Ji'nan, Shandong), a rural area in Beijing, and the summit of Mount Tai (1534 m a.s.l.) in Shandong. The previously reported data collected in a suburban area near Shanghai was also used. The data are examined in terms of the frequency of particle formation, the growth characteristics, and an estimate of the contribution of sulfuric acid vapor to particle growth in highly polluted atmospheres. Despite the large condensational sinks due to the presence of pre-existing particles, intensive formation and growth of particles were frequently (20%-65% of time) observed at the suburban and rural areas, both in surface and the mountain-top sites. The much higher frequency (65%) at Mt Tai suggests that the particle formation and growth is a regional phenomenon occurring not only in the surface but also throughout the planetary boundary layer in eastern China. The observed particle growth rates varied from 3.1 nm/h at the mountain top to 4.5-6.0 nm/h near Beijing and Shanghai. The source rates of condensable vapors were estimated at 0.91+E6 cm-1 s-1 in Mt Tai, 1.80+E6 cm-1 s-1 in Beijing, and 3.08+E6 cm-1 s-1 in Shanghai. The condensation of sulphuric acid vapor contributed about 45% of the particle growth in Shanghai and at Mt Tai, and about 25% in Beijing. These values are higher than those from previous researches at similar latitudes.

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Spatial and temporal variations of aerosols around Beijing in summer 2006: Model evaluation and source

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- 2. Ibaraki University, Ibaraki, Japan.
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- 4. Peking University, Beijing, China.
- 5. Chung Shan Medical University, Taichung, Taiwan.
- 6. University of California-Irvine, Irvine, CA, United States.
- 7. Pacific Northwest National Laboratory, Richland, WA, United States.
- 8. Argonne National Laboratory, Argonne, IL, United States.

Body: Regional aerosol model calculations were made using the WRF-CMAQ and WRF-chem models to study spatial and temporal variations of aerosols around Beijing, China, in the summer of 2006, when the CAREBEIJING-2006 intensive campaign was conducted. Model calculations captured temporal variations of primary (such as elemental carbon, EC) and secondary (such as sulfate) aerosols observed in and around Beijing. The spatial distributions of aerosol optical depth observed by the MODIS satellite sensors were also reproduced over northeast China. Model calculations showed distinct differences in spatial distributions between primary and secondary aerosols in association with synoptic-scale meteorology. Secondary aerosols increased in air around Beijing on a scale of about 1000 x 1000 km2 under an anticyclonic pressure system. This airmass was transported northward from the high anthropogenic emission area extending south of Beijing with continuous photochemical production. Subsequent cold front passage brought clean air from the north, and polluted air around Beijing was swept to the south of Beijing. This cycle was repeated about once a week and was found to be responsible for observed enhancements/reductions of aerosols at the intensive measurement sites. In contrast to secondary aerosols, the spatial distributions of primary aerosols (EC) reflected those of emissions, resulting in only slight variability despite the changes in synoptic-scale meteorology. In accordance with these results, source apportionment simulations revealed that primary aerosols around Beijing were controlled by emissions within 100 km around Beijing within the preceding 24 hours, while emissions as far as 500 km and within the preceding 3 days were found to affect secondary aerosols.

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The Effects of Dust Emissions on the Summertime Climate of Southwest Asia: Incorporating Sub-grid Variability in a Regional Climate Model's Dust

Scheme

M. P. Marcella; 1; E. A. Eltahir; 1;

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Body: Improvements are made to the dust scheme in a regional climate model by representing sub-grid variability of wind speeds and surface roughness lengths. The new module quantifies wind variability by using model meteorology, assuming wind speeds follow a Gaussian distribution. More specifically, wind variability is approximated by dry convective eddies within the boundary layer, forced by surface sensible heat fluxes. Incorporating sub-grid variability of wind increases dust emissions over Southwest Asia by nearly 40% while reducing incoming shortwave radiation by 5-15 W/m^2. Likewise, the dust scheme is modified to include variability of surface roughness length over Southwest Asia. An empirical distribution of roughness length for each grid-cell is calculated based on the USGS's 4km resolution land cover data. However, incorporating roughness length variability does not significantly alter dust emissions. Nevertheless, aerosol optical depth values in simulations including the spatial variability of wind lie closer to observations.

Using the improved dust module in RegCM3, this work also presents the effects of dust on the surface summertime climate of Southwest Asia. It is shown that dust emissions reduce average summertime surface temperatures by approximately 0.5-1.0C while attenuating shortwave incident radiation by 35 W/m^2. Likewise, analysis of the diurnal cycle of energy and surface temperatures yields significant changes caused by the radiative effects of dust. That is, afternoon temperatures are reduced by over 1C while shortwave radiation is reduced by nearly 100 W/m^2. Lastly, results indicate that dust alters not only mean temperature, but also extreme daily temperatures experienced over Southwest Asia.

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Optical Properties of Fine/Coarse Mode Aerosol Mixtures

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- 9. Naval Research Laboratory, Monterey, CA, United States.
- 10. Universite de Sherbrooke, Sherbrooke, QC, Canada.

Body: Several regions of the earth exhibit seasonal mixtures of fine and coarse mode sized aerosol types, which are challenging to characterize from satellite remote sensing. Over land the coarse mode size aerosols (radius >1 micron) originate primarily from arid regions, which generate airborne soil dust, and the dominant fine mode sources are gases and particulates from urban/industrial emissions and from biomass burning. AERONET sun-sky radiometer almucantar retrievals from several years are analyzed for the urban sites of Beijing, China and Kanpur, India (in the Ganges floodplain) where seasonal coarse mode dust particles mix with fine mode pollution aerosol, predominately in the spring. As increasingly more absorbing fine mode pollutants are added to the dust aerosol at both sites, the single scattering albedo (SSA) of the mixtures at 675 nm through 1020 nm decrease as the fine mode fraction of AOD increases, while the 440 nm SSA is relatively constant. Additionally we compare multi-year data from Ilorin, Nigeria where desert dust from the Sahara and Sahel mix with fine mode biomass-burning aerosols. The volume size distribution retrievals from this site often shows tri-modality (third mode centered at 0.6 micron radius), which suggests a different particle source than found for most other arid region AERONET sites, which typically have bi-modal distributions. Comparison of mid-visible single scattering albedo obtained from in situ aircraft measurements during DABEX to multi-year means from the Ilorin site AERONET retrievals show close agreement (within 0.03 or less) over a wide range of Angstrom exponent (0.3 to 1.5). Observed differences in the spectral SSA as a function of fine mode fraction of the optical depth between all three sites are discussed and occur due to differences in absorption for both modes and also due to fine mode particle size dynamics.

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Climate response to black carbon aerosols: Dependence on altitude

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Body: Black carbon (BC) aerosols absorb solar radiation, and are thus thought to cause climate warming. However, here we show that black carbon at high altitudes may decrease surface air temperature despite decreasing planetary albedo.

In this study, we investigate the dependence of climate response on the altitude of BC aerosols. This study was carried out using NCAR Community atmosphere model, CAM3.1 by performing seven idealized simulations, each of which added 1 Mt (10^6 ton) of BC aerosol uniformly around the globe to a different horizontal layer in the atmosphere, ranging from the surface layer to the stratosphere.

It was found that adding 1 Mt of BC to the near-surface bottom layer of the model atmosphere leads to a globally averaged surface air temperature increase of 2.22 +- 0.04 K (relative to the control simulation). As the altitude at which BC was added to the atmosphere increased, less surface warming was observed; BC added in the upper troposphere and stratosphere lead to statistically significant cooling of surface air.

BC in the surface layer increased precipitation by 3.88 +- 0.05%, while BC in all higher layers led to decreased precipitation.

The altitude dependence of BC forcing on surface air temperature and precipitation was the net result of the following combined factors: 1) Proximity to the surface: BC was observed to warm the layer at which it was added. Thus, there was more direct heating of the surface layer from BC located near the surface. 2) Changes in stability of the atmosphere: increased BC concentrations in the bottom layer of the atmosphere increased the vertical lapse rate and thus decreased atmospheric stability with subsequent increases in precipitation and reductions in low cloud cover (warming influence). Increased BC in higher layers increased stability, with subsequent decreases in precipitation and increases in low cloud cover (cooling influence). 3) The aerosol semi-direct effect: for increased BC concentrations near the surface, low cloud cover decreased (warming influence), while for BC in the upper troposphere and stratosphere, high cloud cover decreased (cooling influence). 4) The relative quantity of greenhouse gases above the BC: for BC at low altitudes, its emitted longwave radiation was largely absorbed by the greenhouse gases above, whereas longwave radiation emitted from BC at high altitudes was more likely to escape directly to space. Note that indirect aerosol effects on clouds were not modeled, and thus results here consider only direct and semi-direct aerosol effects.

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Effect of aerosol radiative forcing on the seasonal variation of snow over the northern hemisphere *M. Kim;* ¹; *W. K. Lau;* ²; *W. Lee;* ³; *K. Kim;* ⁴;

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Body: In this study, the effect of aerosol radiative forcing on the seasonal variation of snow is studied based on GCM simulation with prescribed aerosols. Numerical experiments are conducted using NASA fvGCM with McRAS. Monthly mean distribution of five aerosol species (black carbon, organic carbon, dust, sulfate, and sea salt) is obtained from GOCART model. In the control run, all five aerosol species are included. For sensitivity test, we carry out an experiment without any aerosol radiative forcing and three additional runs, which are identical to the control run, except for exclusion of black carbon, of dust, and of sulfate, to show the effect of different types of aerosols.

The results show that atmospheric warming by absorbing aerosols, dust and black carbon, initiate the elevated heat pump (EHP) and subsequently warm the atmosphere and land surface, especially over Tibetan Plateau (TP). As a results snow over TP reduced greatly in April and May, and

the reduction of snow cover decrease surface albedo. Surface energy balance analysis shows that the surface warming due to absorbing aerosol cause early snow melting and further increase surface-atmosphere warming through snow/ice albedo feedback. The similar relations between aerosol radiative forcing and snow melt are also found over other higher latitude region in the Northern Hemisphere. The intensity and duration of earlier snow melt by black carbon aerosol is more significant than that of dust aerosol over the higher latitude in the Northern Hemisphere while over the Tibetan Plateau both black carbon and dust aerosol are important in driving earlier snow melt.

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Long-term trends of aerosol carbon and nitrogen, their stable isotopic compositions, and water-soluble organic carbon in the western North Pacific

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Body: The western North Pacific is an outflow region of Asian dusts and pollutants. A rapid industrial development in China and East Asian countries for last two decades may have seriously altered the air quality of the North Pacific. However, long-term changes of atmospheric composition have not been studied in this region. To better understand long-term atmospheric changes in the western North Pacific, we collected marine aerosols on weekly basis in 2001-2009 at a remote island, Chichijima (27°04'E; 142°13'N) using high volume air sampler and pre-combusted quartz filter. The island is located in the boundary of westerly (winter and spring) and trade wind (summer and autumn) regimes. Here, the aerosol samples were analyzed for total carbon (TC) and nitrogen (TN) and their stable isotopic compositions as well as water-soluble organic carbon (WSOC). Concentrations of aerosol TC and TN were in a range of 0.21-4.58 (average 1.07) ug/m3 and 0.01-3.43 (av. 0.29) ng/m3. These values are ca. 20 times lower than the concentrations in urban Tokyo. C/N weight ratios ranged from 0.50 to 41 (av. 6.6) with summer maxima. Both TC and TN showed a gradual increase with winter/spring maxima probably due to the enhanced anthropogenic activities such as fossil fuel combustion and fertilizer usage in East Asia. On the other hand, stable carbon isotopic ratios (d13C) of TC ranged from -27.3 to -15.1%; however, no systematic trend was detected. Nitrogen isotopic ratios of TN (d15N) ranged from +1.56 to +25.2% with lower values in winter and higher values in summer. Interestingly, d15N values showed a long-term trend of increase by 4% from 2001 to 2009. This may be caused by an increased emission of nitrogen species (NH3, NOx) from biomass burning sources in East Asia and other regions. WSOC (0.05-2.46 ug/m3, av. 0.40 ug/m3) also showed a long-term increase probably due to the increased emissions of organic aerosols and their precursors and subsequent oxidations in the atmosphere during long-range transport. WSOC was found to comprise 10-74% (av. 38%) of aerosol TC.

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Estimating bulk optical properties of aerosols over the western North Pacific by

using MODIS and CERES measurements

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Body: Over the western North Pacific, a large amount of land aerosols from Asian-Pacific countries is transported by the prevailing westerlies. This transport makes the radiative characteristics of these aerosols diverse, particularly when one compares those characteristics over the coastal sea with those over the open sea. In this paper we discuss a method that uses satellite data to obtain the single scattering albedo (ω) and asymmetry factor (g) of atmospheric aerosols for two large-scale subdivisions the coastal sea (within 250 km from the coast) and the open sea (the remaining area) over the western North Pacific (110°E-180°, 20°N-50°N). Our estimation method uses satellite measurements, obtained over a six-year period (2000-2005), of aerosol optical depth (AOD) and shortwave fluxes at both the surface and the top of the atmosphere (TOA); the measurements are obtained using the Moderate Resolution Imaging Spectroradiometer (MODIS) and the Clouds and the Earth's Radiant Energy System (CERES). For the two subdivisions, the estimated annual means of (ω, g) at 630 nm are significantly different: (0.94, 0.65) over the coastal sea and (0.97, 0.70) over the open sea. From a quantitative viewpoint, this result indicates that in comparison with aerosols over the open sea, those over the coastal sea show greater absorption and lesser forward scattering of solar radiation. The estimated optical properties are responsible for the aerosol surface cooling observed by MODIS and CERES, which is approximately 138 and 108 W m-2 per AOD over the coastal sea and open sea, respectively.

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Spectral aerosol light absorption measurement at urban, suburban, and remote sites in Korea

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Body: Spectral aerosol light absorption (babs) was measured by a filter-based seven-wavelength Aethalometer (370, 470, 520, 590, 660, 880, and 950nm) during the winter and spring of 2008 and 2009 at urban (Seoul), suburban (Gwangju), and remote (Deokjeok island) sites in Korea. Aerosol scattering (bscat) was also measured using a Nephelometer. The interference of filter-based absorption measurement by aerosol loading was investigated and corrected for better measurement of babs. Slightly increased loading correction factors were resulted at 880 and 950nm in the urban and remote areas and 370nm in the suburban area, respectively. It was found that original Aethalometer underestimated babs at 370nm by 21.6, 18.9, and 19.5% at the urban, suburban, and remote sites, respectively. From the corrected babs and bscat, aerosol single scattering albedo at 520nm was determined to be 0.82±0.05, 0.86±0.04, and 0.89±0.03 at the urban, suburban, and remote sites, respectively. Atmospheric condition at the remote area was categorized based on satellite based aerosol optical thickness (AOT) and air mass backtrajectory analyses. Ångström exponent of babs in the UV-visible region (370-520nm) (α_UV-vis) generally showed decreasing trend as babs increased. α_UV-vis at the remote area was lower than those obtained at the urban and suburban areas. The measurement site in remote area was frequently influenced by long-range transported pollution from Asian continent. Lower α_UV-vis values, <1.5 were observed at the remote area during the long-range transport pollution (LTP) period.

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Evaluation of a newly developed below-cloud scavenging scheme of regional aerosol simulations: its implication for aerosol budget over East Asia

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Body: Wet scavenging is the most important process for the aerosol removal. It is divided into in-cloud and belowcloud scavenging processes. Although the below-cloud scavenging is less efficient than the in-cloud scavenging, it is important for the removal of coarse and very fine particles from the polluted boundary layer. Important factors determining the efficiency of below-cloud scavenging process by rain droplets are collision efficiency, terminal velocity of a raindrop, raindrop size distributions, and particle size distributions. Complex 3-D models of atmospheric aerosols, however, in general neglect those factors and use a simple parameterization for the below-cloud scavenging in the form of either constant or first-order equations. For example, a Model Inter-Comparison Study for Asia (MICS-Asia) II showed a large range of simulated wet deposition fluxes depending on wet deposition parameterizations of participating models despite of the use of similar meteorological fields. A mechanistic scheme incorporating important factors above to be easily implemented in existing 3-D models is necessary for a better below-cloud scavenging simulation. In this study we test and evaluate a new scheme of the below-cloud scavenging process with Community Multiscale Air Quality (CMAQ) model, accounting for the relationship between the raindrop size distribution and rain intensity along with realistic consideration of other important factors. We conducted regional simulations of CMAQ with the new scheme in East Asia and compared results with other models in MICS-Asia II. We also evaluate the improved CMAQ model by comparing with observations from the Transport and Chemical Evolution over the Pacific (TRACE-P) and the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) aircraft campaigns in spring 2001. Improved wet deposition simulations of aerosols result in a better understanding on aerosol budget and its climatic implication over East Asia.

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Physical and hygroscopic properties and CCN characteristics of the aerosols measured at an island in the northwestern tip of South Korea

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Body: Atmospheric aerosols have drawn great attention in recent decades for their direct and indirect effects on climate. Particularly, Asian regions of rapid industrialization are well-known for transporting enormous air pollution aerosols throughout the Pacific. As an effort to monitor the physical and hygroscopic properties of Asian continental anthropogenic aerosols, we measured total aerosol (CN) concentration, hygroscopicity and cloud condensation nuclei (CCN) concentration for about a month (August) in 2009 at a hilly site in Baekryeong Island, located at the northwestern tip of South Korea (N 37°52′, E 124°53′). Since local anthropogenic disturbance is minimal, the site is ideal for monitoring Asian continental outflow when the prevailing wind is from the west. In this study CCN characterization was also attempted. Critical supersaturation (SC) of ambient aerosols of selected diameters of 75nm and 100nm were measured with a DMA-CCN Counter setting. SC, hygroscopicity, and CCN/CN ratios are combined to estimate the CCN capability of these aerosols. Preliminarily the daily mean CN concentration varied from 2768 to 8626 cm-3 while CCN concentration at 1% supersaturation varied from 1571 to 7492 cm-3. The average of CCN/CN ratio was approximately 0.8. The 75 nm ambient particles had the SC range from 0.20 to 0.50 % whereas the 100 nm ambient particles had the SC range from 0.13 to 0.32. More detailed analysis will be shown at the conference.

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Physicochemical evolution of dust particles observed in March 2007, Seoul, Korea

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- 3. National Institute of Environmental Research, Seoul, Korea, Republic of.

Body: From 26 March to 2 April 2007, two dust episodes were reported in Korea. One is 27-28 March and the other is 31 March-1 April which could be classified into weak and heavy Asian dust events, respectively, based on their PM10 intensities. To examine chemical and physical processes controlling compositions of Asian dust particles, PM10 aerosol samples were collected using PM10 cyclone with teflon filters on the roof of the sixth floor in the Asan science building located in Korea University. PM10 samples were analyzed for water soluble ions including 5 cation species and 3 anion species using ion chromatography. During the heavy Asian dust period, the mass concentration of PM10 reached the maximum value of 637.1 μg/m\$^{3}\$ on 1 April 9-15h and the second maximum value of 412.0μg/m\$^{3}\$ on 31 March 21h-1 April 9h, which can be classified into the relatively strong and the strongest dust storms, respectively. During the strongest dust storm, Ca\$^{2+}\$ and Mg\$^{2+}\$ showed a drastic increase of their levels up to 6.01 μg/m\$^{3}\$ and 0.81 μg/m\$^{3}\$, respectively, suggesting their role as soil dust tracers. In contrast, concentrations of Cl\$^{-}\$, K\$^{+}\$ and Na\$^{+}\$ were the highest, 4.59 μg/m\$^{3}\$, 3.20 μg/m\$^{3}\$ and 2.22 μg/m\$^{3}\$ respectively, during the relatively strong dust storm. During the weak Asian dust period, PM10 mass concentration was 165.5 μg/m\$^{3}\$ and elevated levels of SO\$_{4}\$\$^{2-}\$, NO\$_{3}\$\$^{-}\$ and NH\$_{4}\$\$^{+}\$ in PM10 were observed indicating the significant role of dust particles acting as an effective sink for acidic pollution gases. Detailed discussion including analyses of SEM/EDX, XRF and XRD will be presented in the meeting.

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Comparison of PM 2.5 carbonaceous aerosols in the northeast Asian regions $\underline{\textit{M. Chang;}}^1$; $\textit{M. Lee;}^1$; $\textit{S. Lim;}^1$; $\textit{J. Kim;}^1$;

1. Korea Uni., Seoul, Korea, Republic of.

Body: Aerosols are composed of various chemical species such as ions, organic carbon (OC) and elemental carbon (EC). Among these, organic carbon and elemental carbon were analyzed as 8 fractions (OC1,OC2,OC3,OC4,EC1,EC2,EC3) by TOT/TOR (Thermal Optical transmission/Reflectance method), using IMPROVED protocol method. This study had been performed at Seoul metropolis, Gosan supersite in Jeju and leodo in the East China Sea from August 2007 to June 2008. Mean concentrations of OC and EC in PM_{2.5} are 9.757μg/m³ . 4.648μg/m³ (Seoul), 3.95μg/m³, 1.69μg/m³ (Jeju) and 2.43μg/m³, 1.01μg/m³ (Ieodo). OC to EC ratios were 2.39, 2.63 and 2.82 at Seoul, Jeju and leodo in respectively. Organic matter and elemental carbon are composed 47% and 16% (Seoul), 23.3% and 10.0% (Jeju) and 26% and 5% (leodo). Three stations showed different behaviors of OC and EC. Detailed results for Characteristics of carbonaceous compositions will be discussed in the meeting.

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The relationship between the Asian dust and Arctic Oscillation: An observational investigation

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Body: The Arctic Oscillation (AO) represents the leading empirical orthogonal function of winter sea level pressure (SLP) fields, which resembles North Atlantic Oscillation, but has a more zonally symmetric structure (Thompson and Wallace, 1998; Wallace, 2000; Wu and Wang, 2002). This primary mode of the internal dynamics in the atmosphere predominates the Extratropical Northern Hemisphere circulation from surface to the lower stratosphere showing an equivalent barotropic structure during cold season(November-April) (Thompson and Wallace, 2000). Also, the Asian dust storms show strong interannual variation (Sun et al., 2001; Yoshino, 2002; Zhao et al., 2006; Hara et al., 2006), which are suggested to be associated with climate indices. Therefore, the purpose of this study is to identify the possible connection for the AO and the Asian dust emission during periods from late winter to spring, based on the observational investigations.

In order to examine these complex associations closely, the Aerosol Index (AI) from Total Ozone Mapping Spectrometer (TOMS) are used starting from November 1978 to December 1999, except for data pause from May 1993 to August 1996. SLP and geopotential height at 500hPa (Z500) monthly fields are derived from the European Centre for Medium-Range Weather Forecasts (ECMWF) reanalyses (1958-2001). Also, source regions of Asian dust are mainly focused on three source areas, including the Taklimakan Desert (R1), Badain Jaran Desert (R2), and Mongolia (R3) in this study. In composite study during different phases of AO, the AO exhibits a "negative phase" with relatively high pressure over the polar region and low pressure at mid-latitudes (about 45 degrees North). In strong "negative phase" of AO, decrease of AO is found to be associated with decreases of local tropospheric temperature, U-wind, and geopotential height and strong north-south gradients over the source regions of Asian dust. Therefore, we also notice that a negative index phase in the spring AO results in the strengthened East Asian Trough at 500hPa and a significant pressure decrease in the lower atmosphere, which is favorable for producing the stronger Asian dust emission.

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Solar Radiation and Cloud Change in China Calculated in a 20th Century Simulation with a Climate Model

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Body: Significant long-term trends in various climate elements in China have been observed in recent decades. The elements include surface temperature, precipitation, evaporation, surface solar radiation, cloud cover, etc. It is of particular interest that coexistence of decreasing trends both in solar radiation and cloud cover have been observed over most of China, since a decreasing trend of solar radiation would be expected to be accompanied by an increasing trend of cloud cover that reduces the radiation from the sun. Here, we investigate the cause of this confusing fact with the aid of a 20th century simulation performed with the MIROC climate model.

Simulated trend in the surface solar radiation shows the decreasing trend over entire China, although the trend averaged over China is about -1.0 W/m2 which is smaller than observed trend. While the simulated trend in cloud cover averaged over China is indiscernible as opposed to the observation, a small but significant decreasing trend similar to the observation could be simulated in the southern part of China. In the southern part of China, our model could capture the coexistence of decreasing trends both in solar radiation and cloud cover, which could also be simulated an experiment forced only anthropogenic climate forcings (i.e. increases in greenhouse gases and aerosols). As for the decrease in the surface solar radiation simulated in the model, the dominant cause is the increase in aerosols. Simplified analysis revealed the relative importance of direct and indirect effect of aerosols on the decrease in the surface solar radiation in the model; both effects have comparable contributions in some regions. The increase in greenhouse gasses has some effect on the decrease in cloud cover in the southern part of China in the model.

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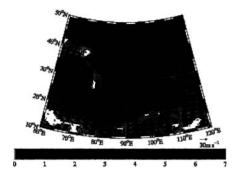
High Concentration of Surface Ozone Observed along the Khumbu Valley Nepal April 2007

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Body: Increasing air pollution in Southeast Asia has the potential for dramatic impacts on the population and climate in relatively pristine regions such as the Himalaya. Recent measurements near Mount Everest indicate the presence of elevated levels of ozone at elevations from 5000m to 9000m that are the result of both the long-range transport of tropospheric pollutants from Southeast Asia as well as the descent of ozone-rich stratospheric air. Here we report on the first surface ozone concentration transect in the Mount Everest region. The data collected at elevations from 2900m to 5200m indicate an increase in concentration with height as well as 8-hour average exposures in excess of 140ppb. Satellite data and meteorological diagnostics suggest a stratospheric source for the high levels observed. The majority of values observed exceed guidelines for human exposure and therefore are of a magnitude to suggest that they are of physiological relevance.

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Potential vorticity (PVU-shading and contours) and horizontal wind (vectors-m/s) from the ERA-I Reanalysis on the 330K potential temperature surface at 06 UTC (approximately noon local time) on April 12 2007 showing the tropopause fold that resulted in the high surface ozone concentration observed along the Khumbu Valley near Mount Everest. The 1 PVU contour is indicated by the thin black line. The Tibetan Plateau is indicated as is the location of Mount Everest by the '+'.

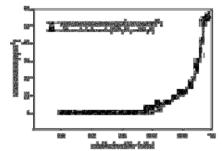
Effect of the Presence of Crustal Elements (Ca2+, K+, and Mg2+) in Estimating Water Content in PM2.5 H. Lee; ¹; Y. Kim; ¹;

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Body: Water content in the ambient aerosols is an important characteristic since it affect various properties of the ambient aerosols such as aerosol hygroscopicity. Measurement of aerosol water content is difficult and usually estimated by using either empirical formula or gas/particle equilibrium model. It has been thought that major inorganic ions (Na+, SO42-, NH4+, NO3-, and Cl-) be the dominant figures in determining aerosol water content. A gas/particle equilibrium model, SCAPE2 was used to estimate the aerosol water content based on the PM2.5 and related gaseous species measurement data at Seoul (Apr. 2001 - Feb. 2002, Nov. 2004 – Jul. 2005) and Gosan (Mar. 1996 - Jan. 2007), the capital of Korea and a background area in Korea, respectively. For each site, seasonal characteristics were examined. Figure 1 shows estimated water content increasing with relative humidity at Gosan in spring. It was found that the increase of estimated aerosol water content curves to relative humidity with crustal elements (Ca2+, K+, and Mg2+) and without them show different behavior at the deliquescence points as shown. Thus, without crustal elements, the aerosol water content estimation might introduce significant errors in aerosol modeling such as 3-D chemical transport modeling.

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Estimated water content increasing with relative humidity of Gosan, Korea in spring

The effects of aerosols on Thunderstorms and Lightning over the Pearl River Delta, China

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- 1. Texas A&M University, College Station, TX, United States.
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Body: The effects of aerosols on precipitation and cloud processes have been investigated using a cloud resolving Weather Research and Forecasting (CR-WRF) with a two-moment bulk microphysical scheme in a cold frontal system developing on 28 Match 2009 in Guangdong, China. The model uses two-domain configuration with a particular focus on the lightning activity occurring the area in the Pearl River Delta (PRD) urban group. The simulated evolution of composite radar reflectivity and accumulated precipitation are qualitatively consistent with the measurements, including automatic gauge station data and C-band weather radar. Sensitivity experiments have been performed for two aerosol scenarios representing 'polluted' and 'clean' conditions. In the polluted case, the aerosol number concentration is initialized with a higher value and the aerosol emission is also considered in the PRD urban area. The results of sensitive simulations suggest that the precipitation is enhanced by as much as 15% in the high aerosol condition. Both the convection zone and the core updraft velocity exhibit similar enhancement as precipitation. More efficient mixed phase processes and intensified convection are speculated to be the significant factors in creating lightning enhancement in PRD urban area, which has been observed by local lightning detection network.

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Insights to the topographic and aerosol roles in the heavy rainfall at the Western Ghats and the scarce rainfall farther east at the rain shadow as observed in CAIPEEX

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- 2. Institute of Earth Sciences, Hebrew University of Jerusalem, Jerusalem, India.

Body: Rainfall processes in the clouds occur in variety of pathways and atmospheric aerosols have been found to play an important role in cloud microphysics. It is important to understand the pathways through which aerosols modify clouds and lead to precipitation. Cloud Aerosol Interaction and Precipitation Enhancement Experiment (CAIPEEX), is an Indian National program conducted by the Indian Institute of Tropical Meteorology (IITM), Pune, India is designed to address the above issues. Under this program aircraft observations of cloud microphysical parameters and aerosol properties are being undertaken over different parts of India. The interior part of the peninsular India is rain shadow region. The seasonal monsoon rainfall there is lower compared to all India mean monsoon rainfall. The rainfall variability is larger and the region is drought prone. The CAIPEEX Phase-I observations provided a suitable opportunity to study the variability in the aerosol and cloud microphysical properties over the rainy regions (Western Ghats) and rain shadow regions to the east. One of the significant results showed that the warm rain initiation starts at early stages (lower cloud depths) over the coastal areas and becomes even lower at the crest of the western slopes, probably due to intense washout of the aerosols by the rain. However, the clouds lose their microphysically maritime character farther east over the rain shadow regions, where the clouds have to grow to higher depths to initiate the process. The aerosol and CCN concentrations support the hypothesis that the shallow convective clouds (i.e., with tops < 6 km) lose their ability to rain over the rain shadow due to increased aerosols concentrations, which are probably contributed to by air pollution

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The interplay between small and large CCN: Impacts on convective cloud microstructure as observed in CAIPEEX.

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Body: Aircraft observations during Cloud Aerosol Interactions and Precipitation Enhancement Experiment (CAIPEEX) conducted over India in 2009 are utilized here. Interplay between the small and large aerosol particles and cloud drop size distributions are investigated with hazy and relatively clean environment. We found that the haze in boundary layer has direct effect on the formation of cloud droplets which could result broad cloud droplet spectra. It may be emphasized that the haze contains giant cloud condensation nuclei (CCN) and could enhance the process of warm rain. However, in polluted environment with small CCN we found narrow cloud droplet spectra at the cloud base. Quantitative relations on the interplay between giant and small nuclei on cloud droplet spectra and formation of warm rain process have been documented and will also be shown.

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Can Aerosol Offset Urban Heat Island Effect?

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Body: The Urban Heat Island effect (UHI) refers to urban skin or air temperature exceeding the temperatures in surrounding non-urban regions. In a warming climate, the UHI may intensify extreme heat waves and consequently cause significant health and energy problems. Aerosols reduce surface insolation via the direct effect, namely, scattering and absorbing sunlight in the atmosphere. Combining the National Aeronautics and Space Administration (NASA) AERONET (AErosol RObotic NETwork) observations over large cities together with Weather Research and Forecasting Model (WRF) simulations, we find that the aerosol direct reduction of surface insolation range from 40-100 Wm-2, depending on seasonality and aerosol loads. As a result, surface skin temperature can be reduced by 1-2C while 2-m surface air temperature by 0.5-1C. This study suggests that the aerosol direct effect is a competing mechanism for the urban heat island effect (UHI). More importantly, both aerosol and urban land cover effects must be adequately represented in meteorological and climate modeling systems in order to properly characterize urban surface energy budgets and UHI.

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Size distributions of secondary and primary aerosols in Asia: A 3-D modeling

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Body: Asian aerosols have received increasing attention because of their potential health and climate effects and the rapid increasing of Asian emissions associated with accelerating economic expansion. Aerosol particles appear in the atmosphere due to either in-situ nucleation (i.e, secondary particles) or direct emissions (i.e., primary particles), and their environmental impacts depend strongly on their concentrations, sizes, compositions, and mixing states. A sizeresolved (sectional) particle microphysics model with a number of computationally efficient schemes has been incorporated into a global chemistry transport model (GEOS-Chem) to simulate the number size distributions of secondary and primary particles in the troposphere (Yu and Luo, Atmos. Chem. Phys. Discuss., 9, 10597-10645, 2009). The growth of nucleated particles through the condensation of sulfuric acid vapor and equilibrium uptake of nitrate, ammonium, and secondary organic aerosol is explicitly simulated, along with the coating of primary particles (dust, black carbon, organic carbon, and sea salt) by volatile components via condensation and coagulation with secondary particles. Here we look into the spatiotemporal variations of the size distributions of secondary and primary aerosols in Asia. The annual mean number concentration of the accumulation mode particles (dry diameter > ~ 100 nm) in the lower troposphere over Asia (especially China) is very high and is dominated (~70-90%) by carbonaceous primary particles (with coated condensable species). Coagulation and condensation turn the primary particles into mixed particles and on average increase the dry sizes of primary particles by a factor of ~ 2-2.5. Despite of high condensation sink, sulfuric acid vapor concentration in many parts of Asian low troposphere is very high (annual mean values above 1E7/cm3) and significant new particle formation still occurs. Secondary particles generally dominate the particles small than 100 nm and the equilibrium uptake of nitrate, ammonium, and secondary organic aerosol contributes significantly to the growth of these particles. The vertical profiles of particle number size distributions at representative locations show significant spatial variations (both horizontally and vertically). Our simulations also indicate substantial seasonal variations of particle size distributions.

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Dust Aerosol Optical Properties Retrieval and Radiative Forcing over Northwestern China during 2008 China-US Joint

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Body: The Atmosphere Radiation Measurements (ARM) Program's Ancillary Facility (AAF/SMART-COMMIT) was deployed to Zhangye (39.082° N, 100.276° E), which is located in a semi-desert area of Northwest China, during the period of late April to mid June in 2008. We selected 11 cases to retrieve dust aerosol optical depth (AOD), Angstrom exponent, size distribution, single-scattering albedo (SSA) and asymmetry parameter (ASY) from Multi-filter Rotating Shadowband Radiometer (MFRSR) measurements. These cases are dominated by large particles with Angstrom exponent values ranging from 0.34 to 0.93. The values of AOD at 0.67 µm range from 0.074 to 0.249. The mean SSA value increases with wavelength from 0.76±0.02 at 0.415 µm to 0.86±0.01 at 0.867 µm, while the mean ASY value decreases from 0.74±0.04 to 0.70±0.02. Before estimating dust aerosol direct radiative forcing, a radiative closure experiment was performed to verify that the retrieved aerosol optical properties and other input parameters to the radiative transfer model appropriately represent atmospheric conditions. The daytime-averaged differences between model simulations and ground observations are -8.5, -2.9, and -2.1 Wm-2 for the total, diffuse, and direct normal fluxes, respectively. The mean difference in the instantaneous reflected solar fluxes at the top of atmosphere (TOA) between the model and CERES observations is 8.0 Wm-2. The solar aerosol direct radiative forcing (ARF), averaged over a 24-hour period, at the surface is-22.4±8.9 Wm-2, while the TOA ARF is small and has an average value of only 0.52±1.69 Wm-2. The daily-average surface aerosol radiative forcing efficiency (ARFE) at 0.5 μm is -95.1±10.3 Wm-2T-1. Our results illustrate that the primary role of dust aerosol is to alter the distribution of solar radiation within the climate system, rather than reflecting solar energy to space. We assess the satellite aerosol optical depth products from MISR and MODIS observations by comparing them with our ground-based retrievals. Reasonable agreements with the ground-based observations are found for the MISR product and MODIS Deep Blue product.

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Implementation of different dust emission schemes into WRF/Chem: An evaluation of dust emisson parameterizations *J. Kang;* ¹; *S. Yoon;* ¹; *Y. Shao;* ²; *E. Jung;* ²;

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- 2. University of Cologne, Cologne, Germany.

Body: Dust modeling plays a key role in understanding dust phenomena. A comprehension of the performance and uncertainty of dust models has become essential for developing dust models. Current models can produce dust phenomena reasonably, but the amounts of simulated dust concentration are much different from each and every model. It is required to estimate how the different parameterizations in several dust emission schemes affect the Asian dust simulations.

Five dust emission schemes (Marticorena and Bergametti;1995, Lu and Shao;1999, Alfaro and Gomes;2001, Shao;2001, Shao;2004) were implemented into WRF/Chem (Weather Research and Forecasting with Chemistry) to compare different dust emission parameterizations and identify the impact of input parameters on dust emission amounts. New surface static data such as vegetation cover and soil texture class were also ingested to make better boundary conditions which can strongly affect dust emission amounts. To avoid the occurrence of dust emission in non-dust source area, calculation of dust emission was confined within the region known as dust source area. Sensitivity tests were conducted focusing on the characteristics of dust emission schemes and the relative importance of input parameters of each scheme. Several case studies for Asian dust events were carried out and the simulation results were compared with observation data.

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Evidence of gravity waves in the PBL above New Delhi, during the total Eclipse of August 2009.

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- 2. Leosphere, Paris, France.

Body: A total eclipse occurred on july the 22th of 2009 over India, starting from Bombay and crossing the country toward the east. During this event, an EZ Aerosol LIDAR™ has been monitoring at the National Physics Lab, making high resolution and continuous observations of the atmosphere at 355nm. Polarisation measurements were also taken.

The eclipse in Indian continent started about 4;30 early in the morning. In Delhi It was between 6:00 AM to 7:00AM on 22-7-09. In this time interval the signature of wave pattern is observed.

These lidar results show some experimental evidence which support the hypothesis that the movement of moon's shadow sweeping the ozone layer at supersonic speed during a solar eclipse, creates gravity waves in the atmosphere.

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Unusual PM episode caused by yellow sand over Taiwan on April 25, 2009

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- 4. Environmental Research and Information Center, Chang Jung Christian University, Tainan, Taiwan.

Body: This study analyzes the unusual particulate matter episode caused by yellow sand (YS), which affected Taiwan on April 25, 2009. During this YS event, the PM10 hourly concentrations higher than 1000 µg m-3, about 20 times higher than local normal concentration, were observed at several sites over northern Taiwan. Moreover, of Taiwan's 76 air quality monitoring stations, 69 stations recorded daily average concentrations of PM10 which exceeded Taiwan's air quality standard of 125 µg m-3. Comparing the time periods when such high PM10 concentrations occurred in different areas in Taiwan, the transport phenomena of PM10 could be shown. The maximum concentrations of the central and southern PM10 occurred about 6 hours and 15 hours later, respectively, than those of the northern region. The maximum hourly values of PM2.5 and PM10 particles at 175 and 814 µg m-3, respectively, were recorded at the Taipei aerosol supersite (TAS) located in northern Taiwan. The ratios of PM10 to PM2.5 decreased from 0.6 to 0.2 during YS period as shown in Fig.1. This YS event not only greatly increased the PM2.5 and PM10 concentrations, but enhanced the sulfate fraction of PM2.5. The PM2.5 sulfate 22 µg m-3 at TAS site during YS period was even higher than that derived from local accumulation. Furthermore, the particles' number size spectra at TAS site showed a decrease in the amount of particles with a diameter of less than 200 nm, indicating that the rise of the mass concentration of PM2.5 might increase along with the increase of PM2.5 particles with diameters larger than 200 nm. Several studies have indicated that the presence of yellow sand may be associated with increased health risks. The results of this study can provide the basis for further investigation into its effects on human health.

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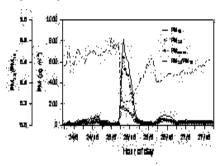


Figure 1. Hourly PM2.5, PM10 and PM2.5 to PM10 ratios at TAS during yellow sand period.

Recent Increase in Black Carbon Concentrations from a Mt. Everest Ice Core Spanning 1860-2000 AD S. Kaspari; ^{1, 2}; M. Schwikowski; ²; M. Gysel; ³; P. A. Mayewski; ^{4, 6}; S. Kang; ^{5, 6}; S. Hou; ^{6, 7};

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Body: Black carbon produced by the incomplete combustion of biomass, coal and diesel fuels can significantly contribute to climate change by altering the Earth's radiative balance. Black carbon in the atmosphere absorbs light and causes atmospheric heating, whereas black carbon deposited on snow and ice can significantly reduce the surface albedo, resulting in rapid melting of snow and ice. Historical records of black carbon concentration and distribution in the atmosphere are needed to determine the role of black carbon in climate change, however most studies have relied on estimated inventories based on wood and/or fossil fuel consumption data. Reconstructing black carbon concentrations in Asia is particularly important because this region has some of the largest black carbon sources globally, which negatively impact climate, water resources, agriculture and human health. We analyzed a Mt. Everest ice core for black carbon using a single particle soot photometer (SP2). The high-resolution black carbon data demonstrates strong seasonality, with peak concentrations during the winter-spring, and low concentrations during the summer monsoon season. Black carbon concentrations from 1975-2000 relative to 1860-1975 have increased approximately threefold, and the timing of this increase is consistent with black carbon emission inventory data from South Asia. It is notable that there is no increasing trend in iron (used as a proxy for dust) since 1860. This is significant because it suggests that if the recent retreat of glaciers in the region is due, at least in part, to the effect of impurities on snow albedo, the reduced albedo is due to changes in black carbon emissions, not dust.

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How do the optical properties of Asian aerosols change when they cross the Pacific?

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Body: Primary and secondary aerosols from Asia may have important climate implications. These aerosols are emitted locally, but can then be lofted into the free troposphere and advected across the Pacific. In this analysis we used observations from the Mount Bachelor Observatory (MBO) in conjunction with satellite data to identify the dominant aerosol types in specific Asian plumes that crossed the Pacific. In situ data from MBO is used to understand the observed changes in radiative properties.

A suite of gas phase and aerosol measurements were made during spring 2008 and spring 2009 at MBO (2763 masl), located in central Oregon. Here we focus on observations of dry sub– μ m aerosol scattering (σ_{sp}) and absorption (σ_{ap}), made with an integrating nephelometer and a particle soot absorption photometer (PSAP). Using a combination of backward trajectory calculations and satellite observations, we identified 7 well defined plumes of Asian origin. These plumes included the highest σ_{sp} (34.8 Mm $^{-1}$ hourly average) and σ_{ap} (4.8 Mm $^{-1}$ hourly average) observed at MBO over the 2008 and 2009 spring campaigns. Of interest in this analysis is 1) whether the intensive optical properties differ between these 7 Asian events, 2) whether these differences can be linked to differences in composition, and 3) whether the intensive optical properties differ from those observed closer to the Asian source region. Preliminary results show that the plumes clustered in terms of their optical properties; plumes hypothesized to contain a large fraction of mineral dust were the most distinct. We also observed larger variability in the average scattering Ångstrom exponent of the plumes and a higher average single scatter albedo than observations closer to the Asian coast. This work will be extended to compare observations at MBO with the most recent observations from Asia as they become available.

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New Discoveries and Insights from Comparisons between Sahara and Asian Dust Using Satellites and a Coupled Climate/Microphysical Model

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Body: This study compares Sahara dust and Asian dust using CALIPSO and other satellite data along with a coupled climate/microphysical sectional model. Our investigations focus on dust lifting, transport mechanisms, physical properties, and optical properties. Satellite retrievals provide excellent spatial and temporal data sets that can be used to explore many of these properties. A three-dimensional coupled microphysical/climate model based on the University of Colorado/NASA Community Aerosol and Radiation Model for Atmospheres (CARMA2.3) and the NCAR Community Atmosphere Model (CAM3) is also used in this study. We use 16 size bins for mineral dust ranging from 0.1 to 10 micron radius. The simulated dust lifting and vertical distribution of Sahara and Asian dust is constrained by CALIPSO (lidar) retrievals in 2006 and 2007. Sahara dust lifting occurs all year long, but Asian dust is lifted mostly in spring due to the low winds at other times as well as it is suppressed by vegetation. Asian dust is also highly variable between years. The Sahara dust layers descend over several days in the cases studied from about 7km to the surface. Asian dust often has multiple layers (two layers in the cases studied) during transport. One layer stays well above boundary layer during transport and shows little descent, while the other, lower, layer descends with time. Sahara dust is generally embedded in tropical easterlies, and interacts with deep convection. Asian dust is primarily in the mid-latitude westerly jet, and is often associated with dynamical features involving widespread precipitation. This study is a step towards a global understanding of dust and its properties.

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REMOTE SENSING MEASUREMENTS OF AEROSOL OPTICAL THICKNESS AND CORRELATION WITH IN-SITU AIR QUALITY PARAMETERS DURING A SMOKE HAZE EPISODE IN SOUTHEAST ASIA

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Body: Transboundary smoke haze due to biomass burning is a major environmental problem in Southeast Asia which has not only affected air quality in the source region, but also in the surrounding countries. Air quality monitoring stations and meteorological stations can provide valuable information on the concentrations of criteria pollutants such as sulphur dioxide, nitrogen oxide, carbon monoxide, ozone and particulate mass (PM10) as well as health advisory to the general public during the haze episodes. Characteristics of aerosol particles in the smoke haze such as the aerosol optical thickness (AOT), aerosol size distribution and Angstrom exponent are also measured or retrieved by sun-tracking photometers, such as those deployed in the world-wide AErosol RObotic NETwork (AERONET). However, due to the limited spatial coverage by the air quality monitoring stations and AERONET sites, it is difficult to study and monitor the spatial and temporal variability of the smoke haze during a biomass burning episode, especially in areas without ground-based instrumentation. As such, we combine the standard in-situ measurements of PM10 by air quality monitoring stations with the remote sensing imagery from the Moderate Resolution Imaging Spectroradiometer (MODIS) on board NASA's Terra and Aqua satellites. The columnar AOT is first derived from the MODIS images for regions where PM10 measurements are available. Empirical correlations between AOT and PM10 measurements are then established for 50 sites in both Malaysia and Singapore during the smoke haze episode in 2006. When available, vertical feature information from the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) is used to examine the validity of the correlations. Aloft transport of aerosols, which can weaken the correlations between AOT and PM10 measurements, is also identified by CALIPSO and taken into consideration for the analysis. With this integrated approach, we hope to enhance and complement current capabilities in monitoring air quality during the haze episodes in Southeast Asia.

This study was completed as a preliminary analysis of the biomass burning situation in Southeast Asia under the Seven SouthEast Asian Studies (7 SEAS) Mission. Through collaborations with scientific partners in Taiwan and various Southeast Asian countries such as Indonesia, Malaysia, Philippines, Singapore, Thailand and Vietnam, 7 SEAS is jointly initiated by NASA's Radiation Science, Tropospheric Chemistry, Air Quality and Oceanography programmes as well as the Office of Naval Research (ONR), the Office of Naval Research – Global (ONRG) and the US State Department in an effort to investigate the complex interactions between aerosols (anthropogenic or natural) and meteorological systems, especially with clouds, and their impacts on air quality in the region. 7 SEAS is a multidisciplinary regional science programme which operates with the integrative support of in-situ measurements, remote sensing and scientific modeling.

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Emission projection and uncertainty analysis of exhaust emissions from global and Asian on-road vehicles *F. Yan*; ^{1, 2}; *E. Winijkul*; ¹; *T. Bond*; ¹; *D. G. Streets*; ²;

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Body: Two of the most notable impacts from emissions of air pollutants are climate change and hemispheric or intercontinental transport. Global emission projections are identified as critical elements in understanding these large-scale impacts. Such projections are required to understand the net response of climate to combined emissions of greenhouse gases, aerosols, and other trace species in the next 30 to 50 years.

Emissions from vehicles vary with introduction of advanced technology and implementation of stringent environmental regulations. We present global emission projections of primary particulate matter emissions including the aerosol components black and organic carbon, from on-road vehicles from 2010 to 2050. These projections are based on a new model of technology that responds to socioeconomic conditions in different economic and mitigation scenarios. The model contains detail about technology stock, such as vintage, and applies exogenous data from economic scenarios to choose new technologies and retire old ones. The driving factors involved in the transitions of technology decision-making include consumption growth rates, retirement rates, timing of emission standards, and generation of superemitters. Asia is a significant contributor to global emissions and its growth rate is expected to be high, so we emphasize the trajectories in this region. Before 2030, the tradeoff between decreasing emission intensity and increasing fuel consumption results in relatively lower rates of increase of PM emissions, although emissions are still increasing. After 2030, we expect that standards will have cleaned up normal vehicles, so emission projections are highly dependent on the behavior of "superemitters."

Changes of technology and policy in the future are uncertain, and their relationship with socioeconomic variables is not well known. This lack of knowledge raises the question: What can be known about future emissions and air quality? We also present sensitivity analyses and Monte Carlo simulations to explore the impacts of these uncertainties on emission projection. We identify the most critical factors affecting our knowledge of emission pathways; these are targets for future research on the interaction between social and economic conditions and technological response

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Aerosol Direct and Semi-Direct Effects on Climate in Asia in the NASA GEOS-5 Model

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- 5. Global Modeling and Assimilation Office (Code 610.5), NASA GSFC, Greenbelt, MD, United States.

Body: Asia is a hot spot for aerosol emissions from anthropogenic sources. Dust and biomass burning aerosols also contribute to the high aerosol loadings in the region. The aerosols in this region reduce the amount of radiation that reaches the surface, and, owing to their absorptive properties in the visible, contribute to radiative heating in the atmosphere. This partitioning of energy between the surface and the atmosphere can have consequences for the regional dynamics and hydrologic cycle. Here we focus on these aerosol direct and semi-direct radiative effects on the regional climate and how feedbacks in turn impact aerosol distributions. We use the NASA Goddard Earth Observing System version 5 (GEOS-5) model, which is an integrated Earth simulation system that includes components for atmospheric and oceanic general circulation and physics, atmospheric chemistry, and oceanic biogeochemistry, land surfac modeling, and data assimilation. We have implemented a version of the Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) module online in the NASA GEOS-5 model; that is, the aerosol and trace gas species are dynamically and radiatively interactive within the atmospheric general circulation model. We do not at present have a treatment of the aerosol indirect effect in GEOS-5. We emphasize particularly the regional climate and hydrological cycle in southern Asia and its sensitivity to aerosol composition and loading. Three classes of simulations are considered: no radiative forcing by aerosols, forcing by a prescribed climatology of aerosols, and fully interactive forcing by the online aerosol distributions. The simulations are evaluated for their representativeness with climatological aerosol observations from MODIS and AERONET.

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Dust Storm Reduction due to Precipitation and Temperature Enhancement in Northwestern China: A Direct Climatic Impact of Absorbing Aerosols

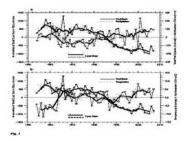
Y. Gu; ¹; K. Liou; ¹; W. Chen; ²; H. Liao; ²;

- 1. Atmospheric and Oceanic Sciences and Joint Institute for Regional Earth System Science and Engineering, University of California, Los Angeles, Los Angeles, CA, United States.
- 2. Institute of Atmospheric Physics, Chinese Academy of Science, Beijing, China.

Body: Dust storms originating in the Gobi desert and northwestern China critically impact weather, climate, and public health in China and neighboring Pacific Rim countries. The frequent occurrence of dust storms has been attributed to both deforestation and the changing environment. Dust storm formation is determined by a number of factors, including dryness, wind field, soil type, and precipitation, with precipitation being the most essential factor. Dust storms normally originate in northwestern China where annual precipitation is less than 400 mm, particularly in extremely dry areas (less than 200 mm), including the Taklamakan Desert, Tarim basin area, and Gobi Desert, where the most severe dust storms have been reported. In the decades between 1954 and 2007, reports of annual dust storm occurrences at 753 Chinese meteorological sites and the corresponding amount of total precipitation show a reduction in the occurrence and intensity of dust storms and clearly demonstrate an inverse relationship between the two. The correlation between dust storm occurrence and temperature in northwestern China also displays a negative trend but is less significant. Using a global climate model, we demonstrate that increased loading of light-absorbing aerosols in China, such as black carbon (BC), is the primary reason for precipitation and temperature increases over northwestern China, and the consequence of reductions in dust storm frequency and intensity. The model-simulated precipitation and temperature changes over northwestern China compare reasonably well with observed trends when a certain portion of absorbing aerosols has been added to the model, which significantly affects regional climate patterns through the heating of the air column.

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Anomalies of the observed annual total dust storm cases during the period from 1954 to 2007 (solid) and the corresponding anomalies of the observed annual mean (a) total precipitation (mm) and (b) surface temperature (C°) (dashed), along with their 5-year mean values.

Constraining aerosol histories with modeled and observed clear-sky surface solar radiation fluxes in Asia and Europe *C. Ruckstuhl;* ¹; *J. R. Norris;* ¹;

1. Scripps Institution of Oceanography, UCSD, La Jolla, CA, United States.

Body: Aerosol radiative forcings are key uncertainties in climate change. IPCC-AR4 20th century simulations, for example, largely fail to reproduce the recent observed trends in surface solar irradiance, even when radiative effects of cloud cover variations are removed. Incorrect aerosol histories are the most likely cause for the disagreement between models and observations. We test this hypothesis by running the Community Atmosphere Model CAM3.1 with four different sulfate and black carbon aerosol histories. Three of the aerosol histories were used in the fourth IPCC assessment report, whereas the fourth aerosol history is prepared for the forthcoming fifth IPCC assessment report. We furthermore constrain the aerosol histories by comparing modeled clear-sky solar irradiance at the Earth's surface with the observed fluxes in China, Japan, and Europe from which cloud cover radiative effects have been removed. The rapid growth of anthropogenic aerosol emissions in China makes this region exquisite to evaluate emission histories. Preliminary results show that CAM3.1 simulations forced with the different aerosol histories all tend to produce clear-sky dimming in China that is much smaller than observed, thus implying that emission increases during the last several decades might be underestimated.

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SULFATE PRODUCTION IN CLOUDS IN EASTERN CHINA: OBSERVATIONS FROM MT. TAI

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- 2. Department of Civil and Structural Engineering, Hong Kong Polytechnic University, Hong Kong, China.
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Body: The fate of China's sulfur dioxide emissions depends, in part, on the ability of regional clouds to support rapid aqueous oxidation of these emissions to sulfate. Sulfur dioxide oxidized in regional clouds is more likely to be removed by wet deposition while sulfur dioxide that undergoes slower gas phase oxidation is expected to survive longer in the atmosphere and exert a radiative forcing impact over a broader spatial scale. Two 2008 field campaigns conducted at Mt. Tai, an isolated peak on the NE China plain, provide insight into the importance of various aqueous phase sulfur oxidation pathways in the region. Single and two-stage cloudwater collectors were used to collect bulk and drop size-resolved samples of cloudwater. Collected cloudwater was analyzed for key species that influence incloud sulfate production, including pH, S(IV), H_2O_2 , Fe and Mn. Other major cloud solutes, including inorganic ions, total organic carbon, formaldehyde, and organic acids were also analyzed, as were gas phase concentrations of SO₂, O₃, and H₂O₂. A wide range of cloud pH was observed, from below 3 to above 6. High concentrations of cloudwater sulfate were consistent with abundant sulfur dioxide emissions in the region. Despite its fast aqueous reaction with sulfur dioxide, high concentrations of residual hydrogen peroxide were measured in some clouds implying a substantial capacity for additional sulfate production. Ozone was found to be an important S(IV) oxidant in some periods when cloud pH was high. This presentation will examine the importance of different oxidants (H2O2, O3, and O₂ catalyzed by trace metals) for sulfur oxidation and the overall capacity of regional clouds to support rapid aqueous phase sulfate production.

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Relating Precipitation Phenomena with MODIS Detected Hot Spots in the Maritime Continent E. M. Ramirez; ¹; J. S. Reid; ²; P. Xian; ²; E. Hyer; ²; J. Turk; ³; M. Flatau; ²; C. Zhang; ⁴;

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- 4. Division of Meteorology and Physical Oceanography, Rosenstiel School of Marine & Atmospheric Science, Miami, FL, United States.

Body: Recent studies of land use practices in SE Asia's Maritime Continent (MC) have raised questions over potential meteorological implications including smoke-cloud interaction and changes in the regional radiation budget. Land management practices on Java, Sumatra, Sulawesi, Borneo, and the Malay Peninsula include biomass burning to clear primary forest as well as to maintain oil palm plantations. Burning is also employed for clearing unwanted remains from previous crops, rice stubble for example. However, burning activity is often dictated by weather, in particular precipitation. We studied 5 years of MODIS active fire hot spot and satellite precipitation data to investigate how observed burning activity correlated with precipitation features at four major scales: 1) Intraseasonal El Nino-Southern Oscillation (ENSO) and Indian Ocean Dipole (IOP); 2) Seasonal migration of the Intertropical Convergence Zone (ITCZ); 3) the 30-90 day Madden Julian Oscillation (MJO); and 4) regional convection from localized weather phenomenon (e.g., orographic, isolated thermal convection, sea breeze, etc...). It was found that observed burn patterns from each of the islands of the MC had differing responses to these four forcings. Observed burning activity on the islands of Borneo and Sulawesi correlated strongly with the ENSO and ITCZ indicies, whereas Sumatra was more influenced by the phase and strength of the MJO. Java, showing a mix of influence, exhibited strong fire activity in the morning rather than afternoon which is unusual for most burning regions. We hypothesize that these observed relationships reflect both physical land use differences and contextual bias/observability issues associated with the region's heavy cloud cover. For further studies on smoke-meteorology interaction, our findings point to the need for a clear understanding of the meteorological context of satellite observations.

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Surface Measurements of dust/local aerosol properties over Northern China during 2008 China-US joined dust field

X. Wang; 1; J. Huang; 1;

1. Lanzhou University, Lanzhou, China.

Body: The objective of this study is to understand the detailed characteristics and underlying mechanisms of aerosol physical and optical parameters over China Loess Plateau and its potential impacts on the regional/global climate. In order to characterize the emission, transport, and removal of atmospheric pollutants emitted from East Asia, the 2008 China-US joined field campaign are conducted from late April to May 2008 focused specifically on the Asian direct measurements of dust and pollution transport, following the plume from the Northern China which from the Taklamakan desert and Gobi desert to the Eastern Pacific and into North America. Such measurements are crucial to understanding how the dust and the pollution plume (including black carbon) are modified as their age. Three sites involved this campaign, including one permanent site (Semi-Arid Climate & Environment Observatory of Lanzhou University (SACOL)) (located in Yuzhong, 35.95N/104.1E), one SACOL's Mobile Facility (SMF) (deployed in Jintai, 37.57N/104.23E) and the U.S. Department of Energy Atmospheric Radiation Measurements(ARM) Ancillary Facility (AAF mobile laboratories, SMART-COMMIT) (deployed in Zhangye, 39.08N/100.27E). Results indicate that the dust plumes are transported from the surface to a long distance from their sources have a significant influence on the air quality in the study area. The meteorological analysis indicates that these polluted layers are not from local sources during dust plume and this large-scale transport of dust and pollutants remains a major uncertainty in quantifying the global effect of emissions from Northern China.

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Retrieving optical properties of dusty clouds from MFRSR and Lidar measurements

T. Wang; 1; J. Huang; 1;

1. Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of Atmospheric Sciences, Lanzhou Univiersity, Lanzhou, Gansu, China.

Body: Based on the scattering properties of nonspherical dust aerosol, a new method is developed for retrieving dust aerosol optical depths of dusty clouds. The dusty clouds are defined as the hybrid system of dust plume and cloud. The new method is based on transmittance measurements from surface-based instruments Multi-filter Rotating Shadowband Radiometer (MFRSR) and cloud parameters from Lidar measurements. It uses the difference of absorption between dust aerosols and water droplets for distinguishing and estimating the optical properties of dusts and clouds, respectively. This new retrieval method is not sensitive to the retrieval error of cloud properties and the maximum absolute deviations of dust aerosol and total optical depths for thin dusty cloud retrieval algorithm are only 0.056 and 0.1, respectively, for given possible uncertainties. The retrieval error for thick dusty cloud mainly depends on Lidar-based total dusty cloud properties. This algorithm was applied to retrieve the dusty cloud properties by using MFRSR and Lidar Measurements, during 2008 China-US joined dust field campaign (March-June 2008). This presentation will provide the preliminary results.

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Tianhe Wang, Lanzhou, Gansu, China, 730000, click here to send an email

Development of an effective lidar retrieval algorithm using lidar measurements during 2008 China-US joined dust field campaign

Z. Huang; 1; J. Huang; 1;

1. Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of Atmospheric Sciences, Lanzhou university, Lanzhou, Gansu, China.

Body: In this study, an effective algorithm was developed to retrieve aerosol optical properties and vertical profile using ground-based lidar measurements. The advantage of this algorithm is that aerosol optical depth retrieving from lidar measurements do not need so-called lidar ratio for same quality retrieved by Sun photometer of AERONET. Also, errors were apparently reduced when retrieving other optical properties using obtained-AOD as constraint. This effective algorithm was applied to retrieve the dust aerosol vertical profiles measured by three MPL-net Micro-Pulse Lidar system, which are located at one permanent site (Semi-Arid Climate & Environment Observatory of Lanzhou University (SACOL)) (located in Yuzhong, 35.95N/104.1E), one SACOL's Mobile Facility (SMF) (deployed in Jintai, 37.57N/104.23E) and the U.S. Department of Energy Atmospheric Radiation Measurements(ARM) Ancillary Facility (AAF mobile laboratories, SMART-COMMIT) (deployed in Zhangye, 39.08N/100.27E)., during 2008 China-US joined dust field campaign (March-June 2008). A dust storm case which widely influenced Northwest China for 2 May, 2008 was studied using the three ground-based lidar and satellite-borne instruments measurements. The results show the different aerosol vertical structures at each site. Characteristics of aerosol vertical structure in spring over Northwest China were also investigated using the new method.

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Radiative Effect Difference between Multi-layered and Single-layer Cloud Derived from CERES ,CALIPSO and Cloudsat

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- 2. Science Systems and Applications Incorporated, Hampton, VA, United States.
- 3. NASA Langley Research Center, Hampton, VA, United States.

Body: Cloud effects on the general circulation through modification of the radiative heating profile within the atmosphere are complex, depending on the height, vertical structure and phase of clouds. In order to determined the instantaneous cloud radiative effect (CRE) induced by multi-layered (ML) and single-layer (SL) clouds, data collected by CALIPSO, CloudSat and the Clouds and Earth's Radiation Energy Budget Scanner (CERES) from March 2007 through February 2008 are analyzed and the differences of CRE between ML and SL clouds at top-of-atmosphere (TOA) and surface are examined. It is found that the zonal mean shortwave (SW) differences of CRE between ML and SL clouds at the TOA and surface are almost positive at most of latitudes, peak in the tropics at 120 W/m2 and drop - 30W/m2 at higher latitudes. It indicates that the ML clouds than SL cloud usually reflect less sunlight to TOA due to their higher cloud top heights, and increase the downward SW radiation at the surface. The zonal mean longwave (LW) differences of CRE between ML and SL clouds at the TOA and surface are relative small, ranging from -30 W/m2 to 30 W/m2. It shows that ML cloud will increase more thermal radiation toward the TOA relative to SL cloud only at tropic, decrease thermal radiation toward the TOA at else latitudes, that is the ML clouds relative to SL cloud tend to cool the atmosphere in the tropics and warm the atmosphere in the middle and high latitudes. The zonal mean net CRE differences are positive at most latitudes and dominated by the SW CRE differences.

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Field observation analysis of atmosphere-land surface interaction over Loess Plateau of Northwest China G. Wang; ¹; J. Huang; ¹; W. Guo; ²;

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- 2. Key Laboratory of Regional Climate-Environment Research for Temperate, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China.

Arid and semi-arid area of Loess Plateau in northwestern China as one of the dust aerosol sources and the climate change sensitive region, have a unique climate characteristic. However, there was lack of long-term atmosphere-land surface interaction observation for this region in the past. In order to understand the energy budget, energy partitioning, surface energy balance and water cycles, data from Jan 2007 to Dec 2008 observed at SACOL (Semi-Arid Climate and Environment Observatory of Lanzhou University, 35°57'N, 104°08'E, elev. 1961m) are used. Seasonal and diurnal variations in radiation components and energy components, especially energy budget, energy partitioning, surface energy balance, sensible heat flux and latent heat flux in different land surface are examined. The results indicate that the highest surface albedo value occurred in winter, as result of snow covered, it can be reached as high as 0.9. But it is a little lower in the growing season (wet season), it mainly because it was covered by the grass, and the higher of soil moisture, the lower of surface albedo. Sensible (latent) heat flux was the main consumer of available energy in winter and spring (summer and autumn). Energy imbalance problem has also been encountered in SACOL. If the soil heat storage in the surface soil and vegetation canopy above the plate is neglected, the energy imbalance can be reached to 22%. However if the surface heat storage was calculated by TDEC (Thermal Diffusion Equation and Correction), the energy imbalance is only 14%. If the soil heat storage is taken into account, the energy imbalance is only 8% in spring, but 15% in the summer and autumn season. It is significantly that the heat storage in vegetation canopy can not be neglected in growing season either. In addition, the sensible heat flux and latent heat flux in different land surface over the semi-arid area of Loess Plateau were analyzed. It is found that the sensible heat flux and latent heat flux show a significant difference in different land surface as result of the differences in vegetation, precipitation and soil moisture.

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Is Anti-Twomey effect real or an artifact?

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- 2. Institute of Earth Sciences, Hebrew University of Jerusalem, Jerusalem, Israel.

Body: Aerosol indirect effects are generally referred to any aerosol induced modification of cloud microphysics, cloud life time, precipitation etc. All these effects are based on examining relative changes in cloud droplet size by aerosol. Twomey hypothesized that droplet size decreases with increase in aerosols for a fixed liquid water content (LWC), which has been supported with several studies. A wide variation is noted for the sensitivity of cloud microphysics to aerosol and some were attributed to the different observational / analysis methods and natural variation. All previous studies showed Twomey effect (droplet size decreases for increase in aerosol) and some studies showed Anti-Twomey effect (cloud droplet size increases by adding aerosol) but did not elaborate on the finding. Using MODIS data, Yuan et al (2008) observed dominant Anti-Twomey effect over southeastern US and southeastern China but did not find any dependence of aerosol and cloud droplet effective radius (DER) over Indian region. In the present study, extensive aircraft measurements were being carried out over Indian subcontinent to document the aerosol and cloud microphysical properties under Cloud Aerosol Interaction and Precipitation Enhancement Experiment (CAIPEEX) during pre-monsoon, active and break monsoon conditions. To investigate the effect of aerosols on DER, simultaneous measurements of aerosol-CCN concentration, DER and LWC are analyzed. The datasets are grouped into different LWC bins and the lowest bin of 0-0.25 gm-3 which is generally found around the cloud base is used. For a given aerosol type, always greater total CCN concentrations were associated with larger cloud drop number concentrations (CDNC) and smaller DER at a given cloud depth above its base. However, when a tail of large haze aerosols was added, it had the opposite effect on the cloud properties. Giant CCNs can reduce the number of cloud droplets under polluted conditions by suppressing the supersaturation reached in a cloud base. The reduced CDNC together with the presence of giant CCNs could lead to increased cloud droplet size and decrease in CDNC. One may state that the giant CCN that reduce cloud base supersaturation have an Anti-Twomy effect in these special conditions. These conditions were noted over the west coast regions during active monsoon conditions, and during the heavy hazy atmosphere during the monsoon over the Indo-Gangetic Plains. The abundance of giant CCN probably incurred the heavy haze that extended up to a height of 4 km. However, to test its significance the data was constrained with fixed cloud thickness versus total CCN. When this is done the Anti-Twomey effect turns in to Twomey effect. From the CAIPEEX measurements, observational evidence to such contrasting aerosol-cloud interactions, error in perception of anti-Twomey effect and accurate quantification of Twomey effect will be presented.

Reference

Yuan, T., Z. Li, R. Zhang, and J. Fan (2008), Increase of cloud droplet size with aerosol optical depth: An observation and modeling study, J. Geophys. Res., 113, D04201, doi:10.1029/2007JD008632.

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Effects of aerosol and greenhouse gasses on the summertime Asian monsoon rainfall trend in the 20th century M. Arai; ¹; T. Miyasaka; ²; T. Nozawa; ³; T. Nagashima; ³; M. Kimoto; ¹;

- 1. Center for Climate System Research, Chiba, Japan.
- 2. Department of Earth and Planetary Science, University of Tokyo, Tokyo, Japan.
- 3. National Institute of Environmental Studies, Tsukuba, Japan.

Body: To evaluate anthropogenic effects on summertime Asian monsoon precipitation changes in the twentieth century, we compared atmosphere-ocean coupled model experiments with different external forcings. A linear trend of precipitation over the Asian monsoon region in each

experiment indicated that the effect of anthropogenic forcing dominated that of natural forcing (Fig1(a), (b)). The contributions of increased greenhouse gases and anthropogenic aerosols were both significant, but their polarity was opposite (Fig1(c), (d)). We found that the twentieth-century precipitation trend remote from the anthropogenic aerosol emission source had a large projection onto the principal mode of natural variability (Fig1(e)). The impact of increased greenhouse gases and anthropogenic aerosols tended to appear as a modulation of the natural variability. Aerosol processes are important to consider for precise evaluations of changes in Asian monsoon precipitation in the twentieth century.

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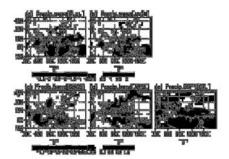


Figure 1. (a-d): 100 year trends of precipitation (colors in units of mm day-1/100 years) and wind at the 850 hPa level (vectors in units of ms-1/100 years) for experiments with (a) full, (b) only anthropogenic, (c) only greenhouse gasses, and (d) only aerosols forcing. Shadings denote the 95% significance level, judged by the Mann-Kendall test. The contour intervals of precipitation are 0.5 mm/day/100 years for (a) and (b), and 0.3 mm/day/100 years for (c) and (d). Panel (e): Precipitation anomalies associated with the leading EOF of precipitation in the control experiment, obtained by regressing seasonal-mean anomalies on to the principal component time series. The area used for the EOF analysis is denoted by a rectangle: 40E–160E, 15S–40N.

Spatial and Interspecies Patterns of PM2.5 at Three Sites in the Pearl River Delta, China: One-Year Observations A. K. Lau; ²; J. Yu; ¹; C. Wu; ²; W. Ng; ²; Z. Yuan; ²; D. Wu; ³;

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Body: The Pearl River Delta (PRD) is among the most economically fast-developing regions in China. The region has been experiencing increasing levels of particulate matter (PM) pollution. In an effort of establishing long-term trend in chemical characteristics of PM2.5 and understanding PM sources important at regional scale, filter-based samples have been collected at three sites in the PRD concurrently in one-in-six-day schedule since August 2007. We here report observation results of PM2.5 over one-year period (August 2007-June 2008). The three sites include an urban downtown location in Guangzhou, Nansha, a rural receptor site at the mouth of the Pearl River, and Tsuen Wan, an urban background site in Hong Kong. Guangzhou recorded the highest annual average PM2.5 concentration of 78.2 μgm-3, followed by Nansha (65.9 μgm-3) and Tsuen Wan (42.8 μgm-3). Organic matter (OM) and sulfate are the top two constituents, accounting for ~70% of PM2.5 mass. The annual average nitrate contributions were similar at GZ and NS (~13%), but lower at TW (~7%). Inter-site correlations of PM2.5 and major constituents indicate that GZ strongly influenced ambient PM2.5 levels at NS, but GZ's influence on TW was much reduced. Sulfate, ammonium, and OM showed strong regional characteristics. To the contrary, EC at the three sites had no correlations, suggesting a dominating local origin. Examples of high PM2.5 episodes are also analyzed to identify the conditions conducive for high PM.

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Seasonal variation of spherical aerosols distribution in East Asia based on ground and space Lidar observation and a Chemical transport model

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Body: The anthropogenic aerosols largely impact on not only human health but also global climate system, therefore air pollution in East Asia due to a rapid economic growth has been recognized as a significant environmental problem. Several international field campaigns had been conducted to elucidate pollutant gases, aerosols characteristics and radiative forcing in East Asia. (e.g., ACE-Asia, TRACE-P, ADEC, EAREX 2005). However, these experiments were mainly conducted in springtime, therefore seasonal variation of aerosols distribution has not been clarified well yet.

National Institute for Environmental Studies (NIES) has been constructing a lidar networks by automated dual wavelength / polarization Mie-lidar systems to observe the atmospheric environment in Asian region since 2001. Furthermore, from June 2006, space-borne backscatter lidar, Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), onboard NASA/CALIPSO satellite, measures continuous global aerosol and cloud vertical distribution with very high spatial resolution. In this paper, we will show the seasonal variation of aerosols distribution in East Asia based on the NIES lidar network observation, Community Multi-scale Air Quality Modeling System (CMAQ) chemical transport model simulation and CALIOP observation over the period from July 2006 to December 2008.

We found that CMAQ result explains the typical seasonal aerosol characteristics by lidar observations. For example, CMAQ and ground lidar showed a summertime peak of aerosol optical thickness (AOT) at Beijing, an autumn AOT peak at Guangzhou and summertime AOT trough at Hedo, Okinawa. These characteristics are mainly controlled by seasonal variations of Asian summer/winter monsoon system. We also examined the CMAQ seasonal average aerosol extinction profiles with ground lidar and CALIOP extinction data. These comparisons clarified that the CMAQ reproduced the observed aerosol layer depth well in the downwind region. Ground lidar and CALIOP seasonal aerosol profiles also showed good agreement with the aerosol layer depth and concentration level. We are also quite successful to present the Asian-scale 3-D seasonal horizontal/vertical distributions of spherical aerosol extinction coefficient based on the composite analysis of CMAQ and CALIOP, which clarify the variation of transport pathway and spherical aerosol layer thickness for each season.

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Spectral Discrimination of Fine and Coarse Mode Aerosol Optical Depth from AERONET Direct Sun Data of Singapore and South-East Asia

S. Salinas Cortijo; 1; B. Chew; 1; S. Liew; 1;

1. National University of Singapore, Singapore, Singapore.

Body: Aerosol optical depth combined with the Angstrom exponent and its derivative, are often used as a qualitative indicator of aerosol particle size, with Angstrom exp. values greater than 2 indicating small (fine mode) particles associated with urban pollution and bio-mass burning. Around this region, forest fires are a regular occurrence during the dry season, specially near the large land masses of Sumatra and Borneo. The practice of clearing land by burning the primary and sometimes secondary forest, results in a smog-like haze covering large areas of regional cities such as cities Singapore, Kuala Lumpur and sometimes the south of Thailand, often reducing visibility and increasing health problems for the local population. In Singapore, the sources of aerosols are mostly from fossil fuel burning (energy stations, incinerators, urban transport etc.) and from the industrial and urban areas. The proximity to the sea adds a possible oceanic source. However, as stated above and depending on the time of the year, there can be a strong biomass component coming from forest fires from various regions of the neighboring countries. Bio-mass related aerosol particles are typically characterized by showing a large optical depth and small, sub-micron particle size distributions. In this work, we analyze three years of direct Sun measurements performed with a multi-channel Cimel Sun-Photometer (part of the AERONET network) located at our site. In order to identify bio-mass burning events in this region, we perform a spectral discrimination between coarse and fine mode optical depth; subsequently, the fine mode parameters such as optical depth, optical ratio and fine mode Angstrom exponents (and its derivative) are used to identify possible bio-mass related events within the data set.

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Carbonaceous and ionic compositions of PM10, PM2.5 and PM1 in Seoul during 2007-2008 *J. Han;* ¹; *M. Lee;* ¹; *S. Lim;* ¹; *J. Kim;* ¹; *I. Park;* ¹; *G. Lee;* ²; *J. Han;* ³;

- 1. Earth&Environmental Sciences, Korea university, Seoul, Korea, Republic of.
- 2. Environmental Science, Hankuk University of Foreign Studies, Young-In, Gyeonggi-Do, Korea, Republic of.
- 3. Atmospheric Research, National Institute of Environmental Research, Incheon, Korea, Republic of.

Body: The size of aerosol is determined by their origin, chemical and physical composition. The goals of this research were to measure the mass concentration of PM₁₀, PM_{2.5}, and PM₁ and their soluble ions and organic matters to understand their chemical property in each size. Here we describe measurements of PM₁₀, PM_{2.5}, and PM₁ in Seoul, South Korea during 2007-2008. Daily-integrated aerosol samples were collected using Cyclone. Water-soluble ions, elemental carbon (EC) and organic carbon (OC) were analysis. In mass concentration, Asian dust plume was well distinguished in PM_{10} while anthropogenic pollution plume in PM_{1} . The variation of $PM_{2.5}$ was similar to that of PM_{10} . Major water-soluble ions such as SO_4^{2-} and NH_4^+ were dominant in PM_1 . but $NO_{3_2}^-$ didn't show clear size dependency. EC(OC) concentrations of PM $_{2.5}$, and PM $_{1}$ were 4.61(9.72), 4.04(9.08) μ g/m 3 . EC and Organic matter $(OM=OC\times1.4)$ constituted 49.9% and 15.8% of PM_1 mass, respectively. In Asian dust event, non-identified matters were increased. Monthly mass fraction of PM₁₀, PM_{2.5}, and PM₁ were correlated with the wind direction. Portion of Carbonaceous and non-identified matters were increased in February and May when dominant westerly wind, which is mainly due to long-range transport. On the other hand, water-soluble ions increased in September when dominant easterly wind.

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A quantitative analysis for nitrogen source-receptor relationships in Northeast Asia based on numerical simulation Y. Sunwoo; 1; Y. Kim; 1; Y. Ma; 1; J. Park; 1; K. Choi; 1; I. Chang; 2; J. Kim; 2;

- 1. Konkuk University, Seoul, Korea, Republic of.
- 2. National Institute of Environmental Research, Incheon, Korea, Republic of.

Body: The long-range transport of air pollutants in Northeast Asia is a very relevant current issue. In recent years, the annual growth rate of nitrogen dioxide(NO2) concentration over the industrial areas in China has significantly increased, as high as about 50% during 1996~2004. In this study, numerical simulations using CMAQ were performed to understand source-receptor relationships for nitrogen from HNO3, NO3- and PAN in Northeast Asia. Modeling domain was arranged with 19°N-45°N, 98°E-147°E and had 60km horizontal grid resolution. SAPRC99 was applied as chemical mechanism. The modeling period was March and July in 2006. Hourly meteorological field and emission data were produced from MM5 and SMOKE, respectively. The emission inventory from the INTEX-B project was used for SO2, NOx, VOCs, CO, PM10. The emissions of NH3 were estimated using TRACE-P emission inventory in 2000 and REAS projection factors. Emission inventory was redistributed to 60km resolution grid using spatial allocation factor. Source-receptor relationships were analysed using EMEP method-3. This method was used based on the deposition amount due to emission in some area. The emissions were computed as a difference between the model run with all emissions and the model run with all emissions except the part of emissions from some area. In this study, Northeast Asia was divided into 5 regions as Korea, Japan and 3 regions(north, central and south) of China. NOx emissions were reduced to 20% over 5 regions, respectively and air quality modeling was simulated. Dry and wet deposition of nitrogen were calculated. Significant higher total deposition amount was found over central and north-east China in July. However, higher deposition amount appeared in north-east China. Wet deposition amount showed about 4 was times of dry deposition in March. However, Dry and wet deposition had similarly amount each other in July. In result of source-receptor relationship analysis, we could find as follows. The high self sourcereceptor relationships were showed in central and south China. The emissions from central China influenced to all receptor region over 30% in March. Seasonal variations of source-receptor relationship for Korea and Japan were high. The impact of central China was over 50% for Korea and about 40% for Japan in March. However, self sourcereceptor relationships were the highest for both sites in July.

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Secondary Organic Aerosol Formation in Urban Air: Temporal Variations and Possible Contributions from Unidentified Hydrocarbons

M. Koike; ¹; H. Matsui; ²; N. Takegawa; ²; Y. Kondo; ²; R. J. Griffin; ³; Y. Miyazaki; ⁴;

- 1. University of Tokyo, Tokyo, Japan.
- 2. RCAST, Univ. of Tokyo, Tokyo, Japan.
- 3. Rice University, Houston, TX, United States.
- 4. Hokkaido Univ., Sapporo, Japan.

Body: Quantitative evaluation of the performance of one of the most advanced mechanistic secondary organic aerosol (SOA) modules/models Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution 2 (MADRID2) in the threedimensional Models-3/Community Multiscale Air Quality (CMAQ), in urban air is made. Model calculations were compared for the Tokyo, Japan, metropolitan area with measurements made using an Aerodyne quadrupole aerosol mass spectrometer (Q-AMS) at an urban site for nine days in July and August 2003. In general, model calculations reproduced absolute values and temporal variations of meteorological parameters, C2-C8 volatile organic compounds (VOCs), NOx (NO + NO2), inorganic aerosols, and O3 concentrations reasonably well at this site. However, model calculated SOA concentrations are a factor of five smaller than observed oxygenated organic aerosol (OOA) concentrations, and calculated total organic aerosol (OA = SOA + primary organic aerosol) concentrations are smaller by a factor of two, indicating missing processes or sources in the current organic aerosol model calculations. On the other hand, observed features of diurnal and day-to-day variations of OOA are captured by our model calculations. Because of the large quantity of unidentified total non-methane VOCs (NMVOCs) in urban air, a possible contribution of SOA formation from high-molecular-weight VOCs is examined through simple sensitivity studies, in which emissions are increased to account for unidentified NMVOCs. It was found that they have potential to be one of the missing SOA sources, demonstrating the importance of reliable measurements of high-molecular-weight VOCs and total NMVOCs. Relationships between SOA and O3, including regional enhancements (~150 x 150 km2) around the Tokyo metropolitan area, also are discussed.

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The biggest control knob: carbon dioxide in Earth's climate history (Invited)

R. B. Alley; 1, 2;

- 1. Geosciences, Pennsylvania State Univ, University Park, PA, United States.
- 2. Earth and Environmental Systems Institute, The Pennsylvania State University, University Park, PA, United States.

Body: CO2 particularly, and greenhouse gases more generally, appear to have played the largest role in controlling Earth's climate history, based on emerging results across a range of subdisciplines. Detection and attribution of climate change are clearly much easier in the instrumental era than earlier, but understanding of these earlier times is improving rapidly. Very strong evidence shows the importance of many climate forcings, including volcanic eruptions, features of Earth's orbit, and continental rearrangement. Extensive speculation is attached to other putative forcings, such as changing cosmic rays, space dust, and magnetic fields. However, with varying levels of completeness ranging from balance-of-evidence to pound-on-the-table near-certainty, attribution studies indicate that major features of the Earth's climate system are primarily explained by changing greenhouse-gas levels, and especially CO2 levels. These climatic features include the long-term persistence of liquid water under a fainter young sun, the snowball-Earth events, the warmth of the Cretaceous, the sudden warming into the Paleocene-Eocene thermal maximum, and amplification of the orbital features during ice-age cycling. The implied climate sensitivity for these slow events is somewhat larger than for the faster changes normally discussed with anthropogenic warming, but the models used to project anthropogenic warming show considerable skill relative to the Earth's climate. A historical perspective thus supports the mainstream scientific assessments of the IPCC, CCSP, etc. strongly, while opening the possibility of somewhat larger changes than often projected.

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Has the Age of Stratospheric Air Decreased over the Past Three Decades (1979-2008)?

Q. Fu; ¹;

1. University of Washington, Seattle, WA, United States.

Body: The age of stratospheric air is the average time to transport an air parcel from tropical tropopause to a given location in the stratosphere, which is a good indictor of the strength of the Brewer-Dobson circulations (BDC). GCMs with good representation of the stratosphere predict an increase in the strength of the BDC in response to an increase of greenhouse gas concentrations and to the ozone depletion. Thus the mean age of stratospheric air is expected to decrease in last three decades. In this talk I will present the observational analyses of satellite MSU lower stratospheric temperatures, indicating an annual mean dynamic cooling (warming) in the tropics (high latitudes) due to the change of the BDC. Based on a unique relationship between the vertical velocity and the temperature in the tropical lower stratosphere, we derived that the BDC (and thus the stratospheric age) has strengthened (decreased) for 1979-2008 by about 13% with a 95 % confident interval of 5-20%. Our observational analysis supports the GCM simulations but contrasts with recently derived long-term trend of mean stratospheric age based on observed SF6 and CO2 concentrations.

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Sensitivity of the Strength of the Brewer-Dobson Circulation to SST Perturbations in an Aqua-planet Model *G. Chen;* ¹; *R. A. Plumb;* ²; *J. Lu;* ³;

- 1. Earth and Atmospheric Sciences, Cornell University, Ithaca, NY, United States.
- 2. Program of Atmospheres, Oceans and Climate, MIT, Cambridge, MA, United States.
- 3. Atmospheric, Oceanic, and Earth Sciences, George Mason University, Fairfax, MD, United States.

Body: Recent studies with chemistry climate models predict a strengthening of the Brewer-Dobson Circulation (BDC) in response to climate change. This BDC change has been attributed to parameterized gravity wave drag and resolved planetary and synoptic wave drag. Here we examine the sensitivity of the BDC strength to the resolved wave drag in an aqua-planet atmospheric model driven by forcings described in the aqua-planet model benchmark (Neale and Hoskins 2000). SST warming is systematically introduced to either low latitudes or high latitudes with different meridional extents, and thus modifies the generation of baroclinic waves in the troposphere.

The tropospheric jet response is quite sensitive to the location and sign of the gradient of SST perturbations with respect to the climatological jet. Roughly speaking, the jet moves towards the flank where the baroclinic wave generation is enhanced due to an increase of anomalous SST gradient. Despite the complexity in the change of resolved waves, the strength of the BDC is increased for all the experiments with low latitude warming, but is decreased only for the experiments with high latitude warming extending to the subtropics. The change of the residual circulation in the stratosphere is consistent with the change in the resolved wave drag. These experiments indicate that the subtropical branch of the BDC is primarily controlled by changes in the subtropical winds that provide waveguides for the upward propagation of resolved synoptic and planetary waves.

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Stratospheric Circulation Changes Inferred From Observations of Ozone and Other Trace Gases

E. A. Ray; 1, 2; F. L. Moore; 1, 2; K. H. Rosenlof; 2; J. W. Elkins; 2; A. Engel; 3;

- 1. CIRES, University of Colorado, Boulder, CO, United States.
- 2. NOAA/ESRL, Boulder, CO, United States.
- 3. J. W. Goethe University, Frankfurt, Germany.

Body: We use several of the very few long-term measurements of trace gases in the stratosphere to infer decadal scale changes in the Brewer Dobson circulation. Inferred perturbations in the circulation due to volcanoes and sea surface temperature changes as well as a long-term trend are consistently evident in TOMS/SBUV total ozone, mean age of air derived from SF6 and CO2, water vapor and photolytic tracer measurements. We also use a simple model of the stratosphere, the tropical leaky pipe model, to estimate the scale and location of circulation changes that can result in the observed stratospheric trace gas changes. The goal of this study is to show that we have observable indicators that give a consistent picture of short-term and long-term stratospheric circulation changes.

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What factors affect planetary wave driving in the Southern Hemisphere in spring?

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- 1. NASA Postdoctoral Program, NASA Goddard Space Flight Center, Greenbelt, MD, United States.
- 2. Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, MD, United States.
- 3. Atmospheric Chemistry and Dynamics Branch, NASA Goddard Space Flight Center, Greenbelt, MD, United States.
- 4. Science Systems and Applications, Inc., Lanham, MD, United States.
- 5. Global Modeling and Assimilation Office, NASA Goddard Space Flight Center, Greenbelt, MD, United States.

Body: The timing of the breakup of the Antarctic vortex is determined by planetary wave driving at Southern Hemisphere (SH) mid-latitudes in spring. A previous modeling study found that weaker than observed heat flux in October and November led to a delayed breakup of the Antarctic vortex. In an attempt to better understand the seasonal cycle of SH heat flux at 100hPa and increase wave driving in version 2 of the Goddard Earth Observing System (GEOS) chemistry-climate model (CCM), this work examines the sensitivity of heat flux to a variety of boundary conditions and model parameters. The inclusion of a quasi-biennial oscillation (QBO), the choice of photolysis rates and the representation of gravity wave drag do not affect October/November heat flux. Furthermore, boundary conditions (such as sea surface temperatures) do not determine the heat flux, as the magnitude and seasonal cycle of 100hPa heat flux differs between CCMs using the same 'recent past' scenario. Heat flux magnitudes in the critical October/November period, and thus the timing of the vortex breakup, are improved when either the horizontal resolution or SH precipitation is increased.

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Static Stability in the Global Upper Troposphere and Lower Stratosphere: Observations of Long-term Mean Structure and Variability using GPS Radio Occultation Data

K. M. Grise; 1; D. W. Thompson; 1; T. Birner; 1;

1. Atmospheric Science, Colorado State University, Fort Collins, CO, United States.

Body: Static stability is a fundamental dynamical quantity that measures the vertical temperature stratification of the atmosphere. The long-term mean static stability field is characterized by the well-known transition from low values in the troposphere to high values in the stratosphere. However, the magnitude and structure of fine-scale static stability features near the tropopause are difficult to discern in temperature data with low vertical resolution. In this study, the authors apply over six years of high vertical resolution Global Positioning System radio occultation temperature profiles to document the long-term mean structure and variability of static stability in the global upper troposphere and lower stratosphere (UTLS).

The results of this study demonstrate that a shallow but pronounced maximum in static stability exists just above the tropopause at all latitudes (i.e., the "tropopause inversion layer," or TIL). This study also uncovers two novel aspects of static stability in the global UTLS. In the tropical lower stratosphere, the results reveal a unique vertically and horizontally varying static stability structure, with maxima located at ~17 km and ~19 km. The upper feature peaks during the NH cold season and has its largest magnitude between 10 and 15 degrees latitude in both hemispheres; the lower feature exhibits a weaker seasonal cycle and is centered at the Equator. The results also demonstrate that the strength of the TIL is closely tied to stratospheric dynamic variability. The magnitude of the TIL is enhanced following sudden stratospheric warmings in the polar regions and the easterly phase of the quasi-biennial oscillation in the tropics.

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Residual circulation trajectories and transit times into the extratropical lowermost stratosphere

T. Birner; 1;

1. Department of Atmospheric Science, Colorado State University, Fort Collins, CO, United States.

Body: Transport into the extratropical lowermost stratosphere (LMS) can be divided into a slow part (time-scale of years) associated with the global-scale residual (Brewer-Dobson) circulation and a fast part (time-scale of days to months) associated with (mostly quasi-horizontal) mixing (i.e. two-way irreversible transport, including stratospheretroposphere exchange). The Brewer-Dobson circulation can be considered to consist of two branches: a deep branch more strongly associated with planetary waves breaking in the middle to upper stratosphere, and a shallow branch more strongly associated with synoptic-scale waves breaking in the subtropical lower stratosphere. In this study the contribution due to the Brewer-Dobson circulation alone to transport into the LMS is quantified using residual circulation trajectories, i.e. trajectories driven by the residual mean meridional and vertical velocities. This contribution represents the reversible (advective) part of the overall transport into the LMS and can be viewed as providing a background onto which the effect of mixing has to be added. Residual mean velocities are obtained from a comprehensive chemistry-climate model as well as from ECMWF reanalysis data. Residual transit times of air traveling from the tropical tropopause to the LMS along the residual circulation streamfunction are evaluated and compared to mean age of air estimates. A clear time-scale separation with much smaller residual transit times into the mid-latitudinal LMS than into polar LMS is found that is indicative of a clear separation of the shallow from the deep branch of the Brewer-Dobson circulation. In contrast mean age of air exhibits a much more homogeneous latitudinal structure. Nevertheless, the residual transit time distribution reproduces qualitatively the observed seasonal cycle of youngest air in the fall and oldest air in the spring.

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Impacts of Asian Summer Monsoon on Seasonal and Interannual Variations of tropical upper tropospheric pollution over South Asia

- 1. Dept. of Atmospheric and Oceanic Sciences, UCLA, Los Angeles, CA, United States.
- 2. Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, United States.

Body: Deep convection associated with the Asian summer monsoon circulation is a primary transport pathway for lifting surface pollution along with water vapor into the upper troposphere and lower stratosphere (UTLS) over South Asia. We investigate the intra-seasonal and inter-annual variabilities of this deep convective transport by analyzing satellite observations of ozone, carbon monoxide (CO), water vapor, ice water content (IWC) and related trace gases from the Microwave Limb Sounder (MLS) and/or Tropospheric Emission Spectrometer (TES). Our analysis focuses on May-September, 2005 to 2008. Outgoing longwave radiation (OLR), South Asian High intensity, and winds derived from the NCEP/NCAR reanalysis data are examined in relation to the satellite observations to understand the role of Asian summer monsoon onset and strength on the convective-lifting of pollution. GEOS-Chem global model simulations, including "tagged" CO simulations and adjoint sensitivity analyses are conducted to quantify the relative contributions from different source regions and types to pollution in the UTLS over South Asia.

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Transport from the Asian monsoon to the stratosphere observed in satellite measurements of hydrogen cyanide W. J. Randel; 1; M. Park; 1; D. E. Kinnison; 1; L. K. Emmons; 1; P. F. Bernath; 2; K. A. Walker; 3; H. C. Pumphrey; 4.

- 1., Boulder, CO, United States.
- 2. University of York, Heslington, United Kingdom.
- 3. University of Toronto, Toronto, ON, Canada.
- 4. University of Edinburgh, Edinburgh, United Kingdom.

Body: Satellite observations of hydrogen cyanide (HCN) from ACE-FTS data is used to identify the transport of polluted air masses from the surface, through the Asian monsoon, deep into the stratosphere. A key factor in this identification is that HCN has a strong sink from contact with the ocean; much of the air in the tropical upper troposphere is relatively depleted in HCN, and hence broad tropical upwelling cannot be the main source for the stratosphere. The ACE-FTS data are combined with MLS observations to show that interannual variations in stratospheric HCN are linked to input from the Asian monsoon region. Results from the WACCM chemistry transport model, incorporating realistic HCN sources, are in reasonable agreement with the satellite observations. These results provide definitive demonstration of transport to the stratosphere through the Asian monsoon.

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On the use of CloudSat and Calipso to Study Clouds Over Sea Ice in the Southern Beaufort Sea.

L. M. Candlish; 1; D. G. Barber; 1;

1. Centre for Earth Observation Science, University of Manitoba, WInnipeg, MB, Canada.

Body: Average atmospheric conditions in the Arctic are often close to algorithmic boundary conditions of many commonly used GCM's, and can cause inaccurate results. A lack of data for atmospheric modeling in the Arctic limits the validity of model output; atmospheric models require field validation. This presentation entails a comparison of two satellites CloudSat and CALIPSO, with empirical data collected during the Circumpolar Flaw Lead System Study (CFL) and the ArcticNet Cruise 2009 in the Western Canadian High Arctic over sea ice.

Satellite-derived parameterizations of Arctic atmospheric conditions at large spatio-temporal scales include surface/cloud temperature, atmospheric temperature, and cloud properties. CloudSat's 94-Ghz cloud profiling radar (CPR) and CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation)'s 532-nm and 1064-nm Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar are the first satellites with the capability to vertically profile the structure of Arctic clouds. The combined radar-lidar cloud detection technique relies on the backscattered energy from the cloud particles. The weak thermal and albedo contrasts between clouds and the ice-covered Arctic surface make other cloud detection techniques insufficient in the Arctic due to their reliance on passive radiances. Validation data including manual observations for cloud fractional coverage, vapor density, and lapse rates from radiosondes, and a ground based microwave vertical profiler are used to quantify forcing mechanisms at seasonal time scales. In-situ atmospheric data continuously collected over the period of November 2007 to August 2008 and July 2009 to November 2009 is categorized to validate CloudSat and CALIPSO data products. The ancillary data product, ECMWF, has been examined and compared with temperature and vapor density profiles from radiosondes and our ground based vertical microwave profiler. Seasonal trend analysis has been performed on CALIPSO's cloud base height data product (CALIPSO Lidar Level 2 1/3 km cloud layer data) and compared with the ceilometer data for low/mid level clouds. The comprehensive collection of field observations and remote sensing analysis provides an accuracy assessment for CloudSat's and CALIPSO's data products in the High Arctic.

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FM09: Question regarding A31A-0074'>click here to send an email

The impact of 21st Century sea ice decline on the hydrological budget of the Arctic

J. J. Day; ¹; J. L. Bamber; ¹; P. J. Valdes; ¹; J. Kohler; ²;

- 1. School of Geographical Science, University of Bristol, Bristol, United Kingdom.
- 2. Norwegian Polar Institute, Tromsø, Norway.

Body: The Arctic is a region particularly susceptible to rapid climate change. GCMs suggest a polar amplification of any global warming signal by about 1.5 due, largely, to sea ice feedbacks. The dramatic recent decline in multi-year ice cover lies outside the standard deviation of the ensemble GCM predictions and has lead to the suggestion that the Arctic Ocean could be ice free in summer as soon as ~2014.

Sea ice acts as a barrier between cold air and warmer oceans during winter, as well as inhibiting evaporation from the water below during the summer. An ice free Arctic would likely have an altered hydrological cycle with more evaporation from the ocean surface leading to changes in precipitation distribution and amount. For example, changes in sea ice cover are thought to have caused changes in the mass balance of Europe's largest ice cap, Austfona, Svalbard, by increasing accumulation.

Using the U.K. Met Office Regional Climate Model (RCM), HadRM3, the atmospheric effects of the observed and projected reduction in Arctic sea ice are investigated. The RCM is driven by the atmosphere only general circulation model HadAM3. Both models are forced with sea surface temperature and sea ice obtained by extrapolating recent changes into the future using bootstrapping based on the HadISST climatology. Here we use an RCM at 25km resolution over the Arctic which captures well the present-day pattern of precipitation and provides a detailed picture of the projected changes in the behaviour of the oceanic-atmosphere moisture fluxes and how they affect precipitation.

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Diurnal albedo of seasonally melting Arctic snow at wavelenghts of UV and visible

O. Meinander; 1;

1. Climate Change, Finnish Meteorological Institute, Helsinki, Finland.

Body: Seasonally melting Arctic snow at Sodankylä (67°22' N, 26°39' E, 179 m a.s.l.), Finland, allows us to study the diurnal and long-term changes in snow albedo and other snow properties, combined with AWS measured meteorological parameters. Albedo of snow is wavelength dependent, and here we concentrate on wavelengths of UV and visible. As the cloud effect for UV wavelengths is smaller than for the whole solar spectrum, UV albedo remains less affected, too. We have found the midday erythemally weighted UV albedo to range from 0.6 to 0.8 in the accumulation period, and from 0.5 to 0.7 during melting. Moreover, we have detected (i) albedo cases with an unexpected diurnal decrease of 0.05 in albedo soon after midday, and recovery thereafter, as well as (ii) albedo cases with SZA asymmetry up to 10 %. The diurnal decrease with the recovery (i) was obvious during the snow melt period, under cases of an almost clear sky and variable cloudiness. Using ancillary data, we suspect this diurnal albedo change to be due to the daily metamorphosis of the surface of the snowpack, in which the temperature of the surface increases, melting some of the snow to liquid water, after which the surface freezes again. In the SZA asymmetry (ii), a diurnal decrease in UV snow albedo as a function of time was found (higher albedo in the morning and lower in the evening). These included cases with either new snow on the previous night, or daytime snow melt combined with refreezing during night, or high relative humidity and low surface temperature during the previous night favorable to frost and higher albedo on the next morning. Further studies on the importance of these diurnal and spectral dependencies to climate-albedo feedback, and to parametrizations of snow albedo in various models, are needed.

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Impact of including fully interactive Greenland and Antarctic ice sheets on the climate sensitivity of an Earth system model of intermediate complexity

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- 3. Laboratoire de Physique Atmosphérique et Planétaire, Institut d'Astrophysique et de Géophysique, Université de Liège, Liège, Belgium.

Body: We use the Earth System Model of Intermediate Complexity LOVECLIM to show the effect of coupling fully interactive ice sheets on the simulated climate response to a 2xCO2 stabilization scenario. For this purpose a number of different parameter sets have been defined for LOVECLIM, covering a wide range of the models sensitivity to greenhouse warming. We analyze the effect and physical mechanism of ice-climate interactions introduced by the dynamic ice sheets for the ensemble of different parameter sets.

We find a lower climate sensitivity of the model when fully coupled ice sheets are included, an effect, which scales with increasing freshwater fluxes from the melting Greenland and Antarctic ice sheets. In both cases, changes in oceanic heat uptake and sea-ice-albedo feedbacks play a mayor role in attenuating the warming.

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Simulations of Vegetation Impacts on Arctic Climate

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Body: Because global warming disproportionately influences high-latitude climate, changes in arctic vegetation are in progress. These land-cover changes include redistribution of local vegetation types as well as northward migration of lower-latitude species in response to the increasing warming. The resulting displacement of low-lying tundra vegetation by shrubs and trees darkens the surface, thus accelerating regional warming. As participants in the U.S. Department of Energy IMPACTS Project, we are investigating the potential for abrupt arctic climatic change resulting from such variations in vegetation, among other mechanisms.

To estimate the relative magnitudes of effects to be expected from changes in high-latitude land cover, we are conducting several numerical experiments with the Community Climate System Model (CCSM). These experiments include:

- 1) A "present-day-climate" control experiment with current atmospheric greenhouse-gas concentrations and climatological monthly sea surface temperatures and sea ice extents prescribed, and with "standard" CLM plant functional types (PFTs) specified;
- 2) A "changed-vegetation-type" experiment that is the same as 1), except that the "standard" PFTs are augmented by additional vegetation types (forbs, sedges, shrubs, mosses, and lichens) that are not presently represented in CLM. This experiment will require information on the location, fractional cover, and physiological parameterizations of these new PFTs.
- 3) A "changed-vegetation-extent experiment" that is the same as 2), except that the spatial extents of selected PFTs (e.g. shrubs or boreal forest PFTs) are shifted northward from their present locations in the CLM.

We will report on the atmospheric climate and land-surface feedbacks associated with these vegetation changes, with emphasis on local and regional surface energy and moisture fluxes and near-surface temperature, humidity, and clouds.

Acknowledgments

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Investigating the effects of Eurasian snow cover on winter climate

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Body: sing NCEP/NCAR and ERA40 reanalysis, the effects of Eurasian (EA) snow cover on the Arctic Oscillation (AO) is better quantified. This teleconnection has been hypothesized to involve three parts: 1. a tropospheric Rossby wave pulse in response to fall snow-forced diabatic cooling; 2. subsequent absorption in the stratosphere, resulting in higher geopotential heights and weaker westerlies; and 3. downward propagation of these anomalies to the surface, culminating in a negative wintertime AO. We find a more significant snow-AO relationship exists during those years with anomalously high EA fall snow cover, suggesting that some minimum areal snow cover extent is required to initiate the response. We also find a more significant snow-AO relationship during those years with an anomalously weak fall polar stratospheric vortex, consistent with more wave absorption by a weaker vortex. Similarly, we find a stronger snow-AO relationship when the Quasi-biennial Oscillation (QBO) is easterly. This is likely due to the QBOs effects on the wave guide for upward propagating planetary waves, so that a weaker polar vortex (and hence more wave absorption) is favored when the QBO is easterly. These insights suggest reasons why General Circulation Models (GCMs) are unable to reproduce the snow-AO relationship. Transient experiments using the Community Atmosphere Model are performed to investigate the possible shortcomings of modeled EA snow cover. We find that experiments with prescribed snow cover yield a marginally improved snow-AO relationship.

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Exploring the Role of Sea-Ice for Seasonal Forecasts

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2. European Centre for Medium-Range Weather Forecasts, Reading, United Kingdom.

3. Norwegian Institute for Air Research, Kjeller, Norway.

Body: Most coupled ocean-atmosphere models used for seasonal forecasting prescribe climatological conditions for sea-ice, which may imply that the predictions are biased and have too weak spread. In the coupled operational seasonal forecast model at ECMWF (IFS/HOPE), the sea-ice normally is relaxed towards climatology after the first month.

We present results from dedicated experiments with the same coupled model set-up where sensitivity of the 2007 circulation to sea-ice conditions is explored by prescribing observed sea ice cover throughout the simulation, rather than climatology. The experimental set-up consists of 5-month long, 5-member ensemble simulations, taking atmospheric and ocean initial conditions from May 2007 and October 2007. The sea-ice is prescribed from 2007, and the sensitivity is further explored by prescribing observed sea-ice from preceding years (6 in total). The objective of this work is to assess whether a sea-ice cover in the models that is more realistic than climatology has an effect on the ensemble spread and gives an improved forecast.

The impact of the Arctic sea-ice on northern hemisphere high- to mid-latitude circulation anomalies and on storm statistics is also examined. Storm statistics is derived from a calculus-based cyclone identification (CCI) algorithm proposed by Benestad and Chen (2006), which also is used to identify high-pressure centres.

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A comparison of polar vortex trend response to Pacific and Indian Ocean warming

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Body: During the past decades the tropical Indo-Pacific Ocean has become warmer than before. Meanwhile, both the northern and the southern hemispheric polar vortex (NPV and SPV) exhibit a deepening trend in boreal winter-half year. Although previous studies reveal that the tropical Indian Ocean Warming (IOW) favors intensifying the NPV and weakening the SPV, how the tropical Pacific Ocean Warming (POW) influences the NPV and the SPV is unclear. In this study, a comparative analysis is conducted through ensemble atmospheric general circulation model (AGCM) experiments. The results show that, for the northern hemisphere, the two warming exert an opposite impact in boreal winter, in that the IOW intensifies the NPV while the POW weakens the NPV. For the southern hemisphere, both the IOW and POW warm the southern polar atmosphere and weaken the SPV. A diagnostic analysis based on vorticity budget reveals that such an interhemispheric different influence in boreal winter between the IOW and the POW is associated with the different roles of transient eddy momentum flux convergence. Furthermore, this difference may be linked to the different strength of stationary wave activity between the hemispheres in boreal winter.

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Modulation in Interannual Sea-Ice Patterns in the Southern Ocean in Association with Large-Scale Atmospheric Mode Shift

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- 3. Institute of Observational Research for Global Change, Japan Agency for Marine-Earth Science and Technology, Yokosuka, Kanagawa, Japan.
- 4. Faculty of Bioresources, Mie University, Tsu, Mie, Japan.

Body: We verified that the synchronous propagations of the spatial patterns of sea ice concentration (SIC) of wavenumber 2 around the Antarctic occurred only for the period 1984 to 1994. An empirical orthogonal function (EOF) analysis of satellite data for 1979–2003 objectively demonstrates that the spatial pattern of SIC propagated eastward only in 1984–1994, in other years, it did not. Our results show that interannual variations in SIC patterns are associated with differences in the dominant large-scale atmospheric patterns. In non-propagating years, variance of the tropospheric Antarctic oscillation (AAO) predominated. However, in propagating years, the AAO variance was subdominant to that of the Pacific South American (PSA) teleconnection pattern having a 4-year period. Such periodic PSA enables the SIC anomalies to propagate eastward with a periodically reinforced dipole pattern. The shift of large-scale atmospheric variability is one possible cause of the modulation in the SIC pattern. The switch of the atmospheric EOF leading mode from the PSA pattern to the AAO in the mid-1990s corresponded to the modulation in the SIC pattern and supports the presence of the atmospheric climate shifts [Udagawa et al., 2009 JGR in press]. We executed model experiments using AGCM to investigate the cause of atmospheric mode shift.

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Atmospheric forcing of Antarctic sea ice

J. A. Renwick; 1;

1. Climate Variability & Change, NIWA, Wellington, New Zealand.

Body: The seasonal variation of Antarctic sea ice extent is one of the largest geophysical changes in the Earth's annual cycle, effectively doubling the size of Antarctica in the winter months. Sea ice forms a natural barrier between the atmosphere and the ocean, modulating surface energy exchanges at high latitudes. Understanding the role of Antarctic sea ice in the climate system, and its response to anthropogenic forcing, is a key issue in climate research.

Beyond the seasonal cycle of ice growth and decay, Antarctic sea ice extent is strongly affected by the atmospheric circulation, on times scales from days to decades. A long-term increasing trend in total sea ice extent has been linked to the Southern Annular Mode, and is likely to have been influenced by ENSO teleconnections. On the seasonal scale, there is a strong ENSO signal in sea ice extent across the Pacific, associated with atmospheric Rossby wave propagation. On daily to weekly time scales, atmospheric waves also modulate sea ice extent, around the whole of the Antarctic continent, but especially across the Pacific sector and into the Weddell Sea.

This presentation will review the climatology of Antarctic sea ice extent, long-term trends and SAM/ENSO-related forcing of sea ice, and will discuss the nature of shorter-term interactions between the atmospheric circulation and sea ice concentration.

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The Feedback of Sea Ice Loss on Arctic Temperatures: 1970-2008

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2. NOAA, NCEP, Camp Springs, MD, United States.

Body: One of the most striking features of high latitude climate change has been the reduction in Arctic sea ice cover. The rate of decline in recent years has been especially surprising, and has been beyond that anticipated from projections by IPCC Fourth Assessment Model simulations. The question we address is the impact of the sea ice decline on temperatures over the tundra of North America and Eurasia during 1970-2008. We diagnose large ensembles of parallel climate simulations, one set using the observed monthly evolution of ocean surface temperature and sea ice conditions, and the second subjected only to the observed ocean temperature variations. The difference between these twin simulations reveals the climate sensitivity to sea ice loss, and illustrates the intensity of feedback of sea ice trends on Arctic terrestrial temperature increases since 1970.

Sea ice loss is found to been responsible for 50%-75% of the overall warming of Arctic land surface temperatures since 1970. Sea ice loss is found to exert a stronger warming of the North American tundra than of the Eurasian tundra. Although the most significant reduction in sea ice has ben observed to occur during summer and early fall, our simulation indicate that the largest impact on Arctic warming has been in winter and spring.

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The Energetics of a Warming Arctic

T. Graham; ¹; A. Ferraro; ¹; M. Vellinga; ¹; P. Wu; ¹;

1. Hadley Centre, UK Met Office, Exeter, United Kingdom.

Body: The inconsistency between observed and simulated rates of recent Arctic sea-ice decline demands a better understanding of energetics of model simulated Arctic warming. Using various ensembles of perturbed HadCM3 simulations, we investigate the sensitivity of sea-ice decline to ocean heat uptake, air-sea fluxes and atmospheric radiative balance and their changing relationships in a warming climate.

In a 22-member ensemble, we find consistent amplification of Arctic surface air temperature changes but much smaller SST changes in both polar regions. This is because the extra solar radiation absorbed by the ocean due to decreasing sea-ice and surface albedo in the summer is mostly lost by increased outgoing long wave radiation in the Autumn with a much reduced air-sea temperature difference.

The change in heat loss from the ocean in fall varies widely between the different perturbed physics ensemble members and is shown to be correlated with both the change in the sea-ice area and initial sea-ice area. The change in solar heat flux into the ocean does not show this dependency and varies much less between the perturbed physics ensemble members.

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Very large warming in Antarctic mid-troposphere in winter: PSCs are implicated.

H. K. Roscoe; ¹; T. A. Lachlan-Cope; ¹;

1. British Antarctic Survey, Cambridge, United Kingdom.

Body: Recent discoveries show that over the last 30 years Antarctic mid-tropospheric temperatures in winter have increased by 0.5 degC/decade. This is very much larger than global or hemisphere-wide trends over the same time period. The reasons for this large warming are not understood: heat advected horizontally into Antarctica shows negligible change; a climate model run with increased greenhouse gases gives a warming of only 0.2 degC/decade. Increased polar stratospheric clouds (PSCs) are a possible cause because PSCs would have their maximum effect in winter, and the increase in greenhouse gases that has taken place in the last 30 years acts to cool the stratosphere and so to increase PSCs. A preliminary model run with increased cloud in the stratosphere showed a warming that partly explains the observations. More model runs, including a proper representation of PSCs, together with increased greenhouse gases and increased PSCs, are needed to confirm that PSCs are indeed responsible.

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On the Interactions of Arctic Sea Ice, Cloud Cover, and Surface Temperature from Satellite Observations

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Body: High latitude climate system contains many complex climate feedbacks, e.g. the albedo-temperature, and surface-cloud feedbacks. Better understanding of the complex interactions between multiple components involved in these feedbacks would improve our understanding of mean climate state, recent dramatic changes, and possible future changes in the high latitudes. With the sharp decrease in Arctic sea ice extent and concentration in recent decades and probable continuous decrease through this century, the impact of sea ice changes along with cloud cover changes on the Arctic surface temperature trends become extremely important. In this study, a linear model is developed and applied to the study of the effect of changes in sea ice concentration (SIC) and cloud cover on surface temperature trends. The influence of the trends in SIC and cloud cover on the trends in surface temperature over the Arctic Ocean from 1982 to 2004 is investigated analytically, and evaluated empirically with satellite products. The results demonstrate that the changes in SIC and cloud cover played major roles in the magnitude of recent Arctic surface temperature trends. After eliminating the effects of the changes in SIC and cloud cover on surface temperature trends, the residual surface temperature trends can be used in a more robust diagnosis of surface warming or cooling in the Arctic. The same model can also be applied to completely different sets of climate variables, whether they are measured or modeled. Meanwhile, the relationship between changes in surface condition and cloud amount are investigated using satellite observations.

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Atmosphere-sea ice interactions over a wide range of climates

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- 1. California Institute of Technology, Pasadena, CA, United States.
- 2. University of Washington, Seattle, WA, United States.

Body: Arctic sea ice is one of the components of the climate system that has changed most during recent decades, and it is projected to continue to change rapidly during the coming century. Yet our knowledge of the system, as indicated by the spread among our best climate models, is currently quite limited. In order to investigate the basic physical relations controlling the sea ice cover, we simulate a wide range of climates using an idealized general circulation model (GCM) above an ocean mixed layer with sea ice. Among the results from these simulations, we find that the interaction between sea ice feedbacks and atmospheric processes cause (i) the area of the seasonal sea ice zone, (ii) the maximum thickness of first-year ice, and (iii) the ice growth versus thickness relationship to all remain approximately constant as the ice edge varies from tropical to polar latitudes. Physical mechanisms responsible for the simulated behavior, as well as implications for simulations of future sea ice retreat with state-of-the-art coupled GCMs, are discussed.

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Parameterization of the Solar Fluxes over Mountain Surfaces for Application to Regional Climate Models W. Lee; ¹; K. Liou; ²; A. Hall; ²;

- 1. Research Center for Environmental Changes, Academia Sinica, Taipei, Taiwan.
- 2. Department of Atmospheric and Oceanic Sciences and Joint Institute for Regional Earth System Science and Engineering, University of California, Los Angeles, Los Angeles, CA, United States.

Body: Surface solar fluxes over a complex terrain are strongly affected by the variations in elevation, slope, and albedo. However, these factors have not been accounted for in the contemporary radiative transfer schemes used in regional climate models, which assume that the lower boundary is flat and homogeneous. To include the effects of shadowing and multiple reflections among mountains, we have developed a parameterization intended for use in regional climate models on the basis of simulations from a three-dimensional Monte Carlo photon tracing radiative transfer program applied to irregular and inhomogeneous terrain. Eighty 10×10 km2 areas with a 90 m resolution in Sierra Nevada Mountains in North America have been selected to compute anomalies of the domain-averaged solar fluxes at mountains with reference to a flat surface under a clear sky condition (with background aerosols). Multiple regression analysis has been carried out to formulate surface fluxes as a function of various topographic parameters. Results of the analysis reveal that more than 70% of the variations in direct and diffuse fluxes can be explained by a simple linear function with only three parameters: solar incident angle, standard deviation of elevation, and sky view factor, while more than 90% of the variation in the reflected fluxes can be explained by a single parameter-terrain configuration factor.

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Effects of cryospheric changes on Earth's solar energy budget *M. Flanner*; ¹;

1. AOSS, University of Michigan, Ann Arbor, MI, United States.

Body: Seasonal snow cover and sea-ice have evolved significantly over the past 30 years, indicating that powerful feedback may be at play in Earth's climate system. Here, the influence of recent cryospheric changes on Earth's surface and top-of-atmosphere (TOA) energy budgets are investigated using satellite observations, the NCAR Community Climate System Model (CCSM), and a radiative transfer model. Three factors strongly modulate the effect of such changes on Earth's TOA solar energy flux. First, regions subject to frequent and thick cloud cover (at any altitude) experience heavily muted TOA responses to snow/ice cover alteration. Second, effects of snow changes over land depend strongly on vegetation cover, with surface and TOA forcings limited in regions where mature forests maintain a low maximum surface albedo during the snow season. Finally, the seasonal timing of cryospheric change is critical. Snow cover and sea-ice changes during local spring and mid-summer, respectively, have the strongest influence because of intense insolation and large capacity for change (in present-day climate). Various climate simulations with and without prescribed snow cover and sea-ice are compared with CERES and ERBE observations, helping constrain modeled effects of cryospheric changes under different cloud and vegetation conditions.

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An Improved Method For Retrieving TOA Albedo Over Polar Regions Using MISR

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1. Physics Department, University of Auckland, Auckland, New Zealand.

Body: The top-of-atmosphere (TOA) albedo is an important factor in determining the Earth's energy budget. However, current methods used by NASA's Multi-Angle Imaging-Spectroradiometer (MISR) instrument to retrieve albedos tend to overestimate polar albedo. This is because the measurements tend to be close to the solar azimuthal plane at high solar zenith angles where there is a lot of forward scattering over snow and ice surfaces, leading to albedo estimates that are occasionally unphysical. In this study we correct such behaviour by using the CERES angular dependence models (ADMs) for snow and ice to provide proxies for the azimuthal angles not sampled by MISR. By utilising MISR's unique viewing arrangement of 9 cameras at different viewing zenith angles we are also able to obtain 9 independent estimates of the albedo for a given scene. These are then averaged to give a best estimate of the albedo. MISR's multiangle approach thus provides additional sampling that directly reduces the uncertainty in regional albedos. It also provides the ability to choose the model which best fits the measured reflectances – removing biases caused by incorrect scene identification. Our preliminary findings indicate the most significant improvements occur over ephemeral sea-ice and when clouds are present over snow or ice. A previous study using MISR albedos indicated that the correlation between top-of-atmosphere albedo anomalies and sea-ice fraction anomalies peaks after 30 days when the sea ice anomalies are lagged against albedo anomalies. This result, suggesting a measurable ice-albedo feedback effect, will be confirmed with the new albedos.

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Storm-induced polar warming and moistening under anthropogenic forcing

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- 2. National Institute of Meteorological Research, Seoul, Korea, Republic of.
- 3. University of Hawaii, Honolulu, HI, United States.

Body: Amplified polar warming and moistening under global warming are critical issues for the climate changes. The authors find that a poleward shift of the westerly jet stream and associated storm feedback play a critical role in enhancing polar warming and moistening. Storm feedback in this case follows a simple rule: providing anomalous heat and moisture fluxes to the left-hand side of the anomalous jet stream. This rule can be used for understanding other climate responses to the anthropogenic forcing.

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Modulation of Orbital Induced Climate Change by Ice, Cloud and Water Vapor Feedbacks

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- 2. Department of Environmental Sciences, Rutgers University, New Brunswick, NJ, United States.

Body: Climate feedbacks are investigated by changing the Earth's obliquity in the GFDL CM2.1 coupled model. A reduction in obliquity increases the meridional gradient of the annual mean insolation, which causes a strengthening of the ocean and atmospheric circulation which transport more heat poleward. The heat transport does not balance the obliquity forcing completely and additional local radiative fluxes are required to maintain equilibrium. The surface temperature increases in the tropics and reduces at high latitudes following the change in the insolation. However, in the subtropics and midlatitudes, the sign of the temperature change is opposite to the forcing, indicating the strong influence of feedbacks. These feedbacks are also responsible for a decrease in the global mean temperature despite the fact that the change in the global mean insolation is zero. The processes responsible are the increase in the ice fraction at high latitudes and the global cloud fraction, both of which reduce the absorbed solar radiation. The reduction in the global greenhouse trapping, due to changes in the distribution of the water vapor content of the atmosphere has an additional cooling effect. The changes in the water vapor content are thermodynamically induced, following the change in the temperature, but also dynamically induced, following a change in the circulation. The findings presented here highlight the ubiquitous role of feedbacks on climate change.

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The rapidly shrinking sea ice cover and ice-albedo feedback effects

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Body: Among the most dramatic changes on the Earth's surface that may be associated with anthropogenic climate change is the rapid decline in the Arctic perennial sea ice cover. In 2007, the perennial ice declined precipitously with its area being 37% lower than climatological average and 28% lower than the previous low established in 2005. In 2008, the perennial ice recovered somewhat because of colder global temperatures but by only about 6% of average value. The trend in the ice area covered by perennial ice is now -12.5% per decade which compared to a previous report of -9% per decade indicates an accelerated decline. Studies of the multiyear ice cover as detected by satellite sensor in winter indicate an even more drastic change. The multiyear ice as detected in winter represents ice that has generally survived two summers and regarded as thicker ice types. Analysis of the thicker multiyear ice types indicates a more rapid decline of 15.5 % per decade. This implies a thinning in the Arctic ice cover as well. Much of the decline occurred in the western region of the Arctic Basin (i.e., Chukchi and Beaufort Seas) where the open water area has been increasing by about 35% per decade. The decline in the sea ice cover causes an increase in low albedo ice free ocean regions that absorb a lot more solar energy than ice covered regions. This causes further decline in the ice cover in a process called ice-albedo feedback. The impact of such process is a warmer mixed layer of the Arctic Ocean. This is consistent with results from analysis of satellite sea surface temperature (SST) data in the region which indicates a warming of 0.52 +/- 0.23 degrees Celsius per decade during the months of August from 1979 to 2008. Results of sensitivity analysis that includes the use of a simple thermodynamic model to assess the net effect of ice-albedo feedback compared to contributions from other factors will be presented.

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Overview Of The Development And Evaluation Of The NCEP Global Aerosol Modeling System: Particulates From

Biomass Burning

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- 4. University of Maryland Baltimore County, Baltimore, MD, United States.

Body: Aerosols particles are abundant in the atmosphere and can absorb and/or reflect the incoming solar radiation. Atmospheric aerosols also can act as cloud condensation nuclei that can change cloud formation and further impact the overall radiation budget. Therefore, the distribution of atmospheric aerosol can be an important element for global meteorology models while computing the global radiation budget. The current NOAA/NWS National Centers for Environmental Prediction (NCEP) Global Forecasting System (GFS) uses a primitive assumption on the spatial distribution of atmospheric aerosol due to limitations from computation resources and a more complete understanding of aerosol radioactive impact. To enhance the forecasting capability of the GFS, NCEP is developing a global aerosol modeling capability. The development began with an offline dust model using the NASA GOCART aerosol model dust component driven by the GFS forecasts. Recent developments include adding the capability to simulate aerosol from biomass burning sources. This offline aerosol model has also been developed in order to serve as an initial benchmark for a planned inline implementation; to provide aerosol lateral boundary conditions for regional air quality forecasting (AQF) systems; and to provide modeled aerosol fields for assimilation of satellite and in-situ data.

NCEP GFS-GOCART global aerosol simulations using GFEDv2.0 for 2006 summer are performed. AERONET and MODIS atmospheric optical depth (AOD) datasets are used to evaluate the model performance. The impact of the aerosols from biomass burning on the NOAA national AQF capability (AQFC) is examined. With the inclusion of aerosol lateral boundary condition from GFS-GOCART, it revealed a dramatic improvement of the U.S. AQF fine particulate matter (PM2.5) simulations when compared with the AIRNOW PM2.5 observations in Texas and Florida during the summer 2006.

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Improve MISR's Capability of Predicting Ground Level PM2.5 Concentrations with Observed Aerosol Vertical Profiles (

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Body: Satellite AOD holds the promise to fill in the gaps between PM2.5 observations in air quality monitoring as ground monitoring networks are costly to operate in the United States, and generally very sparse or not available in the rest of the world. Change of aerosol vertical profiles has been identified as an important source of uncertainties causing the weakening or even breakdown of AOD-PM2.5 relationship. We have demonstrated a scaling approach which uses GEOS-Chem simulated aerosol vertical profile to strength the link between AOD and PM2.5. However, due to computational constraints, the horizontal resolution of global 3-D atmospheric chemistry and transport models such as GEOS-Chem is usually too coarse to capture aerosol structure at scales below about 200 km, and is much coarser than that for the MISR satellite data (~ 17 km). Thus, the GEOS-Chem vertical aerosol profiles used in the AOD scaling may not necessarily be representative at the scales of the MISR data. Recently, the value of groundbased (e.g., MPLNET) and spaceborne (e.g. CALIPSO) lidar data has been demonstrated in limited case studies. In this analysis, we assess the potential of aerosol vertical profiles observed by ground-based or spaceborne lidar in improving the capability of MISR AOD to predict PM2.5 concentrations in the continental US. Specifically, we first match lidar aerosol extinction profiles, MISR AOD data, and EPA PM2.5 measurements in space and time. Then two scaling approaches (simulated vs. observed aerosol vertical profiles) are used to create lower air MISR AOD to be correlated with ground level PM2.5 mass and speciation concentrations. Finally, the performance of the two scaling approaches are compared.

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Use of meteorological data to improve PM2.5 and AOD relationships in a western US valley

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Body: We used MODIS and AERONET AOD to compare with hourly and daily averaged PM2.5 mass concentration at the Fresno, California supersite. MODIS Collection 5 AOD compared much better to AERONET AOD than did MODIS Collection 4 AOD. However, AOD by AERONET or MODIS was not well correlated to PM2.5. We propose that seasonal and diurnal variations in PBL height (mixing depth) cause large variations in the AOD/PM2.5 relationship. By using mixing depth from a meteorological model and doing comparisons seasonally, much improved relationships were obtained. We conclude that for western US valley locations, such as in the California Central Valley, the use of mixing depth from meteorological models can give much more useful information than only using AOD to predict PM2.5. Use of hourly surface relative humidity to account for hygroscopic growth of particles did not significantly improve the relationship, likely due to variations in vertical humidity profiles within the PBL.

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Inferring the composition and concentration of aerosols by combining the AERONET, MPLNET and CALIOP data: comparison with in-situ measurements and utilization to evaluate and improve GCM results.

- D. Ganguly; 1, 2; P. A. Ginoux; 1; V. Ramaswamy; 1, 2;
- 1. NOAA, Geophysical Fluid Dynamics Laboratory, Princeton, NJ, United States.
- 2. Atmospheric and Oceanic Sciences Program, Princeton University, Princeton, NJ, United States.

Body: We present a method to derive the concentration of aerosol components using the spectral measurements of AOD (aerosol optical depth) and single scattering albedo along with their size distribution and extinction profile available from AERONET (Aerosol Robotic Network) and MPLNET (Micro-pulse Lidar Network) stations as well as the space borne CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) lidar [Ganguly et al., 2009a; 2009b]. The technique involves finding the best combination of aerosol concentration by minimizing differences between measured and calculated spectral variation in AOD and single scattering albedo along with the size distribution of aerosols over specific locations. Lidar data on extinction profile provides the vertical constraint on the distribution of aerosols in the atmosphere. Relative humidity from NCEP reanalysis is used to compute the hygroscopic growth factors and associated changes in the optical properties of aerosol components at all vertical levels. The technique has been successfully applied over different regions around the world such as North America, Southern Africa and South Asia. The results have been validated using in-situ measurements of aerosol composition available from the first two regions. Finally, we show how these results are being used to evaluate and improve the GFDL-AM2/AM3 climate model simulations. We believe our technique could also be used for the retrieval of air quality by calculating PM2.5 and PM10 concentrations. This could improve the existing methods by providing a better relation between surface measurements of PM2.5 concentration and satellite data.

References:

Ganguly, D., P. Ginoux, V. Ramaswamy, O. Dubovik, J. Welton, E. A. Reid and B. N. Holben (2009a), Inferring the composition and concentration of aerosols by combining AERONET and MPLNET data: comparison with other measurements and utilization to evaluate GCM output, J. Geophys. Res., 114, D16203, doi:10.1029/2009JD011895.

Ganguly, D., P. Ginoux, V. Ramaswamy, D. M. Winker, B. N. Holben and S. N. Tripathi (2009b), Retrieving the composition and concentration of aerosols over the Indo- Gangetic basin using CALIOP and AERONET data, Geophys. Res. Lett., 36, L13806, doi:10.1029/2009GL038315.

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Experiments with data assimilation in comprehensive air quality models: Impacts on model predictions and observation requirements (*Invited*)

R. Mathur; 1;

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Body: Emerging regional scale atmospheric simulation models must address the increasing complexity arising from new model applications that treat multi-pollutant interactions. Sophisticated air quality modeling systems are needed to develop effective abatement strategies that focus on simultaneously controlling multiple criteria pollutants as well as use in providing short term air quality forecasts. In recent years the applications of such models is continuously being extended to address atmospheric pollution phenomenon from local to hemispheric spatial scales over time scales ranging from episodic to annual. The need to represent interactions between physical and chemical atmospheric processes occurring at these disparate spatial and temporal scales requires the use of observation data beyond traditional in-situ networks so that the model simulations can be reasonably constrained.

Preliminary applications of assimilation of remote sensing and aloft observations within a comprehensive regional scale atmospheric chemistry-transport modeling system will be presented: (1) A methodology is developed to assimilate MODIS aerosol optical depths in the model to represent the impacts long-range transport associated with the summer 2004 Alaskan fires on surface-level regional fine particulate matter (PM2.5) concentrations across the Eastern U.S. The episodic impact of this pollution transport event on PM2.5 concentrations over the eastern U.S. during mid-July 2004, is quantified through the complementary use of the model with remotely-sensed, aloft, and surface measurements; (2) Simple nudging experiments with limited aloft measurements are performed to identify uncertainties in model representations of physical processes and assess the potential use of such measurements in improving the predictive capability of atmospheric chemistry-transport models. The results from these early applications will be discussed in context of uncertainties in the model and in the remote sensing data and needs for defining a future optimum observing strategy.

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Modeling evolution of aerosol mixing state and the associated optical and CCN activation properties (*Invited*) R. A. Zaveri; ¹; R. C. Easter; ¹; J. Barnard; ¹; N. S. Riemer; ²; M. West; ²; A. P. Ault; ³; K. A. Prather; ³;

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- 2. University of Illinois, Urbana-Champaign, IL, United States.
- 3. University of California, San Diego, San Diego, CA, United States.

Body: Primary particles such as urban soot, sea-salt, dust, etc. are externally-mixed when emitted. With time they undergo chemical and microphysical transformations due to coagulation and condensation of many different semi-volatile gases (e.g., organic, sulfuric acid, nitric acid, ammonia) and produce numerous size and mixing-state distributions with widely different optical and CCN activation properties. However, accurately tracking the mixing state in conventional aerosol models requires treating a multidimensional size distribution, which is computationally prohibitive. Consequently, current aerosols models employ simplified treatments that may not adequately resolve aerosol mixing state. In this study, we use the newly developed particle-resolved aerosol model PartMC-MOSAIC to explicitly simulate the evolution of aerosol mixing states in an idealized urban plume scenario. The results provide unprecedented insights into the effects of various processes such as emission, dilution, condensation, and coagulation on the evolution of mixing states of different types of aerosols and their optical and CCN activation properties. The effects of the internal mixture assumption commonly used by most aerosol models on the predicted optical and CCN properties will be presented. A novel approach for constraining and evaluating PartMC-MOSAIC with single particle mass spectrometer and other such measurements will be discussed.

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Rahul A. Zaveri, Richland, Washington, USA, 99352-0000, click here to send an email

The Aerosol Modeling Testbed: A Tool to Facilitate Improved Aerosol Process Modules

J. D. Fast; 1; W. I. Gustafson; 1; E. G. Chapman; 1; R. C. Easter; 1; J. Rishel; 1;

1. Pacific Northwest National Laboratory, Richland, WA, United States.

Body: Predictions of aerosol mass, composition, size distribution, hygroscopicity, and optical properties still contain large uncertainties. For example, the formation and transformation of secondary organic aerosols and the nature of many cloud-aerosol interactions are still poorly understood and consequently inadequately represented in models. When new aerosol treatments are developed, they are usually implemented into an existing aerosol model and evaluated using a limited number of measurements from a specific case study. One consequence of the current modeling paradigm is that the performance and computational efficiency of several treatments for a specific aerosol process cannot be adequately quantified because many other processes among aerosol models are different as well. For example, predictions of aerosol properties from several models have been compared, but different grid configurations, meteorology, and emission rates are often employed so that variations in predicted aerosol properties among the models were not due entirely to the treatment of aerosol processes. Thus, these studies do not quantify the range of uncertainty associated only with the predicted aerosol properties, nor does this type of uncertainty analysis provide much information on which aerosol process needs improving the most. Reducing the uncertainties associated with aerosols predictions requires that we know the advantages and disadvantages of specific aerosol treatments when the meteorology, chemistry, and other aerosol processes are identical.

To address these issues, an Aerosol Modeling Testbed (AMT) has been developed that will simplify and greatly increase the efficiency of evaluating new aerosol treatments for regional and global models. The AMT consists of a modular and user-friendly version of the Weather Research and Forecasting model (WRF-Chem), a series testbed cases consisting of extensive in situ and remote sensing measurements of meteorological, trace gas, and aerosol properties, and a suite of tools to evaluate the performance of aerosol process modules. The first testbed case completed is based on the Megacities Initiative: Local and Global Research Observations (MILAGRO) field campaign conducted in the vicinity of Mexico City during March 2006. In addition to standard in situ measurements of aerosol properties, aerosol composition from several Aerodyne Aerosol Mass Spectrometers, profiles of aerosol backscatter and extinction from the NASA High Resolution Spectral Lidar (HRSL), and aerosol optical depth from satellite instrumentation are included in the MILAGRO testbed case. Two other testbed cases are also being developed. An example of how the AMT can be used to assess the strengths and weaknesses of two aerosol models in WRF-Chem, one using a simple modal approach to represent the aerosol size distribution and the other using a more comple sectional approach, will be presented using both in situ and remote sensing data collected during MILAGRO. How the AMT can be used by the scientific community to foster collaborations and coordinate aerosol modeling research will also be discussed.

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Satellite Remote Sensing of Particulate Matter Air Quality: Progress, Potential and Pitfalls (Invited)

S. A. Christopher; 1;

1. UAHuntsville, Huntsville, AL, United States.

Body: Satellite Remote Sensing of Particulate Matter Air Quality: Progress, Potential and Pitfalls

Abstract. Fine or respirable particles with particle aerodynamic diameters less than 2.5 µm (PM2.5) affect visibility, change cloud properties, reflect and absorb incoming solar radiation, affect human health and are ubiquitous in the atmosphere. These particles are injected into the atmosphere either as primary emissions or form into the atmosphere by gas to particle conversion. There are various sources of PM2.5 including emissions from automobiles, industrial exhaust, and agricultural fires. In 2006, the United States Environmental Protection Agency (EPA) made the standards stringent by changing the 24-hr averaged PM2.5 mass values from 65µgm-3 to 35µgm-3. This was primarily based on epidemiological studies that showed the long term health benefits of making the PM2.5 standards stringent. Typically PM2.5 mass concentration is measured from surface monitors and in the United States there are nearly 1000 such filter based daily and 600 contiguous stations managed by federal, state, local, and tribal agencies. Worldwide, there are few PM2.5 ground monitors since they are expensive to purchase, maintain and operate. Satellite remote sensing therefore provides a viable method for monitoring PM2.5 from space. Although, there are several hundred satellites currently in orbit and not all of them are suited for PM2.5 air quality assessments. Typically multi-spectral reflected solar radiation measurements from space-borne sensors are converted to aerosol optical depth (AOD) which is a measure of the column (surface to top of atmosphere) integrated extinction (absorption plus scattering). This column AOD (usually at 550 nm) is often converted to PM2.5 mass near the ground using various techniques. In this presentation we discuss the progress over the last decade on assessing PM2.5 from satellites; outline the potential and discuss the various pitfalls that one encounters. We discuss the effect of clouds, the lack of global information on the vertical structure of aerosols and how the use of meteorology such as mixing layer heights and humidity can be a tremendous asset for estimating PM2.5 from satellites.

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Effective Lidar Ratios of Dense Dust Aerosol Layers over North Africa Observed by the CALIPSO Lidar

Z. Liu; 1, 2; D. M. Winker; 2; A. H. Omar; 2; M. Vaughan; 2; C. R. Trepte; 2; Y. Hu; 2; C. A. Hostetler; 2; W. Sun; 2; B. Lin; 2;

- 1. National Institude of Aerospac, Hampton, VA, United States.
- 2. NASA LaRC, Hampton, VA, United States.

Body: The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite, a joint US and French mission, was launched three years ago to provide new insight into the role that clouds and aerosols play in regulating Earth's weather, climate, and air quality. A key instrument on board the CALIPSO payload is a two-wavelength, polarization-sensitive backscatter lidar. With its capabilities of depolarization ratio measurement and high resolution profiling, the CALIPSO lidar provides a unique opportunity to study the dust aerosol globally. Currently, a cloud and aerosol discrimination (CAD) algorithm that incorporates five-dimensional probability distribution function (5D-PDF) is being developed for implementation in future data releases. This new 5D-PDF approach allows nearly unambiguous identification of dense dust layers over/near their source regions and therefore enables the study of these layers using a large amount of the CALIPSO data.

Lidar ratio (i.e., extinction-to-backscatter ratio) is an intrinsic optical property of aerosols and a key parameter necessary in the lidar signal inversion to retrieve profiles of aerosol extinction and backscatter coefficients, which are two primary products of the CALIPSO level 2 data. This parameter is usually selected in the CALIPSO lidar level 2 data processing based on the aerosol type identified. (Six types of aerosols have been modeled: dust, polluted dust, marine, continental, polluted continental, and smoke.) As more data is being collected by the CALIPSO lidar, validation studies with the CALIPSO measurements are being performed and are now becoming available. For opaque dust layers, the effective lidar ratio (the product of lidar ratio and multiple scattering factor) can be determined easily from integrated attenuated backscatter over the layer top and apparent base. We have performed an extensive analysis based on the first two and a half years (June 2006 - December 2008) of the CALIPSO lidar nighttime measurement data with the 5D-PDF CAD algorithm applied. The effective lidar ratios computed for the opaque dust layers over the North Africa (12N-30N), one of the largest source regions in the world, have a relatively broad distribution, with a mean/median value of 38.5/36.4 sr at 532 nm and 50.3/47.7 sr at 1064 nm. The experimentally determined values are in good agreement with the modeling results for Saharan dust aerosols. Monte-Carlo simulations have also been performed to examine the impact of multiple scattering. The results show that multiple scattering generally has a small impact on the effective lidar (multiple scattering factor at the layer base > 0.9). However, when the dust extinction is > ~ 2/km, the multiple scattering impact can increase significantly. A closer examination of the depolarization ratio profiles in the dense dust layers shows a general agreement with the multiple scattering simulations.

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Enhanced Aerosol Backscatter in the Vicinity of Marine Boundary Layer Clouds Revealed by Satellite-Based Lidar,

J. L. Tackett; ^{1, 2}; L. Di Girolamo; ²;

- 1. Science Systems and Applications, Inc., Hampton, VA, United States.
- 2. Atmospheric Sciences, University of Illinois at Urbana-Champaign, Urbana, IL, United States.

Body: Numerous studies have found strong correlations between cloud and aerosol properties. However, little has been published on the spatial variability of aerosols in the immediate vicinity of clouds because this is difficult to observe due to insufficient sampling from in-situ observations and 3-D radiative cloud-adjacency effects in passive remote sensing. The satellite-based lidar CALIPSO overcomes these difficulties by providing a large number of high spatial resolution samples that are unbiased by 3-D radiative cloud-adjacency effects when operated at night. Here, we provide the first CALIPSO observations of backscatter as a function of distance to cloud that can be associated with changes in aerosol properties. Though individual backscatter profiles are noisy, when we statistically analyze many transects together, a systematic trend is evident. We use nine months of nighttime CALIPSO data over the tropical western Atlantic to measure backscatter as a function of distance to cloud within the altitudes of 0.5 to 2.0 km. Systematic backscatter and color ratio enhancements are observed as cloud edge is approached, suggesting that aerosol properties are changing due to the presence of clouds. Furthermore, the vertical information provided by CALIPSO provides a unique observation: maximum enhancement occurs near the tops of thick clouds and cloud base, which may be in part due to (1) detrainment of enlarged aerosols through collision-coalescence and (2) stable layer capping of enhanced moisture and vertical moisture advection causing hygroscopic growth. We provide backscatter calculations which suggest our CALIPSO observations adjacent to cloud are best explained by an aerosol size distribution with increased median radius, decreased number concentration and decreased width compared to far from cloud. These observations provide valuable validation data for cloud-resolving models which include dynamic and chemical aerosol processing in efforts to identify the most important processing mechanisms to include in climate models.

URL: http://www.agu.org/pubs/crossref/2009/2009GL039264.shtml

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Development and Initial Testing of a Multi-Sensor Platform for Cloud-Aerosol Interactions in the Lower Troposphere A. R. Nehrir; ¹; D. S. Hoffman; ¹; K. S. Repasky; ¹; B. Todt; ²; T. Sharpe; ¹; C. Half Red; ¹; J. L. Carlsten; ²;

- 1. Electrical and Computer Engineering , Montana State University , Bozeman , MT, United States.
- 2. Physics, Montana State University, Bozeman, MT, United States.

Body: Coupled atmospheric components of the lower troposphere including aerosols and water vapor have a large affect on the chemical processes that drive the earth's complex climate system. Aerosols can affect the earth's global radiation budget directly by absorbing or reflecting incoming solar radiation, and indirectly by changing the microphysical properties of clouds by serving as cloud condensation nuclei (CCN). An increase in CCN results in higher cloud droplet concentration which has been shown to suppress drizzle formation and lead to more reflective clouds. The changes in the cloud microphysical structure due to the interaction of aerosols and water vapor result in more incoming solar radiation being reflected back into space, leading to a net negative radiative forcing in the global radiation budget. The uncertainty in this radiative forcing reflects the uncertainty in the understanding of the aerosol indirect effect and its role in the climate system. To better understand the aerosol direct and indirect effects, lidar measurements of aerosol properties and water vapor distributions can provide important information to enhance our understanding of the role of aerosols in the climate system.

The LIDAR group at Montana State University has initiated a program to simultaneously study aerosols, water vapor, and cloud formation with high spatial and temporal resolution using both active and passive sensors. Aerosol distributions and radiative properties are currently being studied with a two-color LIDAR system at 1064 and 532 nm. In addition, a three color, high spectral resolution LIDAR system at 1064,532, and 355 nm has also been developed and is starting to take initial data. Daytime and nighttime boundary layer water vapor number density profiles are also currently being studied with an external cavity diode oscillator/diode amplifier based micro-pulsed differential absorption lidar (DIAL) instrument at the 830 nm water vapor absorption band. Cloud formation studies are being conducted by a simultaneous, spatially correlated digital sky imaging camera system where aerosol loading and water vapor distributions are monitored as a function of lateral distance to clouds. Furthermore, a commercially purchased sun/sky scanning solar radiometer (CIMEL 318) as part of the NASA run AERONET program is also being used to study aerosol loading and radiative transfer through the atmosphere. A brief description of these instruments will be presented as well as initial simultaneous results showing correlated data between lower tropospheric aerosols and boundary layer water vapor distributions over extended periods if time.

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Influence of humidified aerosol on lidar depolarization observed during SHEBA

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- 1. NASA Goddard Institute for Space Studies, New York, NY, United States.
- 2. Columbia University Center for Climate Systems Research, New York, NY, United States.

Body: Lidar depolarization measurements of mixed-phase Arctic clouds can provide information about ice habits and the relative amounts of liquid and ice hydrometeors. Lidar measurements taken under the base of a mixed-phase stratocumulus deck during the SHEBA campaign show regions with surprisingly low depolarization despite the absence of cloud drops or liquid-phase precipitation. Using forward lidar computations based on large-eddy simulations with size-resolved microphysics, we show that the presence of humidified aerosol can very well explain the distribution of the observed depolarization values. Results indicate that humidified aerosol must be taken into account when interpreting lidar depolarization measurements under conditions such as those observed during SHEBA. We also explore the range of conditions under which haze particles complicate the interpretation of lidar depolarization.

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Measuring Plume Meander in the Nighttime Stable Boundary Layer with Lidar

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- 1. Environmental Sciences, Louisisana State University, Baton Rouge, LA, United States.
- 2. Natural Resources and Enviornment, The University of Connecticut, Storrs, CT, United States.
- 3. NOAA, Oak Ridge, TN, United States.

Body: Complex dynamics of the stable planetary boundary layer (PBL), such as the effects of density currents, intermittent turbulence, surface-layer decoupling, internal gravity waves, cold air pooling, and katabatic flows affect plume transport and diffusion. A better understanding of these effects is needed for nighttime transport model development. The JORNADA (Joint Observational Research on Nocturnal Atmospheric Dispersion of Aerosols) field campaign, conducted in the New Mexico desert during April 2005, sought to address some of these issues The JORNADA data set includes simultaneous micrometeorological measurements of the boundary layer structure, turbulence, and wave activity along with continuous lidar measurement of aerosol plume releases. What makes JORNADA unique is the real-time monitoring of an elevated plume with a lidar. The quantification of plume meander will be presented in this paper. The application of these techniques to the JORNADA data allows for a more complete understanding of the nocturnal boundary layer (NBL). We will present an in-depth analysis of lidar measurements of plume meander and dispersion and their relationship to the complexities of NBL structure.

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An automatic Planetary Boundary Layer height retrieval method with compact EZ backscattering Lidar in the frame of ICOS campaign

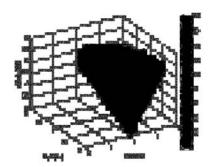
S. Loaec; 1; S. Lolli; 1; I. sauvage; 1; M. Boquet; 1; I. Xueref-Remy; 2;

- 1. Leosphere, Paris, France.
- 2. LSCE, Paris, France.

Body: Bigger strongly urbanized cities in the world are often exposed to atmospheric pollution events. To understand the chemical and physical processes that are taking place in these areas it is necessary to describe correctly the Planetary Boundary Layer (PBL) dynamics and the PBL height evolution. For these proposals, a compact and rugged eye safe UV Lidar, the EZLIDAR™, was developed together by CEA/LMD and LEOSPHERE (France) to study and investigate structural and optical properties of clouds and aerosols and PBL time evolution. EZLIDAR™ has been validated by different remote and in-situ instruments as MPL Type-4 Lidar manufactured by NASA at ARM/SGP site or the LNA (Lidar Nuage Aerosol) at the Laboratoire de Metereologie Dynamique LMD (France) and during several intercomparison campaigns. EZLIDAR™ algorithm retrieves automatically the PBL height in real-time. The method is based on the detection of the slope of the signal linked to a sharp change in concentration of the aerosols. Once detected, the different layers are filtered on a 15mn sample and classified between nocturnal, convective or residual layer, depending on the time and date. This method has been validated against those retrieved by the algorithm STRAT from data acquired at IPSL, France, showing 95% of correlation. In this paper are presented the results of the intercomparison campaign that took place in Orleans, France and Mace Head, Ireland in the framework of ICOS (Integrated Carbon Observation System) project, where the EZ Lidar™ worked under all weather conditions, clear sky, fog, low clouds, during the whole month of October 2008. Moreover, thanks to its 3D scanning capability, the EZLIDAR was able to provide the variability of the PBL height around the site, enabling the scientists to estimate the flux intensities that play a key role in the radiative transfer budget and in the atmospheric pollutants dispersion.

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Active methods to measure multilayer Planetary Boundary Layer Dynamics

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1. City College of New York, New York, NY, United States.

Body: The need to characterize in a robust way Planetary Boundary Layer (PBL) Heights is crucial as in air quality forcast and transport models. Incorrect determination of PBL heights can severely distort the surface level PM2.5 predictions crucial in determining whether New York City is in compliance. It has been amply demonstrated that lidar systems have repeatedly proven to be valuable tools in the study of the process of PBL. Therefore, it is useful to explore automated robust methods for PBL height retrieval. This presentation will focus on the determination of PBL height by assessing a variety of statistical and signal processing methods to assess PBL heights and compare tyhem against meteorologically based estimates of the Mixing layer. In particular, we find that in general, wavelet based techniques are more robust then edge detection filters and are most useful in obtaining multiple layers in low SNR cases. We also find that edge detection methods are very sensitive to noise and work well only under high SNR cases. Applications of these signal processing methods are applied to vertical resolving data from Calipso, Vaisala Ceilometer and Micropulse Lidar and compared against meteorological models from ARL as well as radiosondes. Particular emphasis on the Urban Heat Island effects will be presented.

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Lidar Approach in Estimating Particulate Mass Emissions from a Poultry Production Facility

P. A. Lewandowski; 1; W. E. Eichinger; 1; J. H. Prueger; 2; J. Hatfield; 2;

- 1. IIHR Hydroscience & Engineering, University of Iowa, Iowa City, IA, United States.
- 2. The National Soil Tilth Laboratory, Ames, IA, United States.

Body:

The current conventional particulate and mass emission measurements from livestock facilities rely primarily on point indoor/outdoor measurements. These measurements combined with assumed outflow rates from a building lead to emission rates and emission factors from the building. This approach, well established in the literature, poses accuracy and representation issues.

To overcome the limitations of point measurement emission estimates, a new remote sensing approach is proposed. A scanning elastic lidar was used to estimate the spatially resolved extinction coefficient associated with particulates originating from a poultry production building. Particulate size distribution and wind co-measurements were combined with the lidar extinction coefficient data to estimate particulate mass fluxes and the emission factor from the building. The particulate size distribution was measured continuously since the size distribution changes significantly during the day. Assumptions of constant size distributions may result in errors of a factor of two in derived quantities.

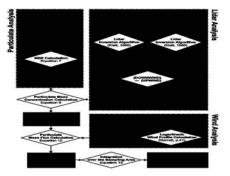
The data analysis from the study showed that the average particulate mass emission value from the poultry production building was 0.13±0.04 g/s (460±150 g/h) and the respective emission factor was 3.0±1.0 g/h AU (per animal unit, 500 kg live weight). The lidar estimated values are lower than the values found in the literature from point measurement studies.

The study demonstrates a new innovative method in measuring emissions using scanning lidar technique. As presented in the study, the method can successfully address the need for a better tool for emission measurements in agricultural applications. The outlined measurement approach can be also applied, with careful considerations, to any non-point particulate emissions measurement needs in industry or in urban environment.

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Lidar, particle sizer and wind anemometer data processing flowchart leading to the particulate mass emission estimates

Automated Raman lidar for day and night operational observation of water vapor for meteorological applications

T. Dinoev; 1; Y. Arshinov; 2; S. Bobrovnikov; 2; I. Serikov; 3; B. Calpini; 4; P. Ristori; 5; H. van den Bergh; 1; V. Simeonov; 1;

- 1. EPFL, Lausanne, Switzerland.
- 2. IAO, Tomsk, Russian Federation.
- 3. MPIMet, Hamburg, Germany.
- 4. MeteoSwiss, Payerne, Switzerland.
- 5. CITEFA, Buenos Aires, Argentina.

Body: We will present the design and results obtained by a fully automated, water vapor /aerosol lidar developed for operational use in the swiss meteorological office. The lidar supplies water vapor mixing ratio using vibrational Raman scattering, and aerosol extinction and backscattered coefficients at 355 nm derived from pure rotational Raman signals. The operational range of the lidar is 50-12000 m (nighttime) and 50-5000 m (daytime) with time resolution of 10-30 min. The spatial resolution varies with height from 15 to 150 m in order to maintain the maximum measurement error of 10%. The system is designed to provide long-term water vapor and aerosol database. The lidar is a part of the EARLINET ASOS network. It is in regular operation since September 2008at the main aerological station of Meteoswiss- Payerne. Upgrade with a temperature channel is ongoing and it will be operational in 2010.

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Very high spatial and temporal lidar for moisture and temperature monitoring

F. Martin; ¹; I. Serikov; ²; P. Ristori; ³; C. W. Higgins; ¹; M. B. Parlange; ¹; V. Simeonov; ¹;

- 1. EPFL, Lausanne, Switzerland.
- 2. MPIMet, Hamburg, Germany.
- 3. CITEFA, Buenos Aires, Argentina.

Body: A new generation solar-blind lidar system with an operational range 10-500 m and high spatial (1.5-6 m) and temporal (x1s) resolutions for simultaneous humidity and temperature, measurements in the lower atmosphere will be presented. To maintain the measurement accuracy while operating with fixed spatial and temporal resolution, the receiver is designed to provide lower than 10 dynamic range of the signals within the distance operational range of the lidar. The lidar has 360° azimuth and 240° scanning ability. The instrument has been used in two field campaigns to study the structure of the lower atmosphere over complex terrains. Selected results from these campaigns will be presented.

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Aerosol remote sensing over land using AATSR

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Body: The Advanced Along Track Scanning Radiometer (AATSR) flying on ENVISAT has been providing information on aerosol properties over widely different areas across the globe, with different land surface properties and different aerosol composition and concentrations. To this end, the single and dual view algorithms have been developed for application over ocean and land, respectively, and the evaluation of the aerosol properties retrieved using these algorithms, by comparison with AERONET data and results from other satellites, in particular MODIS, is being used to continuously improve them. Applications include the retrieval of forest fires smoke plumes over different areas in the world, to very clean areas in Finland, moderately polluted areas in Europe and highly polluted areas over China. The algorithms use look-up tables (LUTs) for different aerosol types and the algorithm has been set up to choose the most likely mixing ratio of two prescribed aerosol types (based on a priori information) using the information on the spectral dependence of the radiations measured at three or four wavelengths, in addition to aerosol optical density and Ångström parameter. Selected results will be presented.

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Diurnal variation of column NO₂ observed from space

<u>L. C. Valin;</u> ¹; A. R. Russell; ¹; R. C. Hudman; ¹; A. K. Mebust; ¹; R. C. Cohen; ¹;

1. College of Chemistry, University of California, Berkeley, Berkeley, CA, United States.

Body: NO₂ vertical column density, accessible from satellite-based UV-VIS observations on board NASA AURA satellite by the Ozone Monitoring Instrument (OMI), evolves in space and time due to emissions, transport and also chemical reaction with hydroxyl radical (OH). Since OH, and thus NO₂ removal rate, is greatest from 10 AM – 2 PM LST, the nominal OMI observation time of ~ 1:15 PM LST is arguably most representative of local-scale reactive nitrogen emissions. We assess the stability of column NO₂ from 12:45 to 1:15 PM LST over the Four Corners Power Plant using long-term averages of OMI observations whose temporal coverage is due to the wide swath angle of OMI and the 16-day orbital repeat pattern of AURA. We evaluate effects of systematic uncertainties and bias in the retrieval on the inferred time variation and use the WRF-CHEM chemical transport model at 4x4 km² grid resolution to compare predictions of meteorological and chemical variability with those observed.

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Validation of GOME-2 UV radiance

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- 2. Graduate University of Chinese Academy of Sciences, Beijing, China.
- 3. Goddard Earth Sciences and Technology Center, University of Maryland, Baltimore, MD, United States.
- 4. Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, United States.

Body: Global Ozone Monitoring Experiment-2(GOME-2) backscattered radiance was validated using a simulation aproach. Normalized GOME-2 radiance was simulated with a Vector Linearized Discrete Ordinate Radiative Transfer(VLIDORT) model and zonal mean Microwave Limb Sounder(MLS) ozone profiles in UV1(280-317nm) and UV2(310-340nm). Measured radiance was normalized by solar mean reference spectrum derived by GOME-2. Comparison of measurements and simulation shows a strong high frequency wavelength dependent bias in both channels and large offset between UV1a and UV1b, the formal might be related to residual ring effect and some atomic emission lines in shorter waves. Large offset in UV1b reveals a polarization correction problem because large angle dependent biases was found in this band. Latitudinal dependence is relatively weak except for high latitudes. In the overlap region of uv1b and uv2b (i.e. 310-316nm), UV2b shows weaker latitude and angle dependency than uv1b. Since we are using climatological ozone below 200 hPa, some of latitudinal dependent biases might be related to the imperfect knowledge of tropospheric ozone. Angle or X-track asymmetry increases and become significant at high latitude conditions especially for the first several X-positions. Our further work is to remove ring effect by fitting a ring spectral and correct all these bias to improve trace gas retrieval.

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Stochastic Modeling of Water Vapor in the Climate System

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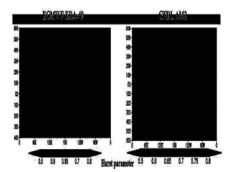
Body: The IPCC2007 assessment highlights one important question of "what the relative contribution of large-scale advective processes (in which confidence in GCMs' representation is high) is compared with microphysical processes (in which confidence is much lower) for determining the distribution and variation in water vapour." Here we present a data-driven, stochastic analysis-based framework to quantify subgrid-scale unresolved moistening processes (chiefly convective moistening) at a given isentropic surface (315K) and apply this framework to both ECMWF ERA-40 reanalysis data and the GFDL AM2 model output.

Based on ECMWF ERA-40 six-hourly data and the Advection-Diffusion-Condensation (ADC) model, we firstly estimate the subgrid convective moistening, which is vital to the water vapor distribution but not resolved explicitly in the model used for ECMWF ERA-40. We then have devised a stochastic modeling approach to parameterize the convective moistening in the ADC model. Convective moistening is represented by a multiplicative correlated (i.e. "colored") noisy process, in terms of a fractional Brownian motion. By means of optimization, parameters (Hurst parameter and noise intensity) are obtained for each individual grid boxes. Hurst parameter indicates the relative importance of large-scale process versus subgrid convective moistening. A Hurst parameter of 0.5 suggests the dominant control of subgrid convective moistening on the evolution of water vapor of the entire grid box while a Hurst parameter of 1.0 suggests nearly total control of large-scale dynamics on the water vapor variation over that grid box. Based on the ADC model and the stochastic representation of the convective moistening, an idealized model for water vapor evolution is then developed and validated by predicting the evolution of specific humidity based on ERA-40 wind fields alone. Compared with the ERA-40 reanalysis humidity, results suggest that the framework can capture various features of observed water vapor distributions and evolutions.

We also apply the same analysis to the 6-hourly water vapor data simulated by the GFDL AM2 model forced by observed SST over 2003-2005. The Hurst parameters derived from the AM2 humidity fields resemble those from ECMWF to a reasonable degree over the extra-tropics. In the tropics the Hurst parameters from the AM2 model is overwhelmingly larger than those from the ECMWF ERA-40 with distinctive spatial patterns. This indicates insufficient convective moistening at this level in the AM2 model compared to the ECMWF ERA-40 reanalysis. Another noticeable feature of AM2 result is that, over the margin of Tibet plateau and North Australia, the Hurst parameters are less than 0.5. The implication this feature is then further explored.

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Water vapor and OLR feedbacks over the recent ENSO cycle

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- 1. Atmospheric Sciences, University of Arizona, Tucson, AZ, United States.
- 2. Jet Propulsion Laboratory, Pasadena, CA, United States.

Body: The El-Nino-Southern Oscillation (ENSO) dominates low latitude climate variability. The ENSO cycle is tied to changes in the patterns and intensity of moist convection and precipitation. As such, water vapor observations across the tropics are critical to understanding and forecasting ENSO.

We report on evidence of a substantial negative feedback between the 2007 El Nino and 2008 La Nina that may be related to why 2008 was a relatively cold year. We focus on this period because free tropospheric vertical water distribution from the COSMIC GPS occultation mission was available to study the low latitude water cycle.

In comparing these two particular realizations of the ENSO phases, we find that the mean free tropospheric water vapor between 30N and 30S was higher during the warm phase by about 3.3%/oC change in sea surface temperature (SST) which is less than the Clausius Clapeyron. Furthermore, the median of the low latitude free tropospheric water vapor is actually slightly smaller in the warm phase than that in the cold phase.

There are indications of two SST thresholds. The zonally averaged SST must be at least 26oC in the cold phase for the positive feedback associated with deep convection to occur in the warm phase. The oceanic thermostat is evident where zonal mean SSTs do not exceed a maximum temperature limit of about 28.5oC indicating some combination of energy input into the oceans and transport of heat away from these latitudes is limiting the low latitude ocean temperatures, at least for this short period of observations.

Based on changes in SST between the two ENSO phases, the observations can be divided into two groups. For latitudes where the warm phase temperatures were greater than the cold phase by 0.4oC or less, the changes in outgoing longwave radiation (OLR) increase at a rate of 9.5 W/m2 per oC change in SST, substantially larger than the 4 W/m2 per oC of Stephan Boltzmann, indicating a negative feedback. A smaller group of latitudes where the SST difference was more than 0.4oC show clear signs of positive feedback with significantly larger free tropospheric water vapor amounts during the warm phase and an overall decrease in OLR with increasing SST. The feedback over the low latitudes is negative overall as it must be for ENSO to be an oscillation. This behavior may be providing clues about how clouds will respond in a warmer world.

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Investigating the Water Vapor Component of the Greenhouse Effect from the Atmospheric InfraRed Sounder (AIRS)

- A. Gambacorta; 1; C. Barnet; 2; F. Sun; 1; M. Goldberg; 2;
- 1. remote sensing, NOAA/PSGS, Camp Springs, MD, United States.
- 2. remote sensing, NOAA/NESDIS, Camp Springs, MD, United States.

Body: We investigate the water vapor component of the greenhouse effect in the tropical region using data from the Atmospheric InfraRed Sounder (AIRS). Differently from previous studies who have relayed on the assumption of constant lapse rate and performed coarse layer or total column sensitivity analysis, we resort to AIRS high vertical resolution to measure the greenhouse effect sensitivity to water vapor along the vertical column. We employ a "partial radiative perturbation" methodology and discriminate between two different dynamic regimes, convective and non-convective. This analysis provides useful insights on the occurrence and strength of the water vapor greenhouse effect and its sensitivity to spatial variations of surface temperature. By comparison with the clear-sky computation conducted in previous works, we attempt to confine an estimate for the cloud contribution to the greenhouse effect. Our results compare well with the current literature, falling in the upper range of the existing global circulation model estimates.

We value the results of this analysis as a useful reference to help discriminate among model simulations and improve our capability to make predictions about the future of our climate.

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Consistent Differences in Climate Feedbacks between Coupled and Atmospheric GCMs

K. M. Shell; 1;

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Body: Climate sensitivity is generally studied using two different types of models. Atmosphere-ocean general circulation models (AOGCMs, or coupled GCMs) include interactive ocean dynamics and detailed heat uptake, but they are computationally expensive to run. Atmospheric GCMs (AGCMs) with mixed-layer oceans cannot fully simulate the ocean's response to and influence on atmospheric changes. However, AGCMs require much less computer time and thus are often used to quantify and understand climate feedbacks and climate sensitivity. This work compares physical climate feedbacks (water vapor, lapse rate, surface albedo, and clouds) between coupled GCMs and atmospheric GCMs with mixed layer oceans from the World Climate Research Programme's (WCRP's) Coupled Model Intercomparison Project phase 3 (CMIP3) multi-model dataset. We use the new "radiative kernel" technique to calculate feedbacks. This technique decomposes each feedback into two parts: 1) the radiative effect of climate variable changes and 2) the response of these feedback variables to an imposed forcing. Since kernels are similar for different models, a single kernel can be used to estimate feedbacks for any model simulation.

We find differences in feedbacks between coupled models and their corresponding atmospheric models with mixed layers. The global-average water vapor feedback is consistently stronger (more positive) in coupled models versus atmospheric models with mixed layer oceans. The global-average lapse rate feedbacks are also consistently stronger (more negative). Zonal-average water vapor feedbacks indicate that coupled models have a larger water vapor increase in the tropics compared to atmospheric models. The differences in lapse rate feedbacks extend over the tropics and midlatitudes and correspond to a larger tropic to global surface temperature change ratio. The northern high latitudes warm faster than the southern high latitudes in coupled models; we see this effect in, for example, the albedo feedback. The feedback magnitudes depend on the particular forcing scenario (e.g., SRESa1b or 1pctto2x). We examine these feedbacks as a function of time to determine if the differences between scenarios decrease as the models approach equilibrium (indicating that the feedback differences are transient) or if the forcing and model's configuration affect the equilibrium feedbacks. These results indicate that care should be taken when climate feedbacks and sensitivities from AGCMs are used to provide information about AOGCM feedbacks and sensitivities.

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Observationally Based Estimates of Climate Feedbacks

N. D. Gordon; ¹; P. Forster; ¹;

1. University of Leeds, Leeds, United Kingdom.

Body: Our study focuses hopes to provide constraints to feedbacks in our climate system from assimilation of observations and a radiative transfer scheme. To make accurate projections of future climate, we need to better understand the effect of major physical processes on the climate, especially the transfer of radiation in the atmosphere. We focus our study on the variability of water vapor in the atmosphere, derived from satellite observations, and calculate the effect that each of these time series of water vapor has on the output of the Edwards-Slingo radiation scheme. By comparing the observed surface temperature perturbation with the simulated perturbation in net top of atmosphere radiation, we can build a linear relationship. Using linear feedback analysis, we try to isolate the effect that each of these elements has on the climate system. Additionally, we compare our results to those obtained using a kernel method, and highlight the differences. By better understanding the effect that variability in elements of the climate system has on the net top of atmosphere radiation, we can better constrain the climate sensitivity.

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Mechanisms for the weakening of tropical circulation

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- 2. Department of Atmospheric Sciences, National Taiwan University, Taipei, Taiwan.
- 3. Department of Earth Sciences, National Taiwan Normal University, Taipei, Taiwan.

Body: As global warming becomes dominant, many climate changes are starting to show. One of the well-known climate changes is a weakening of tropical circulation, which could affect rainfall amount and intensity both in regional and global scales. The global-mean water vapor increases at a rate of 7.5%/K, roughly following the Clausius-Clapeyron thermal expansion at a constant relative humidity, while the global-mean precipitation increases at a rate of only around 2%/K. The fact that the precipitation increases more slowly than the water vapor implies a weakening of tropical circulation (Soden and Held 2006; Vecchi and Soden 2007). In our study, however, we will demonstrate this simple relationship between the global-mean precipitation and water vapor cannot guarantee a weakening of tropical circulation. In fact, tropical circulation could strengthen even though the global-mean precipitation increases much more slowly than the global-mean water vapor. We propose here a new mechanism for this robust climate change in the strength of tropical circulation. Based on a more precise column-integrated moisture budget, atmospheric stability associated with convection depth is the main mechanism. Convection tends to extend higher in a warmer climate, due to an uplifting of the tropopause. The higher the convection, the more stable the atmosphere. This leads to a weakening of tropical circulation.

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An Observed Tropical Oceanic Radiative-Convective Cloud Feedback

M. D. Lebsock; ¹; G. L. Stephens; ¹; C. D. Kummerow; ¹;

1. Colorado State University, Fort Collins, CO, United States.

Body: Anomalies of precipitation, cloud, thermodynamic, and radiation variables are analyzed on the large spatial scale defined by the tropical oceans. In particular, relationships between the mean tropical oceanic precipitation anomaly and radiative anomalies are examined. It is found that tropical mean precipitation is well correlated with cloud properties and radiative fields. In particular, the tropical mean precipitation anomaly is positively correlated with the top of the atmosphere reflected shortwave anomaly and negatively correlated with the emitted longwave anomaly. The tropical mean relationships are found to primarily result from a coherent oscillation of precipitation and the area of high-level cloudiness. The correlations manifest themselves radiatively as a modest decrease in net downwelling radiation at the top of the atmosphere, and a redistribution of energy from the surface to the atmosphere through reduced solar radiation to the surface and decreased longwave emission to space. Integrated over the tropical oceanic domain, the anomalous atmospheric column radiative heating is found to be about 10% of the magnitude of the anomalous latent heating. The temporal signature of the radiative heating is observed in the column mean temperature that indicates a coherent phase-lagged oscillation between atmospheric stability and convection. These relationships are identified as a radiative-convective cloud feedback that is observed on intra-seasonal timescales in the tropical atmosphere.

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Interrelations of AIRS/AMSU-derived anomalies and trends of temperature, water vapor, clouds and OLR *G. I. Molnar*; ^{1, 2}; *J. Susskind*; ²;

- 1. GEST, UMBC, Baltimore, MD, United States.
- 2. Laboratory for Atmospheres, NASA GSFC, Greenbelt, MD, United States.

Body: The AIRS/AMSU instrument is currently the best space-based tool to simultaneously monitor the vertical distribution of key climatically important atmospheric parameters as well as surface properties and has been providing high quality data for more than 7 years. Currently, general circulation models (GCMs) are our best tools for longerterm climate change predictions. However, there are large uncertainties in their climate feedback strengths, in particular those related to atmospheric moisture. Here, we illustrate that even relatively shorter-term observed climate parameter variabilities, measured with high enough quality, could provide useful insights about climate feedbacks and may serve as constraints on theoretical and GCM-computed feedback mechanisms. Anomalies relative to the mean seasonal cycle of 7 years worth of data are indicative of various climate forcings and feedbacks, especially in periods containing strong El Niño/La Niña oscillations. This work is based on publicly available Version 5 Level3 AIRS analysis results produced at the GODDARD/DISC. Following the presentation of AIRS-anomaly validations, using available independent satellite data analysis results, we continue with an assessment of interrelationships of AIRSobserved anomalies of various climate parameters at different spatial scales. We also present AIRS-retrievals-based global, regional and 1x1 degree grid-scale "trend"-analyses of important atmospheric parameters for this 7+ year period. Note that here "trend" simply means the linear fit to the anomaly time series of various parameters at the above-mentioned spatial scales. Correlations among the anomaly timeseries and relevant Hovmoller diagrams reveal how selected climate feedbacks operate on various spatial scales. We find especially meaningful correlations over the El Niño/La Niña affected regions. These observed correlations may also provide constraints on model implementations/manifestations of climate feedback processes and illustrate the usefulness of continuing the high quality AIRS-based climate variability measurements.

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Research activity of the greenhouse gas measurements using optical remote sensing in Japan (*Invited*) *K. Asai*: ¹:

1. Tohoku Institute of Technology, Sendai, Japan.

Body: Japan might be one of the most active countries dedicating themselves to studying the greenhouse gas (GHG) measurements using optical remote sensing not only on the ground but also from space. There are two reasons; one of them ascends to the Kyoto Protocol, agreed in December 1997 in Kyoto, an ancient city of Japan until 19th centuries, was designed to address the international response to serious climate change due to greenhouse gases. The other reason is due to a revision of the Basic Environment Law of Japan in order to meet the Kyoto Protocol in 1998. The State makes efforts to ensure international collaboration so as to effectively promote the monitoring, observation and measurement of the environmental situation with regard to global warming.

Main activities are listed in a Table1. They are divided into two categories, i.e. the Greenhouse gases Observing SATellite (GOSAT), launched on Jan.23, 2009 and active remote sensing using lidar technology. In case of GOSAT, an initial analysis of carbon dioxide and methane concentrations was obtained for clear-sky scenes over land. In the future, after further calibration and validation of the data, observation data and corresponding analyzed products will be made available.

On the other hand, studies of the laser remote sensing for measuring GHG have been actively carrying out to achieve reliable data with a higher accuracy at wavelengths of 1.6micron meter (Tokyo Metropolitan University, JAXA, Mitsubishi Electric Co.) and 2 micron meter (National Institute of Information and Communications Technology). As well-known, one of the most interests regarding atmospheric CO2 measurements is that carbon dioxide molecule measured are due to anthropological emission from fossil fuel burning or due to natural one from forest fires etc. We proposed a newly advanced CO2/CO DIAL using a hybrid of pulsed Tm,Ho:YLF and pulsed OPO pumped by it for better understanding them. Now, our effort is directed to find out the most suitable wavelength pairs to be selected.

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"Greenhouse gases Observing SATellite (GOSAT)", launched on Jan.23, 2009							
Sensor: Fourier Transform Spectrometer + Cloud Aerosol Imager							
	Band 1	Band 2	Band 3	Band 4			
Spectral coverage [µm]	0.758-0.775	1.56-1.72	1.92-2.08	5.56-14.3			
Target species	O2	CO2, CH4	CO2, H2O	CO2, CH4			
FOV/Footprint	15.8mrad / 10.5km						
Active remote sensing using laser technologies DIAL :DIfferential Absorption Lidar, LAS :Laser Absorption Sensor							

Insititution	NiCT	тми	JAXA&MELCO	ТІТ	
Target species	CO2	CO2	CO2	CO2,CO	
System	DIAL	DIAL	LAS	DIAL	
Laser transmitter	Pulsed Tm,Ho:YLF	Pulsed OPO	cw Laser Diode	Tm,Ho:YLF@C O2 Pulsed OPO@CO	
On-line(nm)	2050.967	1572.0178	1572.992	TBD	
Off-line(nm)	2051.250	1572.150	1573.193	TBD	
Accuracy/Stabili ty	0.7% (5min.)	<2%	4ppm (32sec).		
NiCT:National Institute of Information and Communications Technology, TMU:Tokyo Metropolitan University, JAXA:Japan Aerospace Exploration Agency, MELCO:Mitsubishi Electric C0.Ltd, TIT:Tohoku Institute of Technology					

Airborne Validation of Active CO₂ LAS Measurements

<u>E. V. Browell;</u> ¹; J. Dobler; ²; S. Kooi; ¹; Y. Choi; ¹; F. Harrison; ¹; B. Moore; ³; T. Zaccheo; ⁴;

- 1. NASA Langley Research Ctr, Hampton, VA, United States.
- 2. ITT Corporation, Fort Wayne, IN, United States.
- 3. Climate Central, Princeton, NJ, United States.
- 4. AER, Inc., Lexington, MA, United States.

Body: A unique, multi-frequency, single-beam, laser absorption spectrometer (LAS) that operates at 1.57 μm has been developed for a future space-based mission to determine the global distribution of sources and sinks of atmospheric carbon dioxide (CO₂). A prototype of the space-based LAS system was developed by ITT, and it has been successfully flight tested in seven airborne campaigns conducted in different geographic regions over the last four years. Flight tests were conducted over Oklahoma, Michigan, New Hampshire, and Virginia under a wide range of atmospheric conditions. Remote LAS measurements were compared to high-quality in situ measurements obtained from instrumentation on the same aircraft on spirals under the ground track of the LAS. LAS flights were conducted over a wide range of land and water reflectances and in the presence of scattered clouds. An extensive data set of CO₂ measurements has been obtained for evaluating the LAS performance. These LAS test flights resulted in the first demonstration of high-precision, high-accuracy, remote laser measurements of CO₂ from an airborne platform. The LAS CO₂ column measurements were found to have a precision for a 10-s horizontal average (~1 km) of better than 1 ppm of ${\rm CO}_2$ over land and 1.3 ppm over water. Absolute comparisons of ${\rm CO}_2$ remote and in situ measurements showed agreement on average to better than 0.5 ppm of CO₂ with a standard deviation of the agreement to better than 2 ppm of CO₂. In addition, results of recent coordinated aircraft flight tests of different CO₂ LAS systems and different in situ CO₂ measurement systems over Oklahoma and Virginia in July-August 2009 are also discussed in this paper.

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Pulsed Airborne Lidar measurements of Atmospheric CO2 Column Absorption and Line Shapes from 3-13 km

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- 3. Sigma Space Inc, Greenbelt, MD, United States.

Body: We have developed a lidar technique for measuring the tropospheric CO2 concentrations as a candidate for NASA's planned ASCENDS mission. Our technique uses two pulsed laser transmitters allowing simultaneous measurement of a CO2 absorption line in the 1570 nm band, O2 extinction in the Oxygen A-band and surface height and backscatter. The lidar measures the energy and time of flight of the laser echoes reflected from the atmosphere and surface. The lasers are stepped in wavelength across the CO2 line and an O2 line region during the measurement. The receiver uses a telescope and photon counting detectors, and measures the background light and energies of the laser echoes from the surface along with scattering from any aerosols in the path. The gas extinction and column densities for the CO2 and O2 gases are estimated from the ratio of the on- and off- line signals via the DIAL technique. Time gating is used to isolate the laser echo signals from the surface, and to reject laser photons scattered in the atmosphere.

We have developed an airborne lidar to demonstrate the CO2 measurement from the NASA Glenn Lear-25 aircraft. The airborne lidar steps the pulsed laser's wavelength across a selected CO2 line with 20 steps per scan. The line scan rate is 450 Hz, laser pulse energy is 25 uJ and laser pulse widths are 1 usec. The time resolved laser backscatter is collected by a 20 cm telescope, detected by a photomultiplier and is recorded by a photon counting system. We made initial airborne measurements on flights during October and December 2008. Laser backscatter and absorption measurements were made over a variety of land and water surfaces and through thin and broken clouds. Atmospheric CO2 column measurements using the 1571.4, 1572.02 and 1572.33 nm CO2 lines. Two flights were made above the DOE SGP ARM site at altitudes from 3-8 km. These flights were coordinated with DOE investigators who flew an in-situ CO2 sensor on a Cessna aircraft under the path. The increasing CO2 line absorptions with altitudes were evident and comparison with in-situ measurements showed agreements to 6 ppm.

This spring we improved the aircraft's nadir window. During July and August 2009 we made 9 additional 2 hour long flights and measured the atmospheric CO2 absorption and line shapes using the 1572.33 nm CO2 line. Measurements were made at stepped altitudes from 3-13 km over a variety of surface types in Nebraska, Illinois, the SGP ARM site, and near and over the Chesapeake Bay in North Carolina and Virginia. Strong laser signals and clear line shapes were observed at all altitudes, and some measurements were made through thin clouds. The flights over the ARM site were underflown with in-situ measurements made from the DOE Cessna. The Oklahoma and east coast flights were coordinated with a LaRC/ITT CO2 lidar on the LaRC UC-12 aircraft, a LaRC in-situ CO2 sensor, and the Oklahoma flights also included a JPL CO2 lidar on a Twin Otter aircraft. Ed Browell and Gary Spiers led the LaRC and JPL teams. More details of the flights, measurements and their analysis will be described in the presentation.

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Isotopic Tracers to Identify Far-traveled Pollutant and Mineral Aerosols in Northern California (*Invited*)

D. J. Depaolo; 1, 2; J. N. Christensen; 2; S. A. Ewing; 3; S. S. Cliff; 4; S. T. Brown; 2, 1; R. A. VanCuren; 5;

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- 2. Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA, United States.
- 3. Marine St Science Center, US Geological Survey, Boulder, CO, United States.
- 4. Applied Science, University of California Davis, Davis, CA, United States.
- 5. Research Division, California Air Resources Board, Sacramento, CA, United States.

Body: Mineral dust and pollutant aerosols can be lofted into the atmosphere and transported 1000s of kilometers, facilitating intercontinental communication of soil components, biological material (bacteria, viruses) and anthropogenic particulates. Far-traveled aerosols also affect air quality, atmospheric radiation balance and cloud formation. Understanding the sources of aerosols, and how they evolve with climate change, land use changes, and emerging industrial activity, is important for assessing air quality and climate processes in California. A particular concern for California is trans-Pacific transport of mineral aerosols from Asian deserts, and the possibility that industrial and other pollutants accompany them. The geographic sources of mineral and pollutant aerosols can in many cases be determined from their isotopic composition, using for example some combination of elements such as Pb, Sr, Nd, Hf, Zn, N, S, C, O, U, B, and Li. With systematic sample collection and analysis, isotopes can provide quantification of the changing proportions of local versus distant sources. Where the far-traveled components can be identified, comparisons can be made to meteorological data to better understand the factors controlling the efficiency of long-range transport.

With heavy dust storms, such as those that arise in the Sahel/Sahara or the deserts of Asia, aerosols can be tracked in satellite imagery and other approaches may not be necessary. During more common periods of lesser aerosol loading, and where greater transport distances are involved, ground-based methods such as chemical analysis of a time-series of collected PM2.5 are needed to evaluate sources. Pollutants may or may not accompany mineral dust, and may be added along the transport path. Although chemical analysis is useful, relatively fast and inexpensive, more information, and in some cases more definitive conclusions, can be obtained by adding isotopic measurements. By combining multiple isotopic systems (e.g., Pb, Sr, Nd isotopes), the precision and sensitivity of geographic attribution is increased, and different aerosol components can be targeted (e.g., Pb for industrial particulates, Sr and Nd for mineral dust). Isotopes can also give information about aerosol alteration during transport.

As an illustration of these points we will present the results of a time series of isotopic (Pb and Sr), and chemical data for samples collected at several sites in California over the past 1.5 years. In this case the proportion of airborne Pb originating from Asia can be tracked accurately, and shown to vary seasonally and even weekly. The isotopic approach is sufficiently sensitive that the proportion of Asian Pb can be determined even close to urban areas where overall loading is high and local sources are strong. Sr isotopes provide other information, such as the effects of admixed marine aerosols, and based on samples collected in Asia, how materials from different Asian sources are mixed prior to crossing the Pacific.

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Summertime Ozone over California: A Model-Measurement Analysis across Scales

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1., Boulder, CO, United States.

Body: We use the global chemistry transport model MOZART-4 together with the regional-scale model WRF-Chem V3.1 to analyze the characteristic summertime contributions of ozone and ozone pre-cursors over California. Both models employ the same chemistry scheme and emissions allowing for a high level of synergy across model scales with the global model providing the boundary conditions for the regional simulations.

The focus in the analysis is on summer 2008 when the ARCTAS-CARB aircraft campaign, a joint program between NASA and the California Air Resources Board (CARB), took place. Measurements from this field campaign will be used together with in-situ observations from ground (U.S. EPA Air Quality Monitoring System) as well as satellite retrievals (e.g. Aura/OMI NO2 and HCHO, Aura/TES CO and O3, Terra/MOPITT CO) for evaluating the model simulations and support the analysis. We will examine the individual factors impacting ozone concentrations over the California region including contributions from anthropogenic and biogenic sources, wildfires and long-range transport.

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Oxygenated VOCs as indicators of regional-scale photochemistry: comparison of CMAQ model results with measurements from the UC Blodgett Forest Station, California

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- 2. US EPA, Durham, NC, United States.

Body: We present a study of a subset of the processes which influence ozone (O₃) and Secondary Organic Aerosol (SOA) on a regional scale. Using measurements taken at Blodgett Forest Research Station (BFRS) during the BEARPEX 2007 & 2009 campaigns, we examine the role of photochemistry within the canopy and the influences of anthropogenic emissions (ex. NO_x) in a region dominated by biogenic Volatile Organic Compound (VOC) emissions.

Measurements of formaldehyde (HCHO) and glyoxal (GL) are used as tracers for the generalized oxidation of VOC at BFRS. We investigate observed gradients in GL and HCHO with respect to rapid photochemistry within the canopy. The interaction of anthropogenic emissions (in this case the aged anthropogenic effluent from the Sacramento area) with biogenic emissions is examined. The Community Multiscale Air Quality (CMAQ) model is used both for direct comparison to measurement and as a means to generalize the processes under study here to Northern California as a region.

Finally, we employ ratios of compounds (ex. GL / HCHO, GL / MPAN, etc.) in both measurement and CMAQ to study plume evolution and the antecedent conditions of airmasses arriving at BFRS. These reduced-complexity datasets attenuate the influence of meteorology, allowing more complete analysis of measured data and enabling comparison to measurements from other campaigns or platforms and with models.

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Measurements of Greenhouse Gases around the Sacramento Area:

The Airborne Greenhouse Emissions Survey (AGES) Campaign

A. Karion; ^{1, 2}; M. L. Fischer; ³; J. C. Turnbull; ²; C. Sweeney; ^{1, 2}; I. C. Faloona; ⁴; N. Zagorac; ⁴; T. P. Guilderson; ⁵; S. Saripalli; ⁶; T. Sherwood; ⁶;

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- 2. ESRL, NOAA, Boulder, CO, United States.
- 3. Lawrence Berkeley Laboratory, Berkeley, CA, United States.
- 4. University of California, Davis, CA, United States.
- 5. Lawrence Livermore National Laboratory, Livermore, CA, United States.
- 6. Kalscott Engineering, Lawrence, KS, United States.

Body: The state of California is leading the United States by enacting legislation (AB-32) to reduce greenhouse gas emissions to 1990 levels by 2020. The success of reduction efforts can be gauged with accurate emissions inventories and potentially verified with atmospheric measurements of greenhouse gases (GHGs) over time. Measurements of multiple GHGs and associated trace gas species in a specific region also provide information on emissions ratios for source apportionment. We conducted the Airborne Greenhouse Emissions Survey (AGES) campaign to determine emissions signature ratios for the sources that exist in the San Francisco Bay and Sacramento Valley areas. Specifically, we attempt to determine the emissions signatures of sources that influence ongoing measurements made at a tall-tower measurement site near Walnut Grove, CA.

For two weeks in February and March of 2009, a Cessna 210 was flown throughout the Sacramento region, making continuous measurements of CO2, CH4, and CO while also sampling discrete flasks for a variety of additional tracers, including SF6, N2O, and 14C in CO2 (Δ 14CO2). Flight paths were planned using wind predictions for each day to maximize sampling of sources whose emissions would also be sampled contemporaneously by the instrumentation at the Walnut Grove tower (WGC), part of the ongoing California Greenhouse Gas Emissions Measurement (CALGEM) project between NOAA/ESRL's Carbon Cycle group and Lawrence Berkeley National Laboratory (LBNL). Flights were performed in two distinct patterns: 1) flying across a plume upwind and downwind of the Sacramento urban area, and 2) flying across the Sacramento-San Joaquin Delta from Richmond to Walnut Grove, a region consisting of natural wetlands as well as several power plants and refineries. Results show a variety of well-correlated mixing ratio signals downwind of Sacramento, documenting the urban signature emission ratios, while emissions ratios in the Delta region were more variable, likely due to the both natural and anthropogenic sources in that region. Periodic flask measurements of Δ 14CO2 provide additional insight regarding the partitioning of CO2 emissions due to fossil fuel (deficient in 14C) from those of biospheric sources. A strong correlation between fossil-fuel CO2 and CO was measured downwind of Sacramento, suggesting that the continuous measurements of CO can be used to estimate a continuous profile of fossil-fuel CO2 enhancement in this region.

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Aerosol Precursor Emissions, Secondary Aerosol Production, and Climate-Forcing Gas Exchange in the Midwestern United States

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1. Department of Civil and Environmental Engineering/School of Forest Resources and Environmental Science/Atmospheric Sciences Program , Michigan Technological University, Houghton, MI, United States.

Body: Aerosol precursors in the Midwest are generated from a myriad of sources including biogenic emissions of terpenes from the Ozarks region, anthropogenic emissions of volatile and semivolatile aliphatic and aromatic hydrocarbons from the St. Louis airshed, and agricultural emissions of ammonia (NH3), amines, and nitrogen oxides (NOx) from animal husbandry and cropping systems of the Midwest Corn Belt. The deciduous and coniferous forests of the Ozarks region are significant sources of isoprene, monoterpenes, and sesquiterpenes that are sensitive to rising CO2 levels and temperature and generate light-scattering, secondary organic aerosol (SOA). Application of nitrogen fertilizers stimulates emissions of ammonia (NH3), nitric oxide (NO), and nitrous oxide (N2O) from agricultural soils and crops. Nitric acid, generated through photooxidation of NO emissions from fossil fuel combustion in urban air and from soil emissions in agroecosystems, reacts rapidly with NH3 to generate light-scattering, secondary inorganic aerosol (SIA). The atmospheric lifetime of N2O is about 120 years, making the substance a potent greenhouse gas with a global warming potential of 290 for a time horizon of 20 years relative to CO2. Emissions of CO2, N2O, and SIA precursors from the Midwest Corn Belt and surrounding areas are likely to increase in the near future as pastureland and prairie is converted to grow corn and other biofuel crops to meet the demand for renewable fuels. Several large river systems transport nutrients from fertilized fields of the Midwest agroecosystem to the Gulf of Mexico where plankton growth is accelerated. Microbial decomposition of plankton detritus consumes oxygen and creates a hypoxic zone, which might be a significant source of N2O. The presentation will discuss gaps in our knowledge of the production of climate-forcing species in the Midwestern United States.

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Characterizing CH₄ and N₂O Fluxes from a Soybean/Corn Ecosystem in Minnesota

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- 2. Department of Soil, Water, and Climate, University of Minnesota, St Paul, MN, United States.
- 3. Agricultural Research Service, USDA, St Paul, MN, United States.
- 4. College of Applied Meteorology, Nanjing University of Information Science and Technology, Nanjing, Jiangsu, China.

 $\textbf{Body:} \ \text{In order to characterize the budgets of three major greenhouse gases (CO}_2, \ \text{CH}_4 \ \text{and N}_2\text{O}) \ \text{from cropland, we}$ conducted an experiment near Rosemount, Minnesota, in a landscape dominated by soybean and corn faming. The experiment was carried out at the plant, the ecosystem, and the regional scales. A steady-state flow-through chamber was used to measure the fluxes from the plants of soybean and corn. The gradient diffusion method was used to determine the fluxes at the ecosystem scale. Concentration measurements on a tall tower were used to drive a Lagrangian transport model to interoperate the surface fluxes at the regional scale. Measurements of CH₁ and N₂O at each scale were made using tunable diode laser spectroscopy.

The results to date are summarized as follows: 1) Corn plants were a small net sink of N₂O with an average uptake of 4×10^{-4} µmol m⁻² s⁻¹ mainly occurring at night. The N₂O flux of unfertilized soybean plants was below the instrument detection limit, and that of fertilized plants was a net source to the atmosphere at a rate of $5 \times 10^{-3} \, \mu mol \, m^{-2} \, s^{-1}$ with the emission mainly occurring at night. 2) Both the corn and soybean plants showed a slight uptake of CH_{Δ} during the night and release during the day. The daily average CH₄ flux was a small net sink for soybean (5×10⁻⁵ μmol m⁻² s⁻¹) and a small net source for corn (1×10 $^{-4}$ µmol m $^{-2}$ s $^{-1}$). 3) The soybean ecosystem was a source of N $_2$ O, with an emission rate of 1×10^{-4} µmol m⁻² s⁻¹ at night and 5×10^{-4} µmol m⁻² s⁻¹ during the day (The analysis of the ecosystem data for corn is under way). 4) The tall tower measurements indicate a strong source of CH₄ and N₂O at the regional scale. These results will be discussed in the context of a Lagrangian transport model, which is currently under development.

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Quantification of Terpenes by 1DGC-MS and 2DGC-TOF-MS

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1. Michigan Technological University, Houghton, MI, United States.

Body: Biogenic emissions are the primary source of volatile organic compounds in the global troposphere. Deciduous and coniferous forests are the principal emitters of a complex mixture of isoprene (C5H8), monoterpenes (C10H16), and sesquiterpenes (C15H24). Sesquiterpenes are readily oxidized in the atmosphere producing secondary organic aerosols (SOA) with 100% yields. The SOA are hydrophilic and scatter light, and thus, increase albedo and lead to a cooling effect. In addition, both monoterpene and sesquiterpene generated SOA are effective cloud condensation nuclei leading to an increase in the particle number concentration and to the formation of clouds that also increase albedo. To quantify the complex mixture of terpenes and their oxidation products requires development of on-line extraction and comprehensive two-dimensional gas chromatographic techniques. One objective of this work was to compare one-dimensional gas chromatography-mass spectrometry (1DGC-MS) and two-dimensional gas chromatography time-of-flight mass spectrometry (2DGC-TOFMS) for quantifying eight monoterpenes (alpha- and beta-pinene, limonene, 3-carene, linalool, terpinolene, myrcene and ocimene) and eight sesquiterpenes (beta-caryophyllene, humulene, alpha-cedrene, cis-nerolidol, trans-nerolidol, cedrol, camphene and farnesene) in air samples collected in Northern Michigan. Future research involves coupling thermal desorption and supercritical fluid extraction devices to a GC×2GC for routine quantification of the complex mixture of terpenes and their oxidation products in rural and urban air.

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Aerosol Production from the Great Lakes Surface

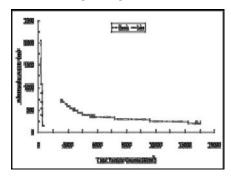
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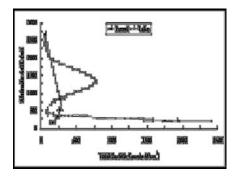
Body: It is well understood that oceans generate airborne particulate matter from mechanical processes such as sea spray and bubble bursting. These particles are primarily composed of salts and other nonvolatile inorganic material; however, the organic mass fraction can vary by location and the extent of biological activity. The size distributions of aerosols in these environments depend greatly on relative humidity with diameters ranging from typically several hundred nanometers to several micrometers. There has been much less discussion of particle formation from fresh water ecosystems, a hub for organic activity, and thus a more likely medium for organic aerosol production. We investigated particle formation over the Great Lakes during the summer of 2009 as a part of the Community Atmosphere-Biosphere Interactions Experiments (CABINEX) at the University of Michigan Biological Station (UMBS) in Pellston, MI. With a scanning mobility particle sizer (SMPS) aboard Purdue University's Airborne Laboratory for Atmospheric Research (ALAR) for size-distribution analysis of accumulation-mode aerosol, we conducted vertical profiles above Lake Michigan and the UMBS deciduous forest, and transects across the peninsula between Lakes Michigan and Huron to study particle formation, transport, and deposition. Preliminary results reveal a well-mixed troposphere above the forest with a mode ~0.1 μm, while in several cases, the total particle concentration over Lake Michigan is an order of magnitude greater than over the forest. There is a consistent bimodal distribution of particle sizes over Lake Michigan the lowest of which is centered at ~0.025 µm, suggesting the possibility of new particle formation. This mode is consistent with the presence of breaking waves on the lake's surface, and this mode and the vertical structure depend greatly on wind speed. We present here evidence for new particle production from breaking waves on fresh water lakes, and discuss the results, including those from detailed chemical analysis of the particulate matter.

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07/09 50 nm mode 07/16 25 nm mode



A Methylmercury Prediction Too For Surface Waters Across The Contiguous United States (*Invited*)

D. P. Krabbenhoft; 1; N. Booth; 1; M. Lutz; 1; M. N. Fienen; 1; T. Saltman; 2;

- 1. Wisconsin Water Science Center, U.S. Geological Survey, Middleton, WI, United States.
- 2. Office of Air and Radiation, U.S. Environmental Protection Agency, Washington, DC, United States.

Body: About 20 years ago, researchers at a few locations across the globe discovered high levels of mercury in fish from remote settings lacking any obvious mercury source. We now know that for most locations atmospheric deposition is the dominant mercury source, and that mercury methylation is the key process that translates low mercury loading rates into relatively high levels in top predators of aquatic food webs. Presently, almost all US states have advisories for elevated levels of mercury in sport fish, and as a result there is considerable public awareness and concern for this nearly ubiquitous contaminant issue. In some states, "statewide" advisories have been issued because elevated fish mercury levels are so common, or the state has no effective way to monitor thousands of lakes, reservoirs, wetlands, and streams. As such, resource managers and public health officials have limited options for informing the public on of where elevated mercury concentrations in sport fish are more likely to occur than others. This project provides, for the first time, a national map of predicted (modeled) methylmercury concentrations in surface waters, which is the most toxic and bioaccumulative form of mercury in the environment. The map is the result of over two decades of research that resulted in the formulation of conceptual models of the mercury methylation process, which is strongly governed by environmental conditions - specifically hydrologic landscapes and water quality. The resulting predictive map shows clear regional trends in the distribution of methylmercury concentrations in surface waters. East of the Mississippi, the Gulf and southeastern Atlantic coast, the northeast, the lower Mississippi valley, and Great Lakes area are predicted to have generally higher environmental methylmercury levels. Higher-elevation, well-drained areas of Appalachia are predicted to have relatively lower methylmercury abundance. Other than the prairie pothole region, in the western US incessant regional patterns are less clear. However, the full range of predicted methylmercury levels are predicted to occur in western US watersheds. Lastly, although this map is being presented at the continental US scale, the principles used to generate the modeled results can easily applied to data sets that represent a range of geographic scales.

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Mercury Isotopic Evidence for Contrasting Mercury Transport Pathways to Coastal versus Open Ocean Fisheries (

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- 2. Dept. of Environmental Health, Harvard School of Public Health, Boston, MA, United States.
- 3. Institute of Biogeochemistry and Pollutant Dynamics, ETH, Zurich, Switzerland.
- 4. Louisiana Universities Marine Consortium, Chauvin, LA, United States.
- 5. National Institute of Nutrition and Seafood Research, Bergen, Norway.

Body: Mercury stable isotopes provide a new method for tracing the sources and chemical transformations of Hg in the environment. In this study we used Hg isotopes to investigate Hg sources to coastal versus migratory open-ocean species of fish residing in the northern Gulf of Mexico (nGOM). We report Hg isotope ratios as δ202Hg (mass dependent fractionation relative to NIST 3133) and Δ 201Hg (mass independent fractionation of odd isotopes). In six coastal and two open ocean species (blackfin and yellowfin tuna), Hg isotopic compositions fell into two non-overlapping ranges. The tuna had significantly higher δ202Hg (0.1 to 0.7‰) and Δ 201Hg (1.0 to 2.2‰) than the coastal fish (δ202Hg = 0 to -1.0‰; Δ 201Hg = 0.4 to 0.5‰). The observations can be best explained by largely disconnected food webs with isotopically distinct MeHg sources. The ratio Δ 199Hg/ Δ 201Hg in nGOM fish is 1.30±0.10 which is consistent with laboratory studies of photochemical MeHg degradation and with ratios measured in freshwater fish (Bergquist and Blum, 2007). The magnitude of mass independent fractionation of Hg in the openocean fish suggests that this source of MeHg was subjected to extensive photodegradation (~50%) before entering the base of the open-ocean food web. Given the Mississippi River's large, productive footprint in the nGOM and the potential for exporting prey and MeHg to the adjacent oligotrophic GOM, the different MeHg sources are noteworthy and consistent with recent evidence in other systems of important open-ocean MeHg sources.

Bergquist, B. A. and Blum, J. D., 2007. Mass-dependent and -independent fractionation of Hg isotopes by photoreduction in aquatic systems. Science 318, 417-420.

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Evidence for the free troposphere as a source of atmospheric mercury measured in Reno, Nevada, U.S.A. *M. S. Gustin;* ¹; *S. N. Lyman;* ^{1, 2};

- 1. University of Nevada, Reno, NV, United States.
- 2. University of Washington, Bothell, WA, United States.

Body: Concentrations of gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) and particulate-bound mercury (PBM) were measured along with other parameters 9 km east and just north of downtown Reno. The northern location was 169 m higher than the eastern site that was situated in the valley containing the city. At both locations higher concentrations of GEM and PBM occurred during periods of low atmospheric mixing and local sources were important for enhancing concentrations above that considered continental background. Concentrations of GOM were higher (maximum of 177 pg m-3) during periods of high temperature and lower dew point. Higher GOM concentrations recorded at the higher elevation site that had less urban impact, along with other data correlations, support the hypothesis that in northern Nevada warm, dry air from the free troposphere is a source of GOM to the surface.

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Global source-receptor relationships for mercury under present and year 2050 anthropogenic emissions scenarios <u>E. S. Corbitt;</u> ¹; C. Holmes; ¹; D. J. Jacob; ^{1, 5}; D. G. Streets; ²; N. E. Selin; ³; A. Sorensen; ⁴; E. M. Sunderland; ^{5, 6}.

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- 3. Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA, United States.
- 4. National Environmental Research Institute, Aarhus University, Roskilde, Denmark.
- 5. School of Engineering & Applied Science, Harvard University, Cambridge, MA, United States.
- 6. School of Public Health, Harvard University, Cambridge, MA, United States.

Body: We use the GEOS-Chem global 3-D model for mercury, including dynamic coupling of the atmosphere with ocean and land reservoirs, to quantify continental and regional source-receptor relationships for mercury under present and future (2050) conditions. The model includes several recent developments such as oxidation of Hg(0) by Br atoms and improved representation of land-atmosphere exchange. Different SRES scenarios are considered for 2050 anthropogenic emissions, thus providing a range of future projections. We use a tagged tracer simulation to track atmospheric emissions of mercury from specific source regions including their cycling with the surface ocean and short-lived land reservoirs. We identify net source and receptor regions, distinguishing regions for which domestic emissions reductions would be most effective from others which receive deposition predominantly from the global atmospheric pool of mercury. The projected future increase in the contribution of Hg(II) to global mercury emissions results in a shift toward more regional source-receptor relationships.

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bess.corbitt@gmail.com?subject=AGU-FM09: Question regarding A44D-04'>click here to send an email

Lake Recovery Following Mercury Deposition Changes

L. Levin; ¹; K. Lohman; ²;

- 1. Electric Power Research Inst, Palo Alto, CA, United States.
- 2. Atmospheric and Environmental Research, San Francisco, CA, United States.

Body: Changes in water column and biota mercury burden in lakes are expected to follow a nonlinear trajectory in time following step changes in deposition. Results from the Canada-U.S. METALLICUS experiment indicate a time-delayed response to watershed deposition changes due to sequestration and retardation of terrain-deposited mercury. Indications from fish samples in New England and Florida are that an initial steep drop in fish content of mercury can soon follow deposition changes. The time to reach full accommodation to changes in deposition remains uncertain. Model experiments using the Dynamic Mercury Cycling Model have tested these trajectory responses to a step change in deposition from U.S. sources overlaid on a global-source upward trend over time. The shape of the time trajectory, especially in its earliest period, is sensitive to the mixing depth of divalent mercury in lake-bottom sediments.

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Production and Cycling of Methylated Mercury Species in Arctic Marine Waters

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- 1. Biological Sciences, University of Alberta, Edmonton, AB, Canada.
- 2. Chemistry, Trent University, Peterborough, ON, Canada.

Body: Monomethyl mercury (MMHg), a vertebrate neurotoxin which bioaccumulates

through foodwebs, is found in some Arctic marine mammals at levels that may be harmful to northern peoples consuming them as food. Unfortunately, sources of MMHg to polar marine food webs remain unknown, in part due to the complex nature of Hq cycling in polar marine waters. Since 2005, we have been sampling the marine waters of the Canadian Arctic Archipelago from the Canadian Coast Guard research icebreaker CCGS Amundsen. Early results demonstrated that elevated concentrations of both MMHg and dimethyl mercury (DMHg, a toxic, gaseous Hg species) are found in sub-surface Arctic marine waters (89±36 pg L-1 and 73±37 pg L-1, respectively) despite low total Hg (THg) concentrations (290±220 pg L-1), suggesting an internal source of methylated Hg. We tested the hypothesis that methylated Hg species are produced directly in the marine water column using stable-isotope Hg tracers. Seawater samples were amended with 198Hg(II) and incubated for 0, 8, 16 or 24 hours to measure the production of MM198Hg, DM198Hg and gaseous elemental 198Hg(0) (GEM) over time. A second tracer, MM199Hg, was also added to quantify MMHg methylation (formation of DM199Hg), demethylation (loss of MM199Hg) and reduction (formation of 199Hg(0)). Preliminary analysis of the data indicates that Hg(II) is methylated in polar marine waters to form both MMHg (first order rate-constant km1 ~6x10-4 d-1) and DMHg (km2 ~5x10-6 d-1). We also found that DMHg production from MMHg is ~50x faster than with Hg(II) as the substrate. Furthermore, at a small number of sites, we measured methylation rates that were elevated by almost a full order of magnitude compared to the average, suggesting that methylation hotspots may exist in Arctic marine waters. However, during the less productive fall season when the CCGS Amundsen cruises were conducted, demethylation of MMHg generally appears to dominate in the water column and can occur via a number of processes, including photodemethylation in surface waters (kpd = 1x10-3 m2 E-1) and dark/biological demethylation (kdm ~0.3 d-1). Using the measured rate constants of methylation and demethylation in a very simple model, we calculate an equilibrium MMHg concentration of 0.003-0.020 ng L-1, which is lower than the actual concentrations measured, suggesting the presence of external sources of MMHg to the water column or that methylation/demethylation activity exhibit a strong seasonality. We are currently examining factors that potentially control the biogeochemical transformations of Hg in marine polar waters, such as productivity and heterotrophic (microbial) respiration. This research will provide valuable input for global and regional Hg models as well as an understanding of the sources of MMHg to Arctic marine foodwebs, which will in turn help design strategies to minimize exposure risks to Northern peoples relying on Arctic animals for food.

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Observations of iodine oxide and reactive gaseous mercury at a coastal site in Pensacola, FL

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- 1. Department of Chemistry and Biochemistry, University of Colorado Boulder, Boulder, CO, United States.
- 2. Electric Power Research Institute, Palo Alto, CA, United States.
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Body: An increasing body of evidence suggests that atmospheric halogens, marked largely by the presence of BrO and IO, are ubiquitous components of the lower troposphere in coastal and oceanic areas. The role of halogen chemistry in mercury oxidation in coastal regions remains unknown. Simulations of atmospheric mercury oxidation suggest that chemistry initiated by atomic Br may have been underestimated. Chlorine is believed to contribute little to the observed ozone mediated-oxidation reactions. Further, synergistic effects of the iodine compounds can be expected: this effect arises from additional halogen-atom formation from IO radicals interacting with BrO radicals to enhance atomic Br and atomic I concentrations; little is known about the possible direct role I atoms may play to oxidize mercury. Finally, other reactive trace gases, such as CH2O and C2H2O2, can suppress the oxidation of Hg by converting bromine-radicals into chemically inert reservoir species. Here we present data from several months of MAX-DOAS measurements of halogen oxides, O4, CH2O and C2H2O2 in parallel with measurements of speciated mercury (Hg0, Hg2+) that are currently being conducted along the U.S. Gulf Coast. Our results demonstrate that reactive gaseous mercury (RGM) is enhanced when the air is coming from the ocean, and also elevated halogen oxide concentrations are being measured from those directions. We discuss our data in context of the sources of halogens that are currently being debated: (1) mixing down of stratospheric bromine into the troposphere, (2) sea-salt mediated air-surface exchange in the marine boundary layer, and (3) biogenic releases.

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Enhanced clear sky reflectance near clouds: What can be learned from it about aerosol properties?

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Body: Studies on aerosol direct and indirect effects require a precise separation of cloud-free and cloudy air. However, separation between cloud-free and cloudy areas from remotely-sensed measurements is ambiguous. The transition zone in the regions around clouds often stretches out tens of km, which are neither precisely clear nor precisely cloudy. We study the transition zone between cloud-free and cloudy air using MODerate-resolution Imaging Spectroradiometer (MODIS) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) measurements. Both instruments show enhanced clear-sky reflectance (MODIS) and clear-sky backscatterer (CALIPSO) near clouds. Analyzing a large dataset of MODIS observations, we examine the effect of three-dimensional radiative interactions between clouds and cloud-free areas, also known as a cloud adjacency effect. The cloud adjacency effect is well observed in MODIS clear-sky data in the vicinity of clouds. Comparing with CALIPSO clear-sky backscatterer measurements, we show that this effect may be responsible for a large portion of the enhanced clear-sky reflectance observed by MODIS. Finally, we describe a simple model that estimates the cloud-induced enhanced reflectances of cloud-free areas in the vicinity of clouds. The model assumes that the enhancement is due entirely to Rayleigh scattering and is therefore bigger at shorter wavelengths, thus creating a so-called apparent "bluing" of aerosols in remote sensing retrievals.

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Emissions of Black Carbon Particles from Biomass Burning and Their Physical and Chemical Properties

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Body: Large amounts of aerosol, including black carbon (BC), are emitted from biomass burning. It is therefore important to understand the chemical composition, rate of emissions, and mixing state of aerosols generated by this combustion process to estimate the impacts of aerosols on climate. Thus far, these physical and chemical quantities have been compiled by combining the data from laboratory and field experiments, but the data from the Arctic region are still very limited. These parameters were measured by an SP2 instrument based on the laser-induced incandescence technique on board the NASA DC-8 during the ARCTAS campaign. Aircraft sampling was made in plumes emitted by wildfires in Canada and the USA, and in those transported over long distances from Russia. First, we extract biomass burning plumes using CH3CN and SO2 data. Then, we derived the slopes of the CO-CO2-CH3CN-aerosol correlations for each burning plume. Based on this, we derive the average CO/CO2, CH3CN/CO2, BC/CO2, and BC/CO ratios together with their variability in the plumes strongly influenced by forest fires over Siberia, California, and Canada. A similar analysis is made for light-scattering particles. Using these relationships, the transport efficiencies of BC particles from the boundary layer to the free troposphere are also estimated. It is found that the BC particles were thickly coated upon emission. From comparison with AMS measurements, the coating materials are found to be mainly composed of organic compounds. This indicates the importance of the enhanced light absorption by BC particles emitted by biomass burning.

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Ammonium Nitrate Formation near the Colorado Front Range

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Body: A significant air quality issue during wintertime temperature inversions along the Colorado Front Range urban corridor is the infamous "Brown Cloud" which is dominated by ammonium nitrate particles. Aerosol composition, size distribution, and gas phase measurements were obtained along with meteorology in Boulder-based ground studies during the winters of 2005 and 2009 and in an airborne survey over the Colorado Front Range urban corridor and northeastern Colorado on April 1, 2008. New in these campaigns was the fast time response data which showed that nitric acid was partitioned mainly into the aerosol phase as ammonium nitrate. During the survey flight, ammonium nitrate mass concentrations were highest on the west side of the urban corridor whereas nitrogen oxide concentrations were highest directly west and south of Denver. Nitric acid concentrations were highest south of the city. The calculated equilibrium gas phase ammonia was highest close to the ground directly around large feed lots near Brush and west of Greeley. These differences are consistent with what is known about the locations of emission sources, the predominant flow during the experiments, and the chemistry. Indeed, the ammonia emissions in the northern part of the region are sufficiently high to cause ammonium nitrate formation to be limited by nitric acid whereas in the southern part of the region ammonium nitrate formation was limited by low ammonia emissions. Although NOx (NO + NO2) emissions in the region are much larger than those for ammonia, NOx must be converted into nitric acid in order for ammonium nitrate to form. In the survey data, aerosol nitrate was correlated with the daytime nitric acid production rate but with higher slopes in the northern parts of the region. In the longer Boulder datasets, the calculated daytime production rate was slow and comparable to nighttime heterogeneous production via N2O5 hydrolysis. During periods of low aerosol surface area, daytime and nighttime production of nitric acid resulted in freshly formed ammonium nitrate particles. These results suggest that reductions in NOx emissions along the northern part of the region are likely to decrease the prevalence of the Brown Cloud.

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Black carbon measurements in the Pearl River Delta region of China

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Body: The Pearl River Delta (PRD) region in southeastern China is one of the most polluted industrial/metropolitan areas in the world. The 3C-STAR campaign (Synthesized Prevention Techniques for Air Pollution Complex and Integrated Demonstration in Key City-Cluster Region), carried out in October-November, 2008, was aimed at improving the understanding and quantification of air pollution in the region, while developing technical capacity for regional air quality monitoring and modeling. We report single-particle soot photometer (SP2) measurements and analyses of refractory black carbon (rBC) at Kaiping, a rural site downwind of the major pollution sources in the PRD area. The rBC mass loadings varied between 0.5 and 10 µg-rBC kg-air-1, and averaged 2.8 µg-rBC kg-air-1. These values are roughly an order of magnitude higher than those measured in the Houston, Texas, a major US metropolitan area. The rBC mass distributions show a primary lognormal peak with a median mass diameter of 0.22 µm volume-equivalent diameter (VED), which is similar to those observed in Houston and other regions with the SP2 instrument. A second mode with a mass median diameter of 0.69 µm VED, has not been observed before. Coatings are found on over 50% of rBC particles, suggesting that they are aged and/or of biomass-burning origin. The high rBC loadings cause significant heating of the atmosphere due to direct solar absorption. A diurnal heating rate of over 0.5 K day-1. is estimated for the average of entire dataset with a maximum heating rate near 3 K day-1.

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The impact of the 1783-84 AD Laki Eruption on Aerosol Formation Processes and Cloud Condensation Nuclei A. Schmidt; ¹; K. S. Carslaw; ¹; G. W. Mann; ²; M. B. Wilson; ¹; T. J. Breider; ¹; S. J. Pickering; ¹;

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Body: The 1783-84 AD Laki flood lava eruption released 122 Mt of sulfur dioxide gas (SO2) over the course of 8 months into the upper troposphere and lower stratosphere above Iceland. The subsequent conversion of SO2 to sulfuric acid caused a widespread "dry fog" of SO4 aerosol affecting the entire Northern Hemisphere. Previous studies have examined the impact of the Laki eruption on the formation of SO4 aerosol and on climate using general circulation models. Here, we study the impact of the Laki eruption on aerosol microphysical processes, including the nucleation of new particles and their growth to cloud condensation nuclei (CCN) using a comprehensive Global Model of Aerosol Processes (GLOMAP). We show that the Laki eruption fundamentally altered the microphysical processes driving the aerosol size distribution in the Northern Hemisphere and that the total particle concentration in the free troposphere (FT) increased by a factor of ~16 over large parts of the Northern Hemisphere during the first 3 months after the onset of the eruption. CCN (supersaturation = 0.22%) concentrations increased by a factor of ~65 in the upper troposphere, giving peak 3-month zonal mean concentrations of ~1400 cm-3 at high northern latitudes. Even though new particle formation mostly occurred in the FT, 3-month zonal mean CCN concentrations in the boundary layer close to the eruption site increased by up to a factor of ~26, with peak concentrations of ~500 cm-3. Thus, the Laki eruption had the potential to profoundly alter cloud microphysical properties and completely dominated as a source of CCN in the pre-industrial atmosphere. Moreover, recognising that such an eruption is likely to occur again, we investigate the sensitivity of aerosol microphysical processes to the timing of the eruption. Our model simulations suggest that the impact of an equivalent wintertime eruption on upper tropospheric CCN concentrations is only about one-third of that of a summertime eruption. Thus, the microphysical processes leading to the growth of particles to CCN sizes are strongly influenced by the season of such a high latitude eruption. Using a global aerosol microphysics model like GLOMAP enables the impact on Cloud Droplet Number (CDN) concentrations to be calculated based on a fully resolved aerosol size distribution, and advances our understanding of how volcanoes influence natural background CCN concentrations.

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The Role of Ions in New Particle Formation in the Upper Troposphere and Lower Stratosphere Using WACCM/CARMA

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Body: Nucleation mode sulfate particles are known to exist in the Upper Troposphere - Lower Stratosphere (UTLS) region; however, the nucleation mechanism(s) and the role of these particles in the aerosol burden in the UTLS region are not well understood. Nucleation caused by the production of ions by solar activity, which can then produce cloud condensation nuclei, has been surmised as a sun-climate link. We use WACCM/CARMA; a three-dimensional chemistry climate model based upon the Whole-Atmosphere Community Climate Model (WACCM) with sectional microphysics from the Community Aerosol and Radiation Model for Atmospheres (CARMA) to simulate the formation and evolution of sulfuric acid aerosols in the UTLS. We will compare the aerosol size distributions predicted by a binary homogeneous nucleation scheme to that of an ion-mediated nucleation scheme.

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Multisite reconciliation of sub- and supersaturated particle water uptake.

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Body: Simplification of water uptake and cloud behaviour are required for large scale models owing to the complexity of a full treatment of all contributory factors to cloud activation. One widely used parameterisation aims to represent a particles hygroscopicity with a single parameter, which may hypothetically be derived from measurements of water uptake at a given relative humidity or supersaturation.

This κ-Köhler approximation represents the behaviour of inorganic salts (such as Sodium Chloride) reasonably well. Its application to ambient multicomponent aerosol is becoming widespread, although a systematic characterisation of its performance in both sub and supersaturated conditions has not been carried out at a comparable level of detail.

In order to probe the single parameter approximation, measured particle sub and supersaturated water uptake have been compared using a variety of instrumentation for multiple field campaigns.

A Hygroscopic Tandem Differential Mobility Analyser (HTDMA) was used to probe the aerosol (dry diameter 26nm<Dp<250nm) water uptake behaviour in the subsaturated regime, typically at 90% RH. Humidigrams were also performed, where the aerosol is subjected to interval steps in RH between 10% and 90%, to better understand the water uptake characteristics.

For a similar range of dry diameters as the HTDMA, a Cloud Condensation Nucleus counter (DMT-CCNc; Lance et al. 2006) was used to examine particle water uptake behaviour in supersaturated environments, between supersaturations of 0.07% - 1%.

The κ-Köhler model can be used to predict critical supersaturations for particles of a given diameter, given their growth factor at a given RH. Data are presented from a collection of field campaigns around the world, showing a difference between measured and predicted data; likely indicating apects of water uptake behaviour not represented in the current κ-Köhler model technique, thus limiting its capabilities with respect to global extrapolation.

We further compare the number of CCN predicted using the κ measured in sub- and supersaturated regimes with that predicted using a constant value for κ in order to evaluate whether a constant single parameter is justifiably applicable for the environments in which we have taken measurements.

Lance et al. Mapping the operation of the DMT continuous flow CCN counter. Aerosol Science and Technology (2006) Petters, M.D. and Kreidenweis, S. M., A single parameter representation of hygroscopic growth and cloud condensation nucleus, Atmos. Chem. Phys., 7, 1961-1971, 2007

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In situ vertical profiles of black carbon aerosol over the Pacific, Arctic, and Antarctic Regions (80°N to 67°S Latitudes)

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Body: In three weeks of January 2009, the NSF /NCAR GV research aircraft flew the HIAPER Pole-to-Pole Observations (HIPPO) mission, with a Single-Particle Soot Photometer (SP2) on board. The SP2 characterized the mass of refractory black carbon (rBC) in individual aerosol particles measured in situ over a total of 138 vertical profiles primarily between 0.3 and 8 km AMSL from 80N to 67 S, including some profiles to 13 km. These measurements of rBC, which are unprecedented in scope, were primarily obtained in the remote central and eastern Pacific, allowing sampling far from sources. Air of extremely low rBC loadings (rBC mass < 0.05 ng/kg air) was detected in both the southern hemisphere lower troposphere and the tropical middle troposphere. Enhanced BC loadings were observed at high northern latitudes associated with Arctic haze and at northern midlatitudes in the central Pacific in air masses tracing back to northern Asia. These results form a powerful constraint on global model treatments of rBC lifetime and transport. A comparison will be made between observed [zonal average is the mean on a latitude circle; I think you mean binned in longitude] profiles and those generated from the AEROCOM suite of global models.

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