

REMOTE SENSING OF TROPOSPHERIC POLLUTION FROM SPACE

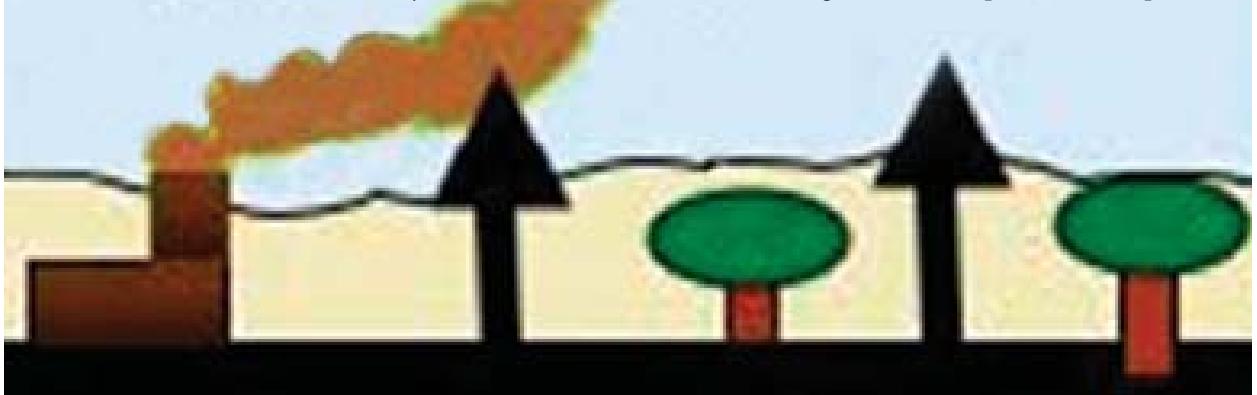
BY JACK FISHMAN, KEVIN W. BOWMAN, JOHN P. BURROWS, ANDREAS RICHTER, KELLY V. CHANCE, DAVID P. EDWARDS, RANDALL V. MARTIN, GARY A. MORRIS, R. BRADLEY PIERCE, JERALD R. ZIEMKE, JASSIM A. AL-SAADI, JOHN K. CREILSON, TODD K. SCHAACK, AND ANNE M. THOMPSON

Geostationary satellite observations of chemically reactive trace gases will provide unique insight into the evolution and extent of air pollution with the temporal resolution necessary to address air quality on a daily basis.

The ability to measure air pollution and other chemically reactive trace gases in the lower atmosphere from satellites has a heritage dating back nearly three decades when the first measurements of carbon monoxide (CO) were made from the space shuttle *Challenger* in November 1981 (Reichle et al. 1986). Since then, numerous satellite-based instruments have provided important measurements from Earth-observing platforms in low-Earth orbit (LEO), giving nearly global coverage of several key trace gases (National Research Council 2008). The National Aeronautics and Space Administration (NASA) *Aura* satellite, originally proposed as a component of the Earth Observing System (EOS) concept in the 1980s, is the most recently launched (2004)

U.S. satellite dedicated to measuring trace gases; with other satellites currently sending back atmospheric composition measurements, this decade is unique with respect to the amount of information coming from space to provide new insight into processes that control the observed distributions.

Despite significant scientific achievements derived from *Aura* and other satellites, there are no plans to launch an *Aura* follow-on. Furthermore the kinds of data that are likely to come from the next generation of satellites will probably be different from what has been seen in the past, according to the National Research Council (NRC), which recently issued a comprehensive report (see “The NRC Report” sidebar) defining the national priorities for space-



based measurements in the forthcoming decade (National Research Council 2007). For atmospheric composition, the report recommended a continuation of the measurement of the trace species already being measured [“top-priority measurements include tropospheric ozone (O_3), CO, nitrogen dioxide (NO_2), formaldehyde (HCHO), sulfur dioxide (SO_2), and aerosols”]. However, the report further stated that a combination of satellites from both LEO and geostationary Earth orbit (GEO) should be launched, with the “highest priority being a GEO mission.” The NRC’s GEO recommendation is consistent with their paradigm that future Earth observations should have direct societal benefit, in addition to providing data primarily for scientific purposes, as has been the priority in the past.

Thus, as a survey of past, present, and future capabilities, the purpose of this paper is threefold. First, we review the progress of the measurements of trace gas distributions that provide the scientific community with the first global distributions of several key species. Second, we show an example of the current research capabilities that now assimilate composition measurements into chemical transport models to provide consistent pictures of multiple species at a specific time. And last, if the next generation of tropospheric trace gas measurements is to supply information that will be used to improve our understanding of air quality, as well as to provide the observations necessary to mitigate harm to society, then such observations must be used in conjunction with operational models. This paradigm is greatly different from the rationale that lay behind the launch of the past and present fleet of satellites. Therefore, in the last part of this paper, we present a preview of research operational air quality forecasting products (Dabberdt et al. 2003; McHenry and Dabberdt 2005) and provide a snapshot of the kind of measurements recommended in the National Research Council (2007) report.

TROPOSPHERIC TRACE GAS MEASUREMENTS FROM SPACE. At lower altitudes, many of the standard techniques that can be used to derive information about the stratosphere by viewing the Earth’s limb (e.g., see Burrows 1999; Kaye and Fishman 2003) cannot be used, and, therefore, nadir viewing is required. Because several tropospheric trace species can also be present in significant quantities in the stratosphere, altitude discrimination between what is present in the troposphere and what is present in the stratospheric overburden becomes a challenge to instrument retrieval development. Key measurable trace gases that are prevalent in both the troposphere and stratosphere include ozone, as well as its most important precursor for in situ tropospheric production, NO_2 . A few important trace gases present predominantly in the lower atmosphere with enough of a spectral signature to be observed from a space platform include CO, HCHO, SO_2 , and the long-lived greenhouse gases carbon dioxide (CO_2) and methane (CH_4).

Tropospheric trace gas measurements from space were originally to be part of NASA’s *Nimbus-7* satellite, launched in 1978. Proposed to be on board was the Measurement of Air Pollution from Satellites (MAPS) instrument, a gas-filter correlation radiometer (GFCR) designed to measure carbon monoxide, which is a species of tropospheric origin and a key player in the reactions of other trace gases through its reaction with the hydroxyl radical (Crutzen and Fishman 1977; Thompson 1992). In the early 1970s, the global sources of CO were in dispute (McConnell et al. 1971; Seiler 1974), with some studies suggesting that natural sources (e.g., oxidation of methane and nonmethane hydrocarbons) were an order of magnitude larger than anthropogenic emissions (National Academy of Science 1977). A global survey of CO from space would determine the relative strengths of these sources. The then-fledgling Environmental Protection Agency would benefit by knowing whether

AFFILIATIONS: FISHMAN, AL-SAAD, AND CREILSON—NASA Langley Research Center, Hampton, Virginia; BOWMAN—Jet Propulsion Laboratory, Pasadena, California; BURROWS AND RICHTER—University of Bremen, Bremen, Germany; CHANCE—Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts; EDWARDS—National Center for Atmospheric Research, Boulder, Colorado; MARTIN—Dalhousie University, Halifax, Nova Scotia, Canada, and Smithsonian Center for Astrophysics, Harvard University, Cambridge, Massachusetts; MORRIS—Valparaiso University, Valparaiso, Indiana; PIERCE—NOAA/NESDIS/STAR, Madison, Wisconsin; ZIEMKE—NASA Goddard Space Flight Center, Greenbelt, Maryland; SCHAACK—University of Wisconsin—Madison, Madison, Wisconsin;

THOMPSON—The Pennsylvania State University, University Park, Pennsylvania

CORRESPONDING AUTHOR: Jack Fishman, Mail Stop 401A, NASA Langley Research Center, Hampton, VA 23681-2199
E-mail: jack.fishman@nasa.gov

The abstract for this article can be found in this issue, following the table of contents.

DOI:10.1175/2008BAMS2526.1

In final form 8 January 2008
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the human component of CO was large enough that regulations on CO would improve air quality.

When NASA's *Nimbus-7* was actually launched in 1978, the MAPS instrument had been removed from the payload. MAPS was modified to fly on the space shuttle and flew four times between 1981 and 1994 (Reichle et al. 1999), providing "snapshots" of CO distributions. These initial glimpses confirmed that CO was highest where human sources from industry and tropical fires dominated. Subsequent to MAPS, the Interferometric Monitor of Greenhouse Gases (IMG) obtained CO measurements for several months aboard the Japanese Advanced Earth Observing System (ADEOS) satellite in 1996–97 (Hadjilazaro et al. 1999; Barret et al. 2005) using a Fourier transform spectrometer. The IMG observations provided confirmation of the findings from

MAPS, but operated for less than a year because of the failure of the ADEOS platform in June 1997.

The Measurement of Pollution in the Troposphere (MOPITT) instrument, a GFCR, has been measuring CO from NASA's *Terra* satellite since 1999. MOPITT's relatively long-term record has been used to determine the impact of localized emissions on global air quality. Among the unanticipated findings was that wildfires in North America and Siberia were surprisingly strong sources of CO (Lamarque et al. 2003; Edwards et al. 2004), which caused ecosystem changes and alterations to the carbon balance over Earth's vegetated areas through removal of above-ground biomass, transport of CO and CO₂ into the atmosphere, and alteration of the carbon uptake by ecosystems. The 2004 Alaskan wildfire season was the region's worst on record because of unusually

THE NRC REPORT

In 2004, NASA, NOAA, and the U.S. Geological Society (USGS) requested that the NRC form a panel to identify and prioritize the next set of observational platforms that should be launched and operated over the next decade. In addition to providing information solely for the purpose of addressing scientific questions, the NRC took the approach that increasing the societal benefits of Earth science research should likewise be high on the priority list of federal science agencies and policy makers, who have long believed that the role of scientific research is not only to expand our knowledge, but also to improve the lives of Americans.

For example, in the U.S. today, an estimated 1.8–3.1 yr of life are lost to people living in the most polluted cities due to chronic exposure to particulates (Pope 2000), and more than 4,000 premature deaths per year occur because of the elevated ozone concentrations now commonly observed in the United States (Bell et al. 2004). Elevated surface ozone concentrations also have deleterious effects on crop production (Mauzerall and Wang 2001; Morgan et al. 2003) costing U.S. agriculture more than a billion dollars annually.

The NRC emphasized that if Earth scientists are to foster applications and extend the societal benefits of

their work, they must also understand the research-to-applications chain, which includes transforming satellite measurements into useful information and distributing that information in a form that is understandable and meets the needs of both public- and private-sector managers, decision makers, and policy makers.

Specifically, with respect to future atmospheric chemistry missions, the National Research Council (2007) recommended that a mission dedicated to the measurement of tropospheric trace gases from a geostationary spacecraft should be launched in the 2013–16 time frame [Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission], and that such a mission would fit into its vision of providing societal benefit. The NRC also called for another satellite similar to *Aura* to be launched again in the 2020 time frame [Global Atmospheric Composition Mission (GACM)].

The use of Earth science data for applications will first require gaining an understanding of how research-level data can be used in a successful operational environment (National Research Council 2007). Extracting societal benefit from spaceborne measurements necessitates, as an equally important second step, the development of a strong link between the measurements and decision makers

who will use such measurements. This linkage must be created and sustained throughout the life cycle of the space mission. In order to implement future missions, scientists who are engaged in research intended to have both scientific and societal contributions must operate differently than they did in the days when the advancement of science was either the primary or only goal of research. Applications development places new responsibilities on agencies to balance applications demands with scientific priorities, and the character of missions may change in significant ways if societal needs are given equal priority with scientific needs. This potential societal benefit was interwoven into the foundation for the NRC requirement of making atmospheric composition measurements from a geostationary platform its highest priority. As this new paradigm evolves, the numbers of published papers, scientific citation indices, or even professional acclamation from scientific peers will not be enough to evaluate the success of the missions that have been recommended. The degree to which human welfare has been improved and the effectiveness of protecting property and saving lives will additionally become important criteria for a successful Earth science and observations program.

warm and dry weather; in central Alaska and Canada's Yukon Territory, more than 11 million acres burned. The location of high CO values measured by MOPITT (Fig. 1) coincided with the location of fires and aerosol plumes seen by the Moderate Resolution Imaging Spectroradiometer (MODIS) that was also on *Terra*. Model analysis using these observations showed that the fires were responsible for ~30 Tg of CO during summer 2004, which is roughly equivalent to the total U.S. anthropogenic CO emissions during that time. Ground-level ozone increased by about 25% in the northern United States and impacted ozone amounts in the free troposphere as far away as Houston, Texas (Morris et al. 2006), and Europe (Pfister et al. 2005). From its first observations of immense CO plumes from forest and grassland fires in Africa and South America that traveled as far as Australia, MOPITT has provided the community with a tool for studying pollution sources, chemistry, and transport in detail.

The Atmospheric Infrared Sounder (AIRS) instrument on board the *Aqua* satellite (launched in 2002) is also capable of providing information about CO (McMillan et al. 2005). Some preliminary products are currently being developed to use this information in near-real time to investigate sources of CO and transport patterns. Comparisons of the CO data from AIRS with concurrent MOPITT observations (Warner et al. 2007) show some promise for these data, which have the advantage of providing near-global coverage on a daily basis. Most recently, the Infrared Atmospheric Sounding Interferometer (IASI) on the European Meteorological Operation *MetOp-A* satellite has also been providing CO measurements as an operational product since 2007 (information available online at http://smc.cnes.fr/IASI/GP_satellite.htm; see also Turquety et al. 2004).

TROPOSPHERIC TRACE GAS INFORMATION FROM SATELLITE MEASUREMENTS DESIGNED TO STUDY THE STRATOSPHERE.

The Total Ozone Mapping Spectrometer (TOMS) instrument aboard *Nimbus-7* was designed to produce global daily maps of total ozone so that stratospheric ozone depletion (a hypothesis in 1978) could be observed and quantified, and the processes leading to it understood (Molina and Rowland 1974). Working on the principle of measuring backscattered ultraviolet (BUV) radiation, TOMS primarily provided information about the distribution of ozone in the stratosphere where ~90% of the ozone in the atmosphere (~15–55 km) is found. The remaining ~10% is located in the troposphere below the tropopause, which is generally located at 10–18-km altitude and varies as a function of both latitude and time of year. In the tropics, the tropopause is located higher in the atmosphere (16–18 km) than at middle latitudes, where its height can be as low as 6–8 km (winter), or as high as 15–16 km (summer).

The amount of ozone in a column of air is expressed in Dobson units (DU), where 1 DU has a value of 2.69×10^{16} molecules of ozone per square centimeter. A representative amount of total column ozone is 300 DU, of which ~30 DU is in the troposphere. At middle latitudes, the synoptic distribution of total ozone is primarily determined by the larger-scale distribution of ozone within the stratosphere; in turn, these patterns generally follow the upper-level airflow on a daily basis, with the polar jet often being the demarcation between higher-stratospheric ozone poleward and lesser amounts at lower latitudes. At low latitudes, therefore, large-scale synoptic patterns of total ozone are often not definitive because of the much weaker pressure gradients generally present

there. In Fig. 2, the monthly distribution of total ozone is depicted. Note that the figure uses a color scale, where the total ozone features at middle and high latitudes are off the scale for better visualization of low-latitude variability (Fishman et al. 1990). The enhancements in total ozone found at low latitudes over the South Atlantic and adjacent continents are generally observed during austral spring (September–

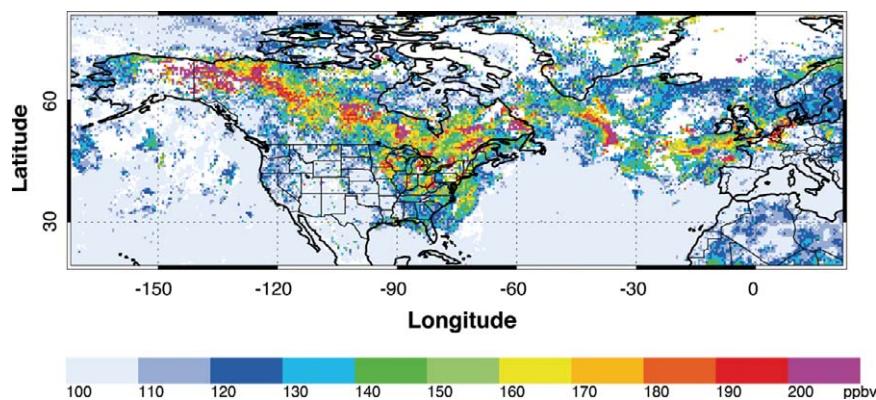


FIG. 1. MOPITT 700-hPa CO mixing ratio for 15–23 Jul 2004, during the INTEX-NA field campaign. The intense wildfires in Alaska produced plumes of pollution that can be traced across North America and the Atlantic Ocean.

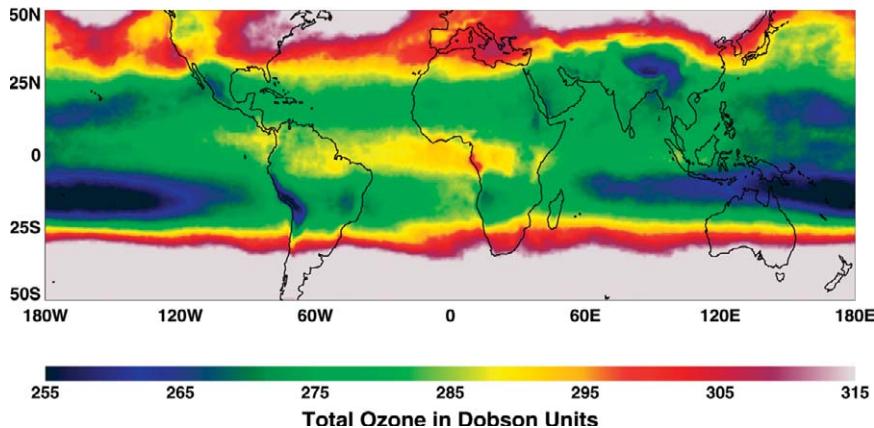


FIG. 2. Total ozone distribution for Sep 1987 as determined from the TOMS satellite (from Fishman et al. 1990).

November). Furthermore, a significant positive correlation was found with CO measurements from the first MAPS shuttle flight in November 1981 (Fishman et al. 1986).

The data depicted in Fig. 2, however, reflect total ozone measurements, of which only a relatively small percentage is found in the troposphere. At higher latitudes, meteorological activity is generally more vigorous, and persistent patterns over periods of a week or two are rare. Therefore, identification of enhanced tropospheric ozone is difficult, and quasi-persistent plumes from Europe and northern Asia would be lost beneath the stratospheric variability. Persistent ozone sources in the tropics offered the first indications that total ozone column satellite information could be used to identify ozone pollution sources.

Extracting tropospheric ozone from total ozone column measurements. The first step in retrieving the amount and distribution of many important tropospheric trace gases through space-based observations such as TOMS is to find a method for separating the tropospheric and stratospheric components. Before that methodology was developed, however, an important ozone analysis was provided in Fishman and Larsen (1987) in which Stratospheric Aerosol and Gas Experiment (SAGE) ozone profiles were used to calculate the stratospheric column ozone (SCO), which showed that stratospheric ozone at low latitudes exhibited little longitudinal variability. TOMS measurements, on the other hand, between 15°N and 15°S, showed that the distribution of total ozone was quite different from the SCO distribution, and was generally highest over the south tropical Atlantic Ocean.

During the 1990s, a number of research groups focused on extracting information about the tropo-

sphere from satellite measurements by assuming that ozone variability in the stratosphere is defined on relatively large spatial scales, compared with the troposphere, so that information could be obtained about the troposphere if this larger-scale stratospheric component could be isolated. Once the stratospheric ozone distribution was determined, the “residual” information [tropospheric ozone

residual (TOR)] could be used to infer information about the troposphere (see Fig. 3).

Using standard tropopause height information from the National Centers for Environmental Prediction (NCEP) in conjunction with collocated TOMS total ozone and SAGE profiles, Fishman et al. (1990) produced the first climatological depiction of tropospheric ozone on a domain that included both the tropics and middle latitudes (from ~50°N to ~50°S). The seasonal climatology of the TOR showed elevated amounts of tropospheric ozone at northern temperate latitudes during the summer, and in the tropics and subtropics in the Southern Hemisphere during austral spring, with the latter being a consequence of widespread biomass burning during that time of the year. This surprising finding in the South Atlantic became the focus of a major field campaign in 1992: Transport and Atmospheric Chemistry near the Equator—Atlantic (Fishman et al. 1996).

Subsequent to the TOMS-SAGE-derived TOR being published in 1990, a number of research groups employed other methodologies to independently

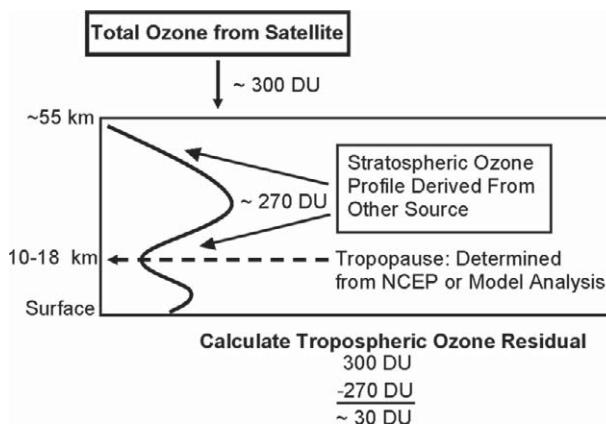


FIG. 3. Schematic of the TOR technique.

calculate SCO values and to derive a quasi-global TOR distribution (e.g., Thompson and Hudson 1999). SCO distributions were derived from the Microwave Limb Scanner (MLS) and the Halogen Occultation Experiment (HALOE) profiles from the *Upper Atmosphere Research Satellite (UARS)* launched in 1991 (Ziemke et al. 1998) and the solar backscattered ultraviolet (SBUV) profiles from *Nimbus-7* and several subsequent National Oceanic and Atmospheric Administration (NOAA) operational satellites (Fishman et al. 2003; Fig. 4).

Other techniques used characteristics of the TOMS measurements alone to develop a means of constructing an independent SCO value using information about how the TOMS made its total column measurement. Two of these techniques compare the amount of total ozone at locations that are affected by the presence of clouds with total ozone amounts in nearby cloud-free regions. The difference in the column amounts is then defined as the tropospheric column ozone (TCO). Furthermore, by having independent information about the height of the clouds, upper-tropospheric ozone profiles can then be calculated (Ziemke et al. 1998, 2001). In a somewhat similar technique, TCO has been derived using total column measurements above and adjacent

to mountains of known heights, with the difference identified as the TCO in the layer up to mountaintop height (Newchurch et al. 2001).

Direct measurement of tropospheric ozone. Direct determination of ozone profiles, including tropospheric ozone, from nadir measurements in the UV and visible was developed for the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument on the European Environmental Satellite (ENVISAT) by applying a retrieval algorithm for ozone using information from differential penetration into the atmosphere. An altitude dependence was then determined by fitting the absorption to the temperature-dependent ozone Huggins bands (Chance et al. 1991, 1997; Bhartia et al. 1996). The method was originally applied to the Global Ozone Monitoring Experiment (GOME) spectral measurements (Munro et al. 1998; Hoogen et al. 1999) and has now been used to determine a multiyear climatology of tropospheric ozone (Liu et al. 2005, 2006). SAGE also obtained direct measurements of ozone in the upper troposphere at low latitudes by using an occultation technique when cumulus clouds are not present (Wang et al. 2006). Each of the direct measurement techniques

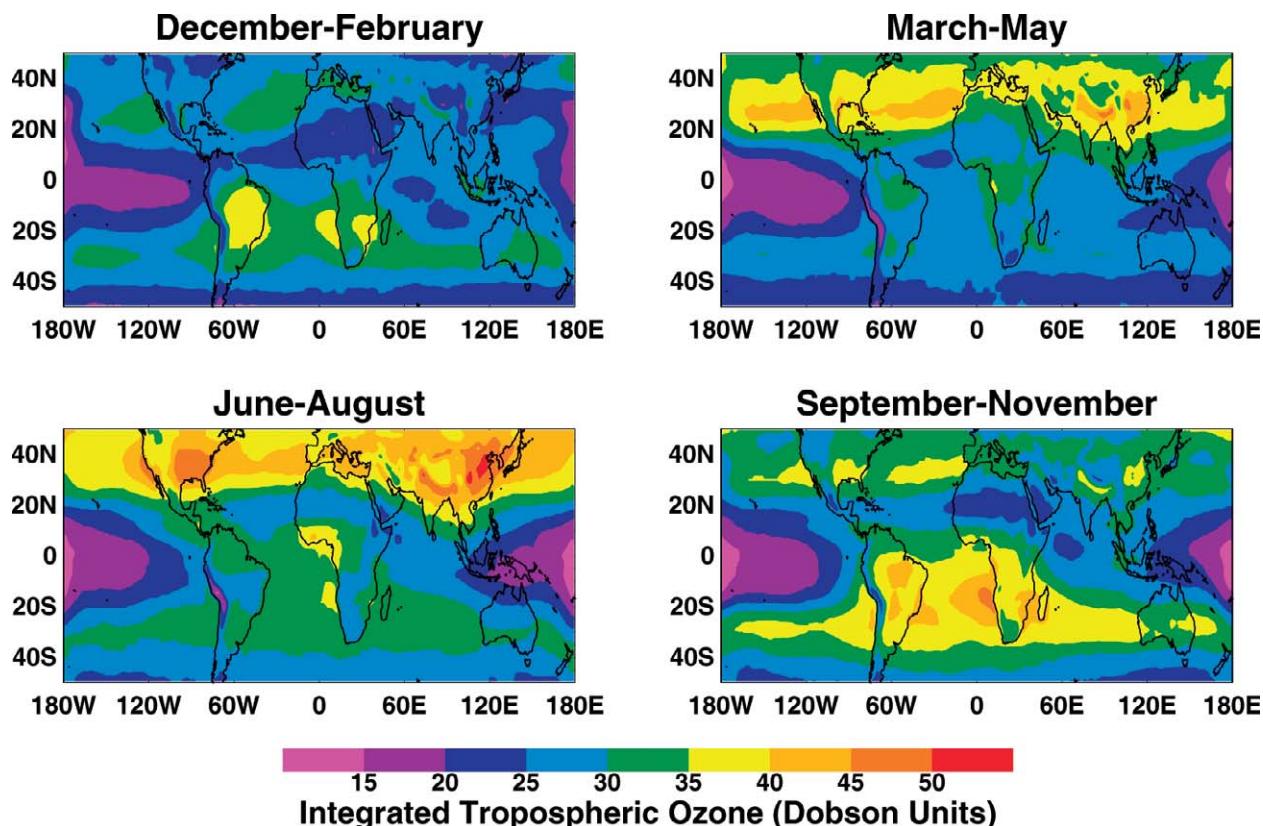


FIG. 4. Climatological depiction of TOR using TOMS and SBUV measurements (Fishman et al. 2003).

produces tropospheric ozone distributions consistent with the features originally found using the TOR methodology.

TROPOSPHERIC NO₂. Although significant quantities of NO₂ are found in both the stratosphere and troposphere, the amount can vary by more than two orders of magnitude, with nearly all of this variability in the troposphere. The amount in the stratosphere is first determined empirically using the knowledge that stratospheric NO₂ is most dependent on the length of day (i.e., the integrated photolysis of the reactive nitrogen reservoirs), and thus has a strong latitudinal dependence. Defining a value of NO₂ found over a region far from pollution sources (generally over the Atlantic or Pacific) as the stratospheric component of the total column at that latitude is the first step for determining how much NO₂ is in the troposphere. This quantity is then subtracted from the observed NO₂ total column for the rest of the day at the same latitude to derive how much of that NO₂ is in the troposphere (Richter and Burrows 2002). Alternatively, the stratospheric NO₂ contribution can also be estimated using chemical transport models (CTMs) or assimilated NO₂ fields. Tropospheric NO₂ columns have been retrieved independently from GOME, SCIAMACHY, and the Ozone Monitoring Instrument (OMI) by several groups (Martin et al. 2002; Richter and Burrows 2002; Beirle et al. 2003; Boersma et al. 2004; Bucsele et al. 2006). Figure 5 shows tropospheric NO₂ columns retrieved from SCIAMACHY (Martin et al. 2006). Pronounced enhancements are evident over major urban and industrial regions. The high degree of spatial heterogeneity provides empirical

evidence that most of the tropospheric NO₂ column is concentrated in the lower troposphere and close to local emissions. As a result, tropospheric NO₂ columns retrieved from satellite observations show considerable prospects for inferring surface nitrogen oxides (NO_x) emissions through inversion of the NO₂ observations (Leue et al. 2001; Martin et al. 2003; Jaeglé et al. 2005).

FORMALDEHYDE. Formaldehyde was hypothesized to exist in ubiquitous quantities in Levy's (1971) classic paper on tropospheric photochemistry as an intermediate product of methane oxidation through reactions with the hydroxyl radical (OH). Background HCHO concentrations coming directly from CH₄ oxidation are too small to be observed from present-day satellites, but the oxidation of other volatile organic compounds (VOCs) that react quickly with OH also produces large amounts of HCHO. Thus, HCHO serves as a major proxy for these species when they are present in large quantities. Formaldehyde was first measured from GOME spectra during the intense biomass burning over Asia in 1997 (Thomas et al. 1998). Development of more sensitive measurement capabilities has led to a global HCHO climatology that is used to determine the distribution of VOC emissions (Chance et al. 2000; Abbot et al. 2003; Shim et al. 2005; Fu et al. 2007). Figure 6 depicts the average HCHO distribution during August 2005 from OMI. The major source region over the United States is in the southeastern states where densely populated deciduous forests permeate the landscape. Although anthropogenic VOC emissions are also important for the generation of photochemical pollution, the

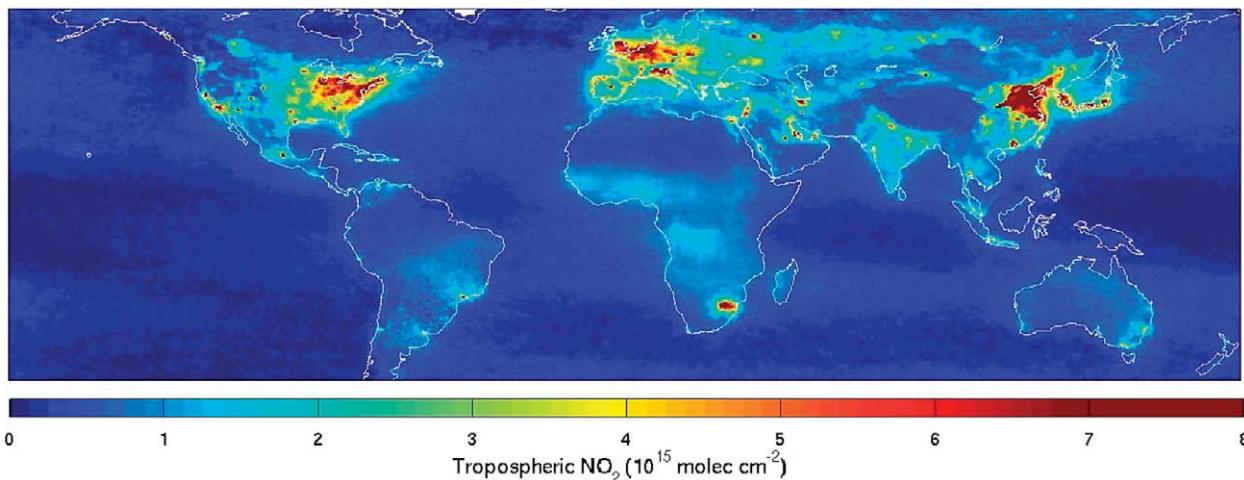


FIG. 5. Tropospheric NO₂ columns retrieved by Martin et al. (2006) from the SCIAMACHY satellite instrument for Jan 2003–Dec 2005 inclusive.

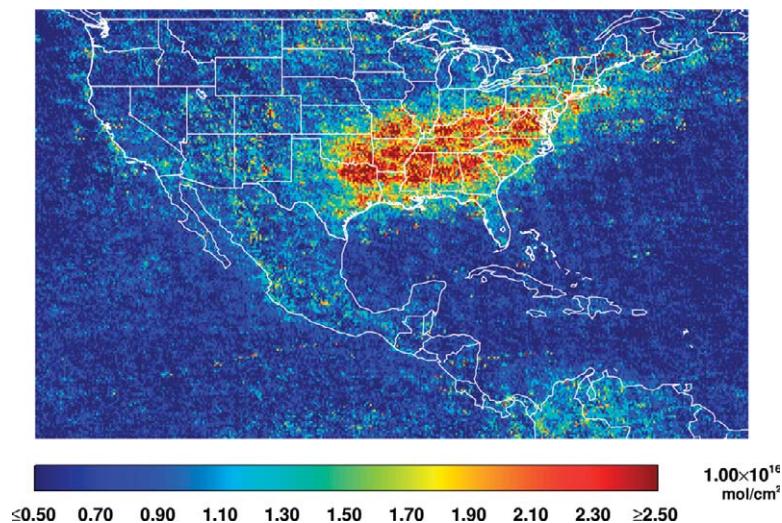


Fig. 6. HCHO distribution during Aug 2005.

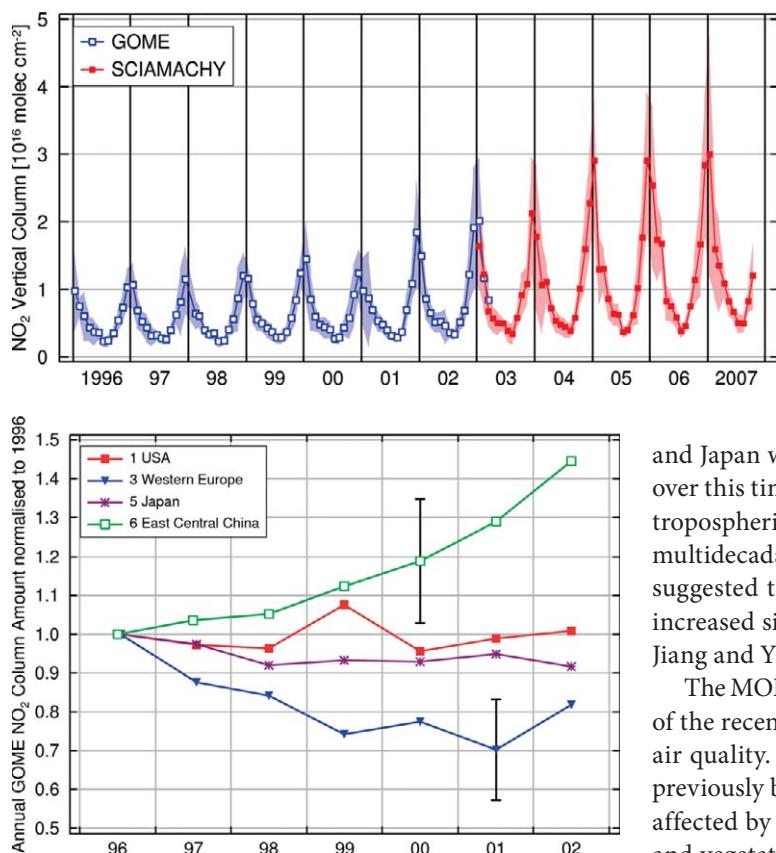


Fig. 7. (top) The monthly averages of tropospheric vertical columns of NO_2 over east-central China defined for the area bounded by $30^\circ\text{--}40^\circ\text{N}$ and by $110^\circ\text{--}123^\circ\text{E}$ are shown. Both GOME (blue open circles) and SCIAMACHY (red filled circles) are shown. (bottom) Annual trend information for the above region (China), a region over Europe, one over the United States, and one over Japan (after Richter et al. 2005) are shown.

HCHO distribution illustrates the dominance of the natural sources in this region. Recently, the glyoxal radical, CHOCHO, has been measured from space, using OMI and SCIAMACHY spectra (Kurosu et al. 2005; Wittrock et al. 2006); glyoxal is another proxy for VOC sources with shorter lifetimes, and likely serves as a better indicator of urban sources (Volkamer et al. 2005).

PROVIDING GLOBAL INSIGHT INTO TRENDS AND INTERANNUAL VARIABILITY.

As tropospheric trace gas data records extend to a decade or longer, important trend information about some emissions in specific regions of the world is now becoming available. For example, NO_2 measurements from the GOME and SCIAMACHY instruments (Fig. 7) show an increase in tropospheric NO_2 columns over industrial China during the last decade, confirming substantially larger regional emissions than a decade earlier (Richter et al. 2005). This same dataset also shows that emissions have decreased over much of Europe and that trends in the United States

and Japan were near zero, if not slightly downward, over this time frame. From the analysis of long-term tropospheric ozone column measurements using the multidecadal record of TOMS, several studies have suggested that the tropospheric ozone column has increased since the early 1980s (Ziemke et al. 2005; Jiang and Yung 1996).

The MOPITT dataset now provides a global record of the recent interannual variability of tropospheric air quality. The observed variability is greater than previously believed due to large fire emissions being affected by climatic conditions that control rainfall and vegetation drying (Fig. 8). These data form the foundation of a long-term observational record that is used to investigate links among chemistry, climate, and other components of the Earth system (Edwards et al. 2003, 2004; Yurganov et al. 2005). For example, strong correlations have been observed between Southern Hemisphere CO zonal average loading, fire activity in Indonesia, and El Niño conditions (Edwards et al. 2006).

Regional interannual variability of tropospheric ozone has been linked to El Niño–Southern Oscillation in both western Africa and northern India (Fishman et al. 2005). As seen in Fig. 9, considerably more tropospheric ozone is present in northeastern India in June during an “El Niño” year (1982) than over the same region during a “La Niña” year (1999). The primary reason for this is the delay of the onset of the summer monsoon during El Niño years, thereby providing more favorable conditions for the photochemical generation of ozone pollution throughout the entire month. Also seen in this figure is the generally higher amount of tropospheric O_3 observed over eastern China in 1999 compared with that in 1982. Fishman et al. (2005) showed that there was no correlation between TOR amounts and ENSO indices over eastern China, but the difference shown in these panels is consistent with the increasing trend found downwind over the Pacific (Ziemke et al. 2005), as well as with the sizeable increase of NO_2 observed by GOME and SCIAMACHY over eastern China (Richter et al. 2005).

TROPOSPHERIC MEASUREMENTS AND DATA ASSIMILATION. Launched in 2004,

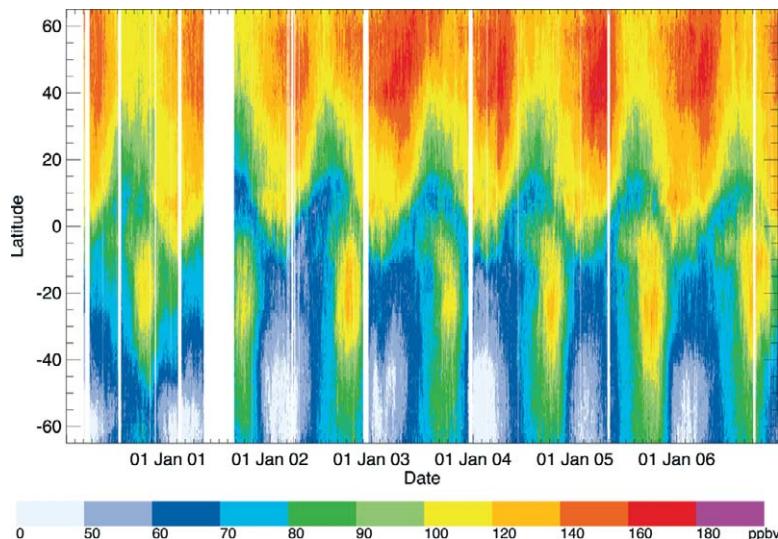


FIG. 8. Zonal plot showing the CO 700-hPa mixing ratio at different latitudes over recent years. In the Southern Hemisphere, there is a large amount of CO emitted from agricultural fires in Africa and South America every year in the late summer. In the Northern Hemisphere, CO is produced by industry, urban activity, and wildfires, and it is usually removed from the atmosphere by photochemistry during the summer. However, western Russian fires in the late summer of 2002 and Siberian fires in the spring of 2003 produced so much CO that pollution built up to produce a very “dirty winter.”

Aura is an entire satellite devoted to atmospheric chemistry. The Tropospheric Emission Spectrometer (TES), a Fourier transform spectrometer whose heritage traces back to the Infrared Interferometer Spectrometer (IRIS) aboard the *Nimbus-4* spacecraft (Hanel and Conrath 1969), provides simultaneous observations of CO and tropospheric O_3 vertical profiles (Beer 2001). Simultaneous observations of

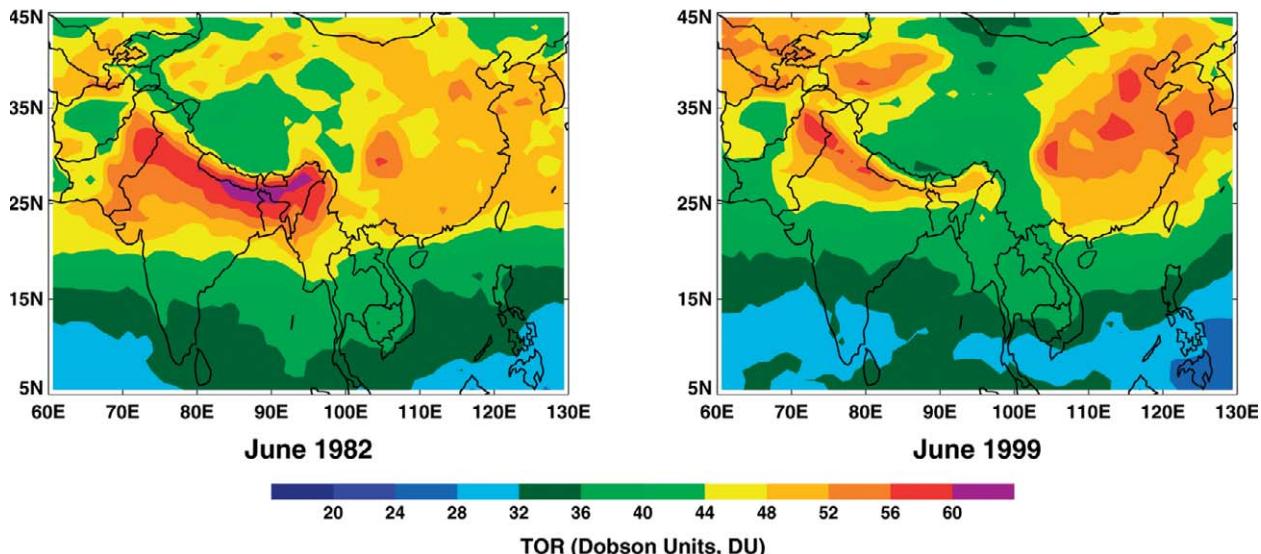


FIG. 9. Monthly distributions of TOR (DU) during Jun 1982 (El Niño year) and Jun 1999 (La Niña year).

these two species are particularly valuable for distinguishing between natural and anthropogenic sources of ozone (Fishman and Seiler 1983). IASI, a French instrument aboard the European *MetOp-A* satellite (Turquety et al. 2004), started providing concurrent CO, tropospheric O₃, and CH₄ in 2007, in addition to the GOME-2 NO₂ product that is also aboard *MetOp-A*. The spatial and vertical information from these measurements is critical for understanding the complex interplay between dynamics and chemistry

that determines the global distribution of ozone in the troposphere.

The value of concurrent measurements is further enhanced through data assimilation. For example, the tool used to derive the fields in Fig. 10 is from the NASA Langley Research Center/University of Wisconsin (LaRC/UW) Regional Air Quality Modeling System (RAQMS; Pierce et al. 2003, 2007). The assimilation of OMI data provides an overall constraint on total column ozone, while the assimilation

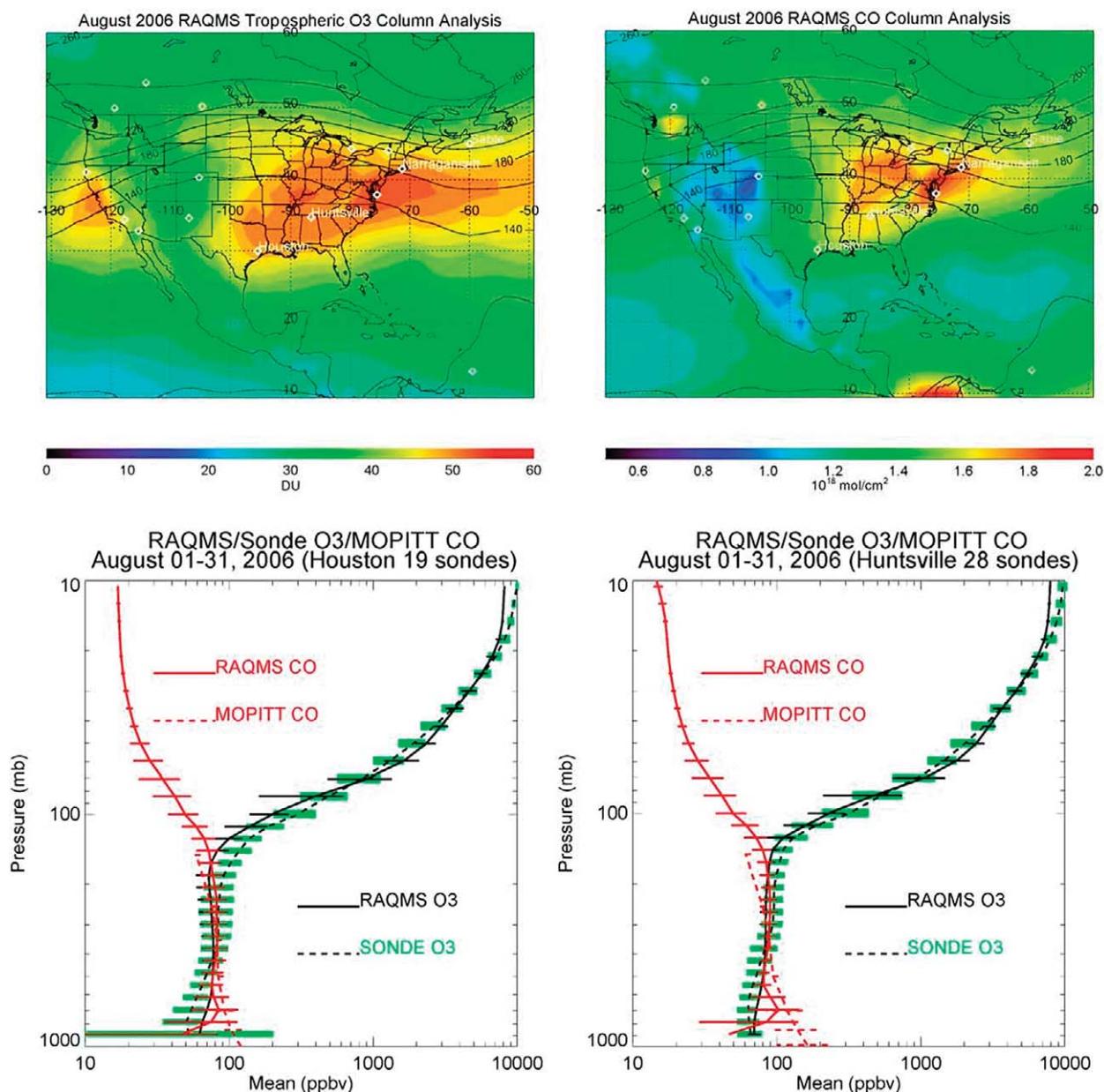


FIG. 10. (top) Mean distribution of O₃ and CO based on 6-hourly RAQMS analyses over the continental United States during 2006 is shown. The location of the IONS sites are indicated by white diamonds with selected sites labeled. The mean pressure of the analyzed thermal tropopause is contoured. (bottom) Comparisons for Houston and Huntsville IONS sites are shown. RAQMS (solid) and MOPITT (dashed) mean CO profiles and standard deviations (red), RAQMS (solid) and IONS (dashed) mean O₃ profiles (black) and std devs (green) are shown.

of profile retrievals constrains lower-stratospheric and tropospheric O₃ and CO from TES. The RAQMS meteorological forecasts are initialized from NOAA's Global Forecast System (GFS) analyses at 6-h intervals, and the RAQMS OMI column assimilation likewise occurs at 6-h intervals. The RAQMS TES O₃ and CO assimilation occurs at 1-h intervals.

Daily ozonesonde launches were conducted throughout the continental United States during August 2006 as part of the Intercontinental Chemical Transport Experiment Ozonesonde Network Study (IONS) program (Thompson et al. 2007). The average profile distributions in Fig. 10 show the August mean

RAQMS-analyzed tropospheric O₃ and total CO columns along with comparisons between RAQMS and IONS O₃, and RAQMS and MOPITT CO for selected IONS sites. Mean MOPITT CO profiles at the IONS ozonesonde sites were obtained through composites of all daytime MOPITT observations within 1° of an IONS site during August 2006.

The analyzed tropospheric ozone columns seen in Fig. 10 show a broad maximum in excess of 50 DU over the south-central and eastern United States and the western Atlantic, and a localized ozone maximum off the California coast. The analyzed total column CO distribution shows a similar CO maximum over

BASIC TROPOSPHERIC CHEMISTRY AND WHAT CAN BE OBSERVED FROM SPACE

Tropospheric ozone is the central character that drives the chemistry of the lower atmosphere. From an atmospheric chemist's point of view, if the global distribution of tropospheric ozone and the evolution of that distribution are understood, then many of the basic questions driving the science can be answered. The primary challenge is the fact that ozone is both a natural and a man-made component of the lower atmosphere (as opposed to the stratosphere, where ozone is produced only naturally). The two major sources of O₃ are its transport from the huge stratospheric reservoir and its in situ photochemical production from the release of anthropogenic and biogenic precursors that are oxidized in the atmosphere to eventually become ozone. Most of these trace gases are initially oxidized by the OH radical and its abundance and distribution, in turn, are most dependent on the O₃ present in the troposphere. Thus, if the distribution of O₃ is well known, then we can get a good handle on the oxidizing capacity of the atmosphere and the global extent of air pollution, two of the "grand challenges" put forth in the integrated global observations strategy for atmospheric composition

(Barrie et al. 2004). Global observations of trace gases using satellites have already provided the community with a unique set of observations that have yielded important insights by being able to measure CO, NO₂, HCHO, and O₃, itself.

Tropospheric O₃ is formed when certain precursor trace gases are oxidized in the atmosphere by the OH radical. The two most important precursor trace gases are CO and a class of gases known as VOC species. However, the amount of O₃ that forms is most critically dependent on the available concentration of NO_x, which is primarily the sum of NO and NO₂. Putting it into a chemical reaction formulation, there are two potential pathways:

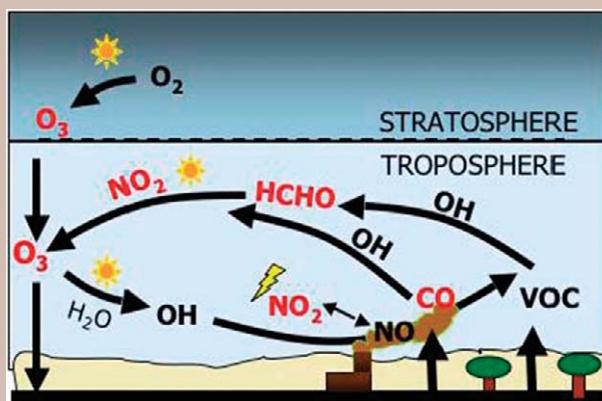


or



From satellites, we can measure HCHO (a surrogate for VOC), CO, O₃, and NO₂ (one of the two primary components of NO_x).

FIG. SB1. Schematic diagram of simplified tropospheric photochemistry. Species in red can be measured using existing satellite instrumentation. CO, NO, and VOCs are emitted in the planetary boundary layer from both anthropogenic and biogenic sources. NO₂ is rapidly converted in the troposphere from emitted NO and the two NO_x establish an equilibrium ratio between each other depending on a number of atmospheric variables, such as the intensity of sunlight and the amount of O₃. HCHO is an intermediate product of VOC oxidation, and its measurement is a good indication of the amount of VOCs present. The amount of O₃ produced is directly proportional to how much NO₂ is present when sunlight is available (denoted by the "sun" symbol). Ozone can also be transported to the troposphere from the stratosphere where it is produced naturally by the photolysis of molecular oxygen (O₂) because of high-energy photons in the upper atmosphere (λ < 242 nm) that are capable of breaking apart the O₂ molecule. Lightning is also a natural source of NO_x in the free troposphere.



the south-central and eastern United States, but no significant CO enhancement off the California coast. The collocated tropospheric O₃ and CO enhancements are due to photochemical ozone production and emissions associated with power generation facilities located east of the Mississippi and large urban centers along the Great Lakes and northeastern United States. The tropospheric O₃ enhancements without significant CO enhancements are due to accumulation of stratospherically influenced air within the persistent subtropical high pressure system off the California coast.

With the capability to discriminate more than one layer in the troposphere, observations from TES have also provided new insight into how the ozone maximum found off the west coast of Africa (Fishman et al. 1990, 1996) evolved (see Fig. 4). Subsequent to its discovery using satellite data, in situ observations from ship cruise campaigns (Thompson et al. 2000) and model studies (Edwards et al. 2003; Martin et al. 2002) showed that the early-year tropospheric ozone distribution over the tropical Atlantic Ocean is characterized by two maxima—one in the lower troposphere north of the intertropical convergence zone (ITCZ) and one in the middle and upper troposphere south of the ITCZ. This feature could not be resolved from the original analysis using the remote-sensing capabilities available in the 1990s. In Jourdain et al. (2007), TES vertical ozone retrievals provided the first space-based observations that characterized this complex altitude-dependent interplay between chemistry and transport. In particular, an enhanced layer of O₃ was observed by TES below 600 hPa that was advected by Harmattan winds from burning over western and central Africa.

THE RESEARCH-TO-APPLICATIONS PARADIGM FOR AIR QUALITY MEASUREMENTS FROM SPACE: RECOMMENDATIONS FROM THE NRC DECADAL SURVEY.

As shown from the preceding discussion, the wealth of information from the instruments aboard *Aura* is being used in conjunction with data assimilation models to provide insights into processes that take place on synoptic scales. This new paradigm for using spaceborne atmospheric composition measurements was tested in near-real time during the 2006 Intercontinental Chemical Transport Experiment (INTEX)-B and Texas Air Quality Study (TexAQS) field campaigns. Initially the foci of these multiagency integrated satellite-aircraft campaigns were distinct and different: the objective of INTEX-B was to investigate the transport and transformation of pollution

from Asia to North America, whereas the TexAQS-2006 campaign focused on the effects of distant and local sources on air quality in Texas. Collectively, however, these studies provided an exciting opportunity to investigate global and regional impacts of air quality using a variety of spaceborne sensors. The integration of these measurements with in situ data, models, and assimilation techniques has yielded rich gains in our understanding of air quality (see information online at www.espo.nasa.gov/intex-b/index.html; <http://esrl.noaa.gov/csd/2006/>). In addition, the National Research Council (2007) promotes this approach as a new paradigm for air quality prediction analogous to the pioneering efforts of the weather community during the second half of the twentieth century when hourly observations from geostationary platforms were looped through computer animation to provide unique insights into cloud formation and water vapor transport.

With respect to atmospheric composition, the first operational products have become a reality in recent years (Al-Saadi et al. 2005; see also information online at <http://idea.ssec.wisc.edu/>; <http://airnow.gov/>). This information is used to advise people to remain indoors if they are susceptible to respiratory stress when exposed to elevated levels of pollution. The use of future satellite technology to improve forecasts of detrimental aspects of air quality underlay the NRC's recommendation for the next atmospheric composition mission to be from geostationary orbit.

The atmospheric chemistry-composition community has traditionally not faced the challenge of using tropospheric trace species measurements in a forecast mode, and as the NRC report clearly indicates, the tools currently do not exist to use such information. The most obvious missing piece is the lack of sufficient temporal resolution. An example of this shortcoming is shown in Fig. 11, which summarizes model results and measurements obtained during an *Aura* validation campaign on 22 and 23 June 2005. The two OMI NO₂ column measurements over Houston are depicted by the red circles on the plot and were made at ~1900 UTC on each day; the distribution at the time of the measurement for the 2 days is shown in the center and right panels above the plots. The left-top panel shows the NO₂ column distribution calculated with the Community Multiscale Air Quality (CMAQ) model (Byun and Ching 1999). On the hourly plot, there are two sets of calculated quantities: surface NO₂ concentration (magenta) and integrated tropospheric NO₂ (dark blue), the latter being measured by OMI. Because its distribution is dominated by local sources (see Fig. 5), the NO₂ observed by the

satellite is closely linked to its concentration at the surface. Furthermore, the diurnal behavior of these quantities is also closely linked. If an instrument with spectral resolution comparable to that of OMI were in geostationary orbit, hourly observations of the type indicated by the blue diamonds during daylight hours (yellow shading) would be possible. Thus, an instrument using today's measurement capability that "stares" at a region throughout the course of the day would capture the most significant part of the diurnal variability that is totally missed by the low-Earth-orbiting *Aura* platform, which finds NO₂ values of 8 and 4 × 10¹⁵ molecules cm⁻², for 22 and 23 June, respectively.

To improve the ability to measure ozone in the lower atmosphere, and especially near the surface, important advances in instrument capability may also be needed (Edwards 2006). Recently, Worden et al. (2007) showed the potential of combining UV and IR spectral radiances from both TES and OMI in a unified retrieval algorithm with significantly improved sensitivity in the troposphere. This approach may lay the groundwork for a new class of instruments that incorporate wavelengths across the ultraviolet, visible, and infrared spectrums that could revolutionize our observational capability from space of CO and O₃ within the planetary boundary layer, and usher in a new era of air quality forecasting and prediction. The use of the Geostationary Imaging Fourier Transform Spectrometer (Smith et al. 2005) for trace gas measurements, active remote sensing for trace gases such as ozone (Browell et al. 2004) from low-Earth orbit, or the placement of an instrument with the powerful spectral resolution of an instrument like IASI in geostationary orbit provide the promise of additional techniques that will advance measurement capability beyond the time frame addressed in the decadal survey.

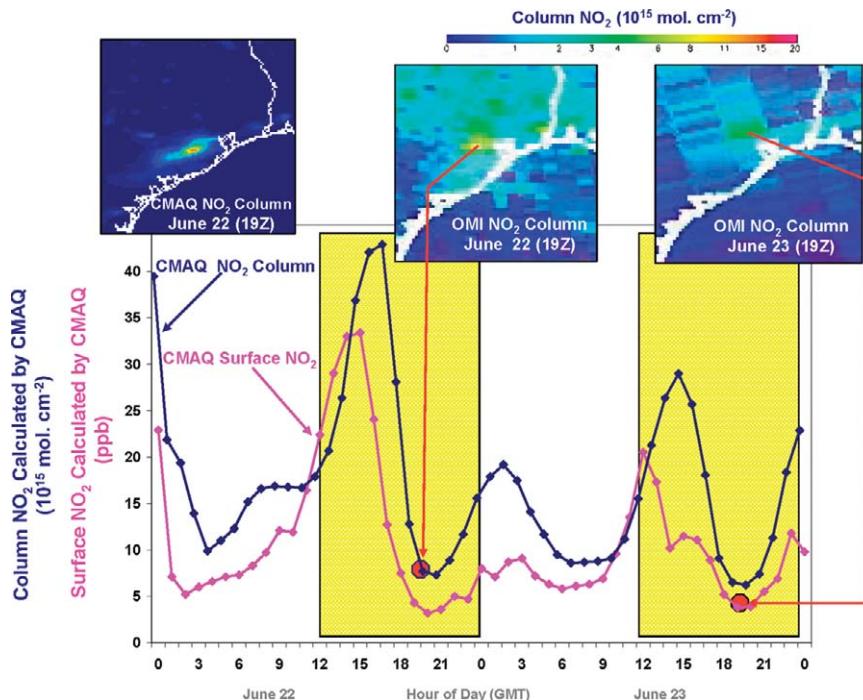


FIG. 11. Two OMI NO₂ column measurements over Houston are depicted by the red circles at the time of the measurement (~1900 UTC 22 and 23 Jun 2005). The three depictions at the top portion of the figure are NO₂ column distributions: (left) calculated using CMAQ with a 12-km resolution and (middle, right) from OMI for the two respective days; all three depictions use the color scale above them. The diurnal calculations shown by the curves are the NO₂ surface concentrations (magenta) and the NO₂ columns (dark blue) calculated by the CMAQ model for Houston and are plotted hourly over this 2-day period. The yellow shading indicates daylight hours and illustrates when data using the solar backscattered technique (as used by OMI) could be obtained from a geostationary orbit.

Regardless of the instrumentation that eventually resides in geostationary orbit to provide continuous high-spatial-resolution measurements of tropospheric trace gases (and aerosols), a substantial challenge also awaits the general atmospheric chemistry/composition community to develop the mathematical tools that use this information. This point is emphasized in the Integrated Global Atmospheric Chemistry Observations strategy (Barrie et al. 2004), which cited the multipronged necessity of a strong modeling component as part of an integrated approach that will provide insight into tropospheric chemical and transport processes. Additionally, the modeling component will serve as the foundation for the eventual development of a robust air quality forecasting capability. As part of Global Earth Observation System of Systems (GEOSS; online at www.epa.gov/geoss), the satellite system of measurements needs to be integrated with data from ground-based observational (atmospheric composition) sites

to develop a seamless three-dimensional field that can be used for both research and operational purposes. As pointed out by the air quality/atmospheric chemistry satellite science community (Edwards 2006), the biggest piece missing in the grandiose scheme of GEOSS is the type of measurement capability that can be obtained only from a geostationary satellite. The NRC provides a roadmap that recommends such a mission in the 2013–16 time frame.

The basic technology specifically mentioned in the decadal survey already exists and has been successfully demonstrated from LEO platforms. From the traditional meteorological perspective, the use of satellite information made a quantum leap when sensors were placed upon geostationary platforms. There is no doubt that similar advancements will be realized when sensors devoted to atmospheric composition measurements are likewise put on a geostationary platform.

ACKNOWLEDGMENTS. The views, opinions, and findings contained in this report are those of the author(s) and should not be construed as an official National Oceanic and Atmospheric Administration or U.S. Government position, policy, or decision.

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