The Remote Sensing of Tropospheric Composition from Space

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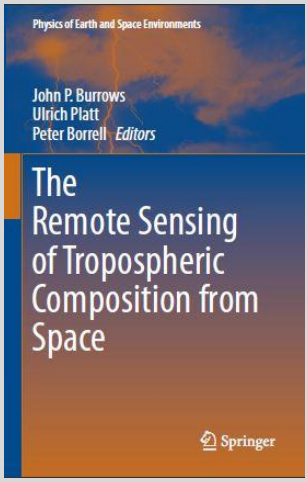

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Chapter 8

Applications of Satellite Observations of Tropospheric Composition

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Chapter 8
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Paul S. Monks and Steffen Beirle

8.1 Introduction

The advent of satellite measurements of the troposphere has taken us from a local/regional view of composition previously available from ground-based measurements to a global view. This revolution in tropospheric research has only occurred in the last three decades. Although instruments for surface mapping and meteorological parameters were recognized as remote sensing applications from the start of the space age, the remote sounding of tropospheric constituents by satellite instrumentation, often conceived for stratospheric measurement, has been used initially to give a new global view of tropospheric composition.

The time scales of atmospheric processes range from seconds to decades. For example, a pollution episode may only be apparent for a week or less, but the wider impact of such an episode may last much longer. This reflects the fact that processes such as the emission rate to the planetary boundary layer, chemical production, homogenous (*e.g.* radical–molecule, radical–radical, photolysis) and heterogeneous reactions, including both wet and dry deposition, determine the production and removal rates of species within the atmosphere. Remote sounding of the troposphere from satellites yields measurements of atmospheric composition, which give regional and global views on spatial and temporal scales not available from any other observing system. The number and type of measurements of a species required to provide a true global representation depends on the atmospheric lifetimes of the species involved.

The challenge of remote sensing the troposphere from satellite platforms is substantial and it is only with the current generation of satellite instruments and improvements in retrieval that a view of the troposphere has become available from space. An era is dawning in which long time series measurements of the troposphere

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from space will become available leading to new understanding of regional and global change. One of the challenges currently being tackled is the assimilation and fusion of different data streams to give a more holistic view of chemical and physical processes in the troposphere.

After the launch of a satellite for each instrument there are a number of scientific stages for the application of satellite data from the first-light in space to the calibration and validation of the satellite product which is a continuous process. The next stage is an observational stage, where the scientific discovery comes from an appreciation of the temporal and spatial distribution of the trace compound, the final stage becomes quantitative analysis with a model.

The aim of this chapter is to give an overview about the utility of satellite observations for measuring tropospheric composition. The focus will be on probing the chemical composition of the atmosphere. The retrieval and applications of aerosols are considered separately in Chapter 6 and the methodologies for the retrieval and validation of the trace species are described in Chapters 2, 3 and 7. This chapter summarises the tropospheric chemical species that can be measured from space and looks at the generalised applications of these measurements at the primary observational level. For a full list of satellites that have measured atmospheric composition in the troposphere and stratosphere readers are referred to the table in Appendix A of this book and also to Burrows (1999) and Martin (2008).

8.2 Overview of the Tropospheric Chemical Species Measured from Space

In this section the tropospheric gases that can be measured from space will be reviewed. Table 8.3 lists the tropospheric chemical species that have been measured from space. A recent overview of the main scientific questions and drivers for tropospheric composition can be found in Monks et al. (2009).

8.2.1 Tropospheric Ozone, O_3

Ninety percent of atmospheric O_3 can be found in the stratosphere; on average only about 10% resides in the troposphere. While stratospheric O_3 determines the amount of short wavelength radiation available to initiate photochemistry (Monks 2005), tropospheric O_3 acts as initiator, reactant and product in much of the oxidation chemistry that takes place in the troposphere.

Tropospheric O_3 was one of the first chemical species (other than water) to derived from space-based total O_3 column observations based on the UV radiances measured by the TOMS instrument combined with stratospheric measurements and/or SAGE and SBUV to give tropospheric residuals (Fishman 1991b; Fishman et al. 1990; 1996). This technique utilises back scattered solar radiation and was refined

by Hudson, Thompson and co-workers (Hudson et al. 1995; Hudson and Thompson 1998; Kim et al. 1996) to give a tropical tropospheric O_3 product. Ziemke and co-workers have advanced the residual-type retrieval methodologies using cloud slicing techniques as well as synergistic use of TOMS (Ahn et al. 2003; Ziemke et al. 1998; 2000; 2001; 2003) and latterly OMI (Choi et al. 2008; Schoeberl et al. 2007) with stratospheric O_3 measurements from HALOE and MLS. These methods have also been advanced using assimilation techniques (Stajner et al. 2008).

Measurements of tropospheric O_3 were expanded using GOME-1 (Hoogen et al. 1999; Liu et al. 2006b; Munro et al. 1998). A number of groups have gone on to improve the retrievals of tropospheric O_3 using a variety of methodologies (Del Frate et al. 2005; Iapaolo et al. 2007; Liu et al. 2007; Muller et al. 2003; Tellmann et al. 2004; van der A et al. 2002). There has been an extensive study to compare GOME-1 O_3 profiles from nine different algorithms (Meijer et al. 2006).

Recent tropospheric O_3 products have become available from TES (Nassar et al. 2008; Osterman et al. 2008; Richards et al. 2008; Worden et al. 2007a; 2007b) that offers direct measurements of tropospheric O_3 from mid-IR spectra. Using IASI data for example, Eremenko et al. (2008) have shown the ability to map out the O_3 distributions during the European heatwave of 2007.

The general distribution and inter-annual variability of tropospheric O_3 as measured from space has been shown using a range of satellite sensors (Fishman and Brackett 1997; Fishman et al. 2005; Liu et al. 2006b; Ziemke et al. 2006). Satellite derived tropospheric O_3 has been used to investigate the influence of stratospheric air masses on tropospheric vertical O_3 columns over the Pacific (Ladstatter-Weissenmayer et al. 2004). Various tropospheric and stratospheric O_3 satellite data has been combined with O_3 sondes to produce O_3 climatologies (Lamsal et al. 2004). Early work indentified tropospheric pollution episodes (Fishman et al. 1987) which have recently been improved to show regional pollution (Fishman et al. 2003).

In the tropics, there has been a comparison of tropical O_3 columns from GOME with a model (Valks et al. 2003), as well as studies of O_3 over Africa (Meyer-Arnek et al. 2005a). The impact of biomass burning (BB) on tropical Atlantic O_3 has also been assessed (Jourdain et al. 2007). Long terms trends in satellite derived tropospheric O_3 over the Pacific have been derived showing a significant upward trend in the mid-latitudes of both hemispheres but not in the tropics (Ziemke et al. 2005). O_3 has been used in combination with other tracers to investigate lightning (Martin et al. 2007), oxidant budgets over the Indian ocean (Ladstatter-Weissenmayer et al. 2007a), pollution flows from north America (Choi et al. 2008) and the effects of the 2006 El Nino (Logan et al. 2008).

The distribution of O_3 and other trace gases are governed by the complex interaction of dynamical, chemical and radiative processes. Feedbacks within the chemistry climate system, in particular the impact of changing O_3 on the Earth's climate system via radiative forcing in the upper troposphere/lower stratosphere region which is sensitive to such perturbations, has been thought to be of particular importance (Worden et al. 2008). There are number of satellite instruments that give solely upper tropospheric (and lower stratospheric) views of O_3 (and a range of other tracers, see for example Section 8.2.6) (Coheur et al. 2005; Fischer et al. 2008;

Hegglin et al. 2008; Nardi et al. 2008; Raspollini et al. 2006; Rozanov et al. 2007). Studies of upper tropospheric O_3 have included assessing the impact of biomass burning (Clarmann et al. 2007).

8.2.2 Nitrogen Dioxide, NO_2

Nitrogen oxides ($NO_x = NO + NO_2$) are released into the troposphere from a variety of biogenic, anthropogenic and physical sources including fossil fuel combustion, biomass burning, microbial activity in soils and lightning discharges. There is still some debate about the exact magnitude of the various sources and sinks for NO_x (Lerdau et al. 2000). According to present estimates, about 30% of the global budget of NO_x comes from fossil fuel combustion with almost 86% of the NO_x emitted in one form or the other into the planetary boundary layer from surface processes. Other major sources are biomass burning ca. 19%, microbial release from soil 32% and lightning 13% (Schumann and Huntrieser 2007). Typical NO/NO_2 ratios in surface air are 0.2–0.5 during the day tending to zero at night. Over the timescales of hours to days NO_x is converted to nitric acid and nitrates, which are subsequently removed by rain and dry deposition.

Satellite measurements of tropospheric NO_2 have found widespread utility. Observational analyses have demonstrated the strong weekly cycles in the observed NO_2 (Beirle et al. 2003), the influence of biomass burning (Burrows et al. 1999; Ladstatter-Weissenmayer and Burrows 1998; Thomas et al. 1998) and continental scale outflow (Leue et al. 2001; Richter and Burrows 2002).

Owing to the importance of nitrogen dioxides in air quality (DEFRA 2003) there has been a focus on comparisons of tropospheric NO_2 measurements with ground sites for regional air quality (Blond et al. 2007). Measurements have been compared to ground based measurements in Kyrgyzstan (Ionov et al. 2006), St Petersburg (Poberovskii et al. 2007), the Moscow region (Timofeev et al. 2000), Switzerland (Schaub et al. 2005; 2007), the Milan area (Ordonez et al. 2006) and in the urban UK (Kramer et al. 2008). Comparisons have also been made with airborne data over the south-eastern USA (Martin et al. 2004b), the Alps/Mediterranean (Heue et al. 2005), over the Atlantic (Bucsela et al. 2008), and over Shanghai (Chen et al. 2009). On a regional scale, there have been comparisons with regional models over North America (Chun'e and Baoning 2008; Kim et al. 2009), Asia (Han et al. 2009; Uno et al. 2007), Africa (Meyer-Arnek et al. 2005a) and Europe (Konovalov et al. 2005). The data over Europe showed varying agreement with bottom-up emission inventories highlighting both apparent over and under-estimates (Konovalov et al. 2006; 2008).

Tropospheric NO_2 data have been used to quantify NO_x emissions from soil (Bertram et al. 2005; Jaegle et al. 2004), shipping (Beirle et al. 2004a; Richter et al. 2004; Franke et al. 2009), power plants (Kim et al. 2006; 2009) and lightning

(Beirle et al. 2004c; 2006; Boersma et al. 2005; Choi et al. 2005; Hild et al. 2002; Martin et al. 2007; Sioris et al. 2007; Thomas et al. 2003).

The length and quality of the space-based NO_2 records has allowed the observation of trends. Trends in tropospheric column densities of NO_2 can highlight the effectiveness of legislative abatement methods as well as the accuracy of emission inventories. There has been particular interest in the NO_2 trend over the developing countries in eastern Asia (Irie et al. 2005) and, in particular, China (Richter et al. 2005; van der A et al. 2006; He et al. 2007; Zhang et al. 2007) as well as the global trend (Richter et al. 2005; van der A et al. 2008; Hayn et al. 2009). Richter et al.

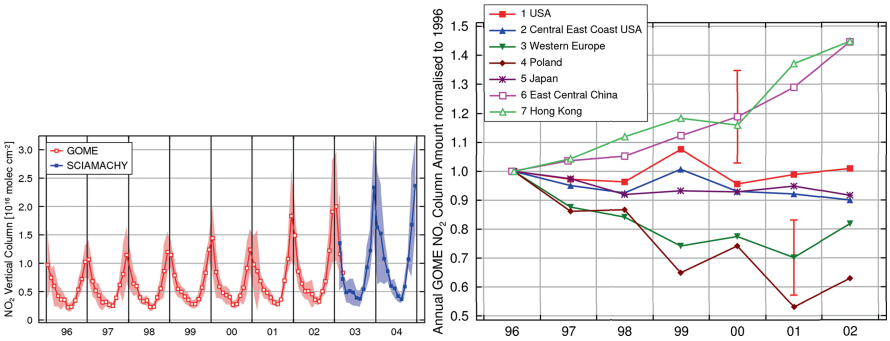

Fig. 8.1 *Left*: monthly averages of tropospheric vertical columns of NO_2 over eastern central China. The temporal evolution of tropospheric NO_2 columns from GOME and SCIAMACHY. *Right*: the mean annual NO_2 column amount normalized to that in 1996 for the geographical regions USA, central east coast USA, western Europe, Poland, Japan, eastern central China, and Hong Kong. The *error bars* represent the estimated uncertainty (s.d.) for an individual year, the values over China being larger as a result of the poorer knowledge and therefore larger uncertainty of the aerosol loading and its change (Richter et al. (2005) (Reprinted by permission of Macmillan Publishers Ltd)).

(2005) showed that there had been a 50% increase in the emissions of NO_2 over China in the period 1996–2004 (see Fig. 8.1 and Section 8.3.3).

Owing to the nature of NO_x emissions there have been many studies that use the satellite data in combination with other data sources and models to constrain emission budgets. For example, Jaegle et al. (2005) used inverse methods to take GOME NO_2 data and partition the emissions between fossil fuel, biomass burning and soil emissions. A number of groups have inverted the space data in order to derive emissions for China (Wang et al. 2007a), Europe (Konovalov et al. 2008) and globally (Jaegle et al. 2005; Konovalov et al. 2006; Muller and Stavrakou 2005; Stavrakou et al. 2008). Stavrakou et al. (2008) inverted the 10 year GOME/SCIAMACHY record such that the largest emission increases were found over eastern China, and in particular in the Beijing area (growth rate of 9.6% per year), whereas appreciable emission decreases were calculated over the United States (−4.3% per year in the Ohio River Valley), and to a lesser extent over Europe

(−1.4% per year in Germany, −1.0% per year in the Po Basin). They noted that the emission changes result in significant trends in surface O_3, amounting to increases of more than 15% per decade over large parts of China in summertime.

A number of groups have used a combination of satellite data and models to estimate global emissions of NO_2 (Ma et al. 2006; Martin et al. 2003; Toenges-Schuller et al. 2006; Zhang et al. 2007) without the formal inversion of the data using emission inventory (bottom-up) methods. Over China, Ma et al. (2006) have compared the satellite data to a model constrained by differing emission inventories, while Zhang et al. (2007) used a dynamic methodology to fit the observed trends over China to the emission inventories. Non-model based statistical methods have been used to infer global emission budgets from GOME measurements (Leue et al. 2001). The data have been used to assess model performance (Savage et al. 2004). The distribution and budget of tropospheric NO_x over Asia, especially India, are examined using a global 3-D chemistry-meteorology model and GOME NO_2 columns (Kunhikrishnan et al. 2004a). Pollution and the influence of stratospheric input in the Mediterranean (Ladstatter-Weissenmayer et al. 2003; 2007c) region has been explored using multi-tracer satellite observations.

A global comparison of NO_2 data with models has been undertaken showing the utility of these data (Lauer et al. 2002; Velders et al. 2001). The approach has been extended with an extensive multi-model comparison (van Noije et al. 2006) that highlighted a combination of model uncertainty and retrieval bias affected the goodness of fit.

The role of NO_2 in O_3 budgets has been investigated globally (Choi et al. 2008; Edwards et al. 2003), regionally over the Mediterranean (Ladstatter-Weissenmayer et al. 2007c), the tropics (Sauvage et al. 2007) and over China (Zhao et al. 2006).

As recently stated by the HTAP report "Observations from the ground, aircraft and satellites provide a wealth of evidence that ozone (O_3) and fine particle concentrations in the UNECE region and throughout the Northern Hemisphere are influenced by intercontinental and hemispheric transport of pollutants." Tropospheric satellite observations of composition have been key observational indicators and quantitative constraints (Keating and Zuber 2007).

Damoah et al. (2004) have shown that Russian forest fires produced NO_2 and other products as well as CO plumes which can be transported appreciable distances. Spichtinger et al. (2001; 2004) have observed the long-range transport (LRT) of NO_2 from Canadian boreal fires (Fig. 8.2). Stohl et al. (2003) used the space-data to characterise express pathways for LRT over the North Atlantic. Wenig et al. (2003) have tracked the long-range transport of South African power plant emissions across the Pacific. Kunhikrishnan et al. (2004b) have examined the export of NO_2 plumes from Africa and Indonesia over the central Indian Ocean.

Guerova et al. (2006) have used a combination of model and satellite data to estimate the impacts of transatlantic transport episodes on summer O_3 in Europe. More recently Bousserez et al. (2007) used model and satellite data to characterise both the anthropogenic and biomass burning influences in the North Atlantic LRT during the summer of 2004.

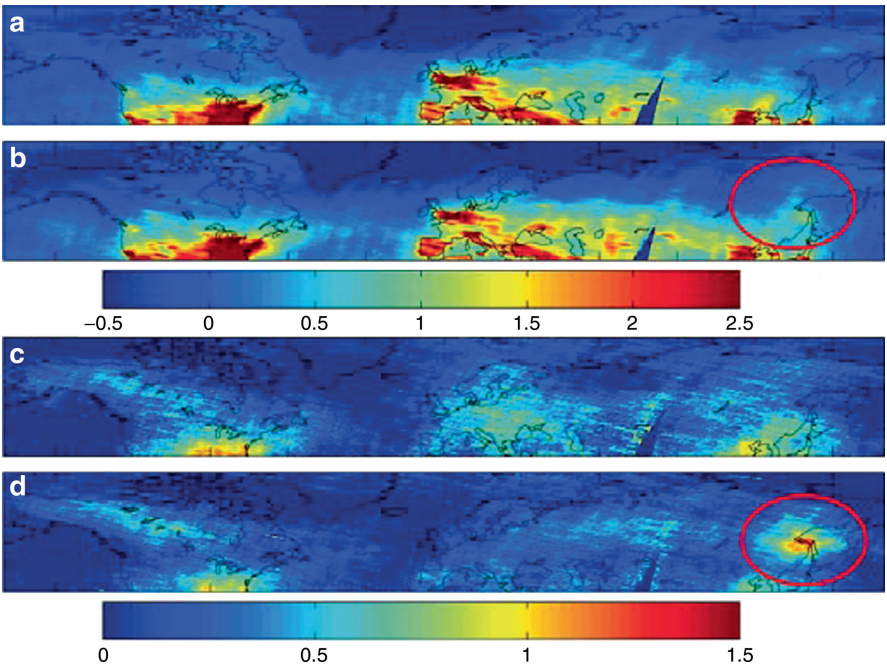

Fig. 8.2 GOME tropospheric NO_2 and HCHO columns averaged from July to August 1997 (**a** and **c**), and 1998 (**b** and **d**). The red circles mark enhanced NO_2 and HCHO columns over eastern Siberia where the strongest fire activity occurred in 1988 (Spichtinger et al. 2004).

8.2.3 Carbon Monoxide, CO

CO plays a central role in tropospheric chemistry, being a universal product of hydrocarbon photochemical degradation chemistry as well as a primary pollutant. Owing to its relatively long atmospheric lifetime (in the order of months) it is also a useful tracer for tropospheric dynamical phenomena. In the background atmosphere a combination of CO and CH_4 are the main loss routes for OH. The main sources of CO are the oxidation of CH_4 and other non-methane hydrocarbons (NMHC) with 40–60% of surface CO levels over the continents, slightly less over the oceans, and 30–60% of CO levels in the free troposphere, being estimated to come from NMHC oxidation (Poisson et al. 2000). The other major source of about equal magnitude is the incomplete combustion of either fossil fuels or biomass.

The first global space-based CO measurements were made from the MAPS instrument on board the space shuttle (Connors et al. 1999; Newell et al. 1999). Many of the gross features of global CO e.g. biomass burning, anthropogenic pollution, NH/SH gradients where identified (Connors et al. 1999; Newell et al. 1999). The data were used in early satellite data assimilation experiments (Lamarque et al. 1999). Fig. 8.3 shows CO data taken from the IMG on ADEOS which flew for

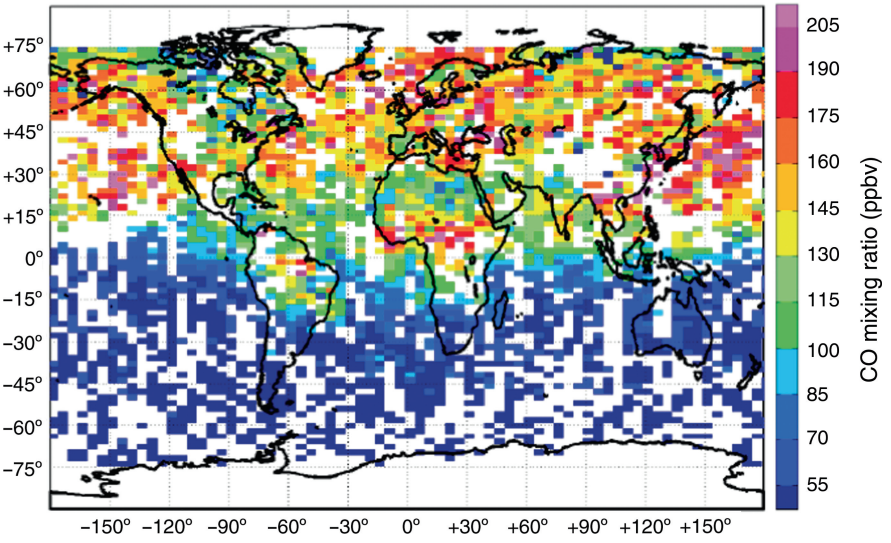

Fig. 8.3 CO volume mixing ratios (ppb) in the lower troposphere (1.2 km) for 1st–10th April 1997 retrieved from the cloud filtered IMG spectra (averages on a 2° × 5° grid) (Barret et al. 2005). Compare this with Fig. 8.13c.

a period in 1996/7 (Barret et al. 2005) giving a glimpse of the global distribution and the role of fires.

With the advent of MOPITT measurements (Deeter et al. 2003; 2004; Drummond and Mand 1996; Edwards et al. 2004; Emmons et al. 2004) global measurements of tropospheric CO have revolutionised our understanding of the natural and man-made tropospheric pollution. As an example, Fig. 8.4 shows average CO mixing ratios at the surface level, as derived from the measurements from March 2000 to June 2007 over China and parts of India and Japan, which are among the most populated areas of the globe (Clerbaux et al. 2008b). Daytime observations over land were used as the highest thermal contrast is expected (Deeter et al. 2007b) giving maximum information content along with an increased sensitivity towards the surface.

These measurements can be taken down to city scale, for example in Mexico City (Massie et al. 2006). Extensive validation of MOPITT has taken place (Emmons et al. 2007). MOPITT has been shown to be able to capture the influence of synoptic processes on the horizontal and vertical distribution of CO (Liu et al. 2006a).

Measurements from SCIAMACHY (Buchwitz et al. 2004; 2005b; 2006; de Laat et al. 2006; Frankenberg et al. 2005b; Gloudemans et al. 2008) which measures in the NIR, is more weighted to the surface than thermal IR measurements and shows enhancements of CO over urban regions (Buchwitz et al. 2007a). A comparison of SCIAMACHY data has been made with MOPITT data (Buchwitz et al. 2006, 2007a; Gloudemans et al. 2005; Straume et al. 2005; Turquety et al. 2008).

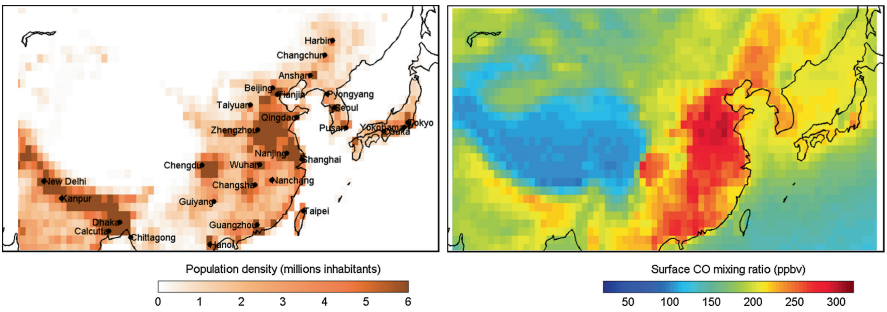

Fig. 8.4 *Left panel*: Population density (source CIESIN, in million inhabitants http://sedac.ciesin.columbia.edu/gpw) over China and surroundings. All cities with more than two million inhabitants are indicated. *Right panel*: MOPITT CO mixing ratios at the surface level (obtained by averaging the MOPITT L2 measurements from March 2000 to June 2007). Adapted from Clerbaux et al. (2008b).

Comparison of SCIAMACHY CO data has been made with models showing that the seasonal variation of the model is very similar to that of the SCIAMACHY measurements. For certain locations, significant differences were found, which are probably related to modelling errors owing to CO emission uncertainties (de Laat et al. 2006; 2007). Validation of the measurements has been undertaken with FTIR (Dils et al. 2006) and ship-borne observations (Warneke et al. 2005).

CO measurements are also available from AIRS (McMillan et al. 2005) and have been used to look at long-range transport of pollution (Stohl et al. 2007; Zhang et al. 2008). Comparisons between AIRS CO and MOPITT and ground-based remote sensing measurements have been made (Warner et al. 2007).

Recently, TES has provided profile measurements of CO (Rinsland et al. 2006b) which have been validated against aircraft measurements (Luo et al. 2007a) and MOPITT (Luo et al. 2007b) observations. Vertically-resolved CO concentration profiles have been retrieved from ACE, extending from the mid-troposphere to the thermosphere (from about 5 to 110 km) (Clerbaux et al. 2005; 2008c; Hegglin et al. 2008). Similarly, there are upper tropospheric CO measurements made from MLS (Filipiak et al. 2005; Livesey et al. 2008; Pumphrey et al. 2007). Observations from MLS have been used to show persistent maxima in CO and minima in O_3 within the anticyclone in the upper troposphere-lower stratosphere (UTLS) throughout summer, and variations in these tracers are closely related to the intensity of underlying deep convection (Park et al. 2007).

A range of new observations of CO are just becoming available from IASI (Clerbaux et al. 2009).

a General Transport Phenomena

Owing to the nature of CO as a marker of anthropogenic pollution and biomass burning, it has been used in a number of studies to assess the importance of

transport pathways. The data has shown evidence of vertical transport of CO (Kar et al. 2004) in the Asian summer monsoon. The transport pathways of CO in the African upper troposphere during the West African monsoon have been investigated through the assimilation of CO observations by the Aura Microwave Limb Sounder (Barret et al. 2008). Assimilation of CO into a chemical transport model was also used to evaluate spring time transport over West Africa (Pradier et al. 2006). MOPITT CO data in combination with trajectory analysis has been used to show that transport from the tropics to the extratropics is a comparatively slow process, giving rise to the appearance of "transport barriers" in the subtropics (Bowman 2006).

Tropical climatologies of water vapour, O_3, CO, and nitric acid from a variety of satellite, aircraft, and balloon-based measurement platforms have been used to test convective parameterisations (Folkins et al. 2006a). Satellite and sonde observations have been use to explore the seasonal cycles of O_3, CO and convective outflow at the tropical tropopause (Folkins et al. 2006b). Bhattacharjee et al. (2007) have explored the influence of a dust storm on CO and water vapour over the Indo-Gangetic Plains showing that these can lift surface CO rapidly throughout the troposphere.

An intriguing feature has been observed in daytime measurements of CO over the Middle East during spring and summer. Enhanced CO is observed over the Zagros Mountains of Iran, following the local topography over this region. It has been argued that this feature is formed by the processes of mountain venting by thermal winds caused by strong daytime differential heating (Kar et al. 2006).

b Hemispheric Transport of Air Pollution

Satellite CO data has found great utility in the investigation of the magnitude and impact of the hemispheric transport of air pollution (Keating and Zuber 2007; Singh et al. 2006). For example, Asian outflow and trans-Pacific transport of CO and O_3 pollution has been tracked using an integrated satellite, aircraft and models (Heald et al. 2003). High CO levels measured at surface background stations and by satellite in/over China (Zhao et al. 2007) have been shown to arise from LRT of biomass burning and biofuel burning areas located in the border areas of Pakistan and India.

Satellite observations of CO columns from MOPITT and of aerosol optical depths from MODIS have been useful in mapping North American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclones (Li et al. 2005). Guerova et al. (2006) have used a combination of models and satellite data to assess the impact of transatlantic transport episodes on summertime O_3 in Europe. Interlaced long-range pollution events of contrasting origin and age the tropical Atlantic Ocean have been delineated with a combination of *in situ* measurements, satellites, trajectories, emission inventories and global models (Gros et al. 2004). A combination of two satellite CO measurements has been used to look at long-range transport of CO over the land and ocean (Warner et al. 2007).

Satellite CO data in combination with *in situ* aircraft data and models has been used to characterize the Asian chemical outflow and relate it quantitatively to its sources as well as determining its chemical evolution during transport (Allen et al. 2004; Jacob et al. 2003). The relationship between satellite and *in situ* observations of CO has been explored in pollutant outflow based on a large-scale feature sampled over central northern Pacific (Crawford et al. 2004). Observations from multiple satellite sources of CO (and other tracers) has been used to investigate emission and export from Asia (Turquety et al. 2008) showing that, compared to observations, there seem be an underestimation of emissions, especially in eastern Asia. Zhang et al. (2008) used an ensemble of aircraft, satellite, sonde, and surface observations to better understand the mechanisms for transpacific O_3 pollution and its implications for North American air quality using CO measurements from TES and AIRS.

c Emission Estimates

CO measurements from space have been used to estimate global emission sources of CO (Arellano et al. 2004; Lin et al. 2007; Petron et al. 2004; Stavrakou and Muller 2006). There has been many attempts to look at regional emission budgets for Europe (Pfister et al. 2004) and Asia (Heald et al. 2004; Peng and Zhao 2007; Tanimoto et al. 2008; Yumimoto and Uno 2006). Emissions in Asia for a range of compounds have been predicted to rise sharply owing to increasing industrialisation (Monks et al. 2009).

For Asia, a combination of satellite and aircraft observations have been used to derive estimates of emissions of 361 Tg y^{-1} for CO (Heald et al. 2004). Tropospheric CO budget analysis suggests that in northern China, surface emission is the largest source of tropospheric CO (Peng and Zhao 2007). Further, model results underestimate CO by 23% in northern China (Peng and Zhao 2007). Arellano et al. (2004) noted that CO emissions in eastern Asia are about a factor of 1.8–2 higher than bottom-up estimates. Adjoint inverse modelling of satellite CO data gives annual anthropogenic (fossil and biofuel combustion) CO emissions over China of 147 Tg (Yumimoto and Uno 2006). Inverse estimates of the CO emissions from China up to 2005 suggested an increase of 16% since 2001, in good agreement with MOPITT satellite observations and the bottom-up estimates up to 2006 (Tanimoto et al. 2008).

Global estimates for the total anthropogenic surface sources of CO (fossil fuel + biofuel + biomass burning) have been derived using inverse modelling of the satellite observations (Petron et al. 2004; Yudin et al. 2004). From this approach sources of CO (all in Tg(CO) per year) were determined to be 509 in Asia, 267 in Africa, 140 in North America, 90 in Europe and 84 in Central and South America (Petron et al. 2004). Emission changes in CO have been inferred from two sets of satellite observations separated by 10 years (Shindell et al. 2005). In a case study using SCIAMACHY CO, it has been possible to estimate the emission of CO on a country scale for the UK (Khlystova et al. 2009).

d Fires (Biomass Burning)

Biomass burning (BB) is a significant process in the earth system which, as a stochastic process, is difficult to model. CO measurements from space have been used for a range of scientific purposes such as the estimation of global biomass burning emission sources of CO (Arellano et al. 2006), the identification of CO plumes from forest fires (Lamarque et al. 2003; Lee et al. 2005) (see Table 8.1) and the impact of biomass burning on regional air quality (Choi and Chang 2006a).

An extensive study has been undertaken by Edwards et al. (2006a) to investigate southern hemisphere BB using a combination of satellite CO and aerosol measurements coupled to models. Further in a separate study, Edwards et al. (2006b) used a 5 year CO data record to examine the inter-annual variability of the southern hemisphere CO loading and show how this relates to climate conditions which determine the intensity of fire sources. The observations showed an annual austral springtime peak in the SH zonal CO loading each year with dry season BB emissions in South America, southern Africa, the maritime continent, and northwestern Australia. Although fires in southern Africa and South America typically produce the greatest amount of CO, the most significant interannual variation is due to varying fire activity and emissions from the maritime continent (SE Asia including the Philipines, Indonesia and Malaysia) and northern Australia (Edwards et al. 2006b; Rinsland et al. 2008). In related work (Gloudemans et al. 2006), model results show a large contribution of South American BB CO over Australian BB regions during the 2004 BB season of up to 30–35% and up to 55% further south, with smaller contributions for 2003. BB CO transported from southern Africa contributed up to a similar to 40% in 2003 and around 30% in 2004 (Gloudemans et al. 2006). Elevated SH upper tropospheric CO as well as a range of small molecule organic tracers (C_2H_6, HCN, and C_2H_2) has been detected from a combination of biomass burning emissions and long-range transport (Rinsland et al. 2005). A combination of ship-based FTIR observations and satellite observations has been used to investigate the BB over the South Atlantic (Velazco et al. 2005) with observations of recurring enhancements of CO in the upper troposphere (10–15 km) in the equatorial regions and the South Atlantic.

Table 8.1 Overview of regionalised biomass burning episodes identified using CO from space

Fire Area	Year	Reference
Idaho-Montana Forest	August 2000	Lamarque et al. (2003)
East coast of Korea	April 2000	Choi and Chang (2006b)
N.W. USA Forest	2000	Liu et al. (2005)
SE Asia	2001	Zheng et al. (2004)
Russian Forest	May 2003	Lee et al. (2005)
Russian Forest	2003	Generoso et al. (2007)
Central Asia	2003	Wang et al. (2006)
Alaska	2004	Pfister et al. (2005), Turquety et al. (2007)
Indonesia	2006	Rinsland et al. (2008)
European Arctic	2006	Stohl et al. (2007)

Using MOPITT data and emission inversion techniques Pfister et al. (2005) showed that the Alaskan wildfires of 2004 emitted 30 ± 5 Tg CO during June to August 2004 which is comparable to the anthropogenic emissions of the continental US. Similarly, Turquety et al. have shown the importance of peat burning and pyroconvective injection (Turquety et al. 2007) for the same fires. Turquety et al. (2007) have made an estimate of North American fire emissions during the summer of 2004 to be 30 Tg CO, that includes 11 Tg from peat burning.

A temporary increase in northern hemispheric tropospheric CO burden in 2002 and 2003 (Yurganov et al. 2005) has been ascribed to boreal fires in Russia. Using CO data from MOPITT and AIRS, Yurganov et al. (2008) have concluded that fires can explain a substantial fraction of the interannual variability of CO_2.

Large differences in CO and O_3 have been measured over Indonesia and the eastern Indian Ocean in October to December 2006 relative to 2005, in 2006 O_3 was higher by 15–30 ppb (30–75%) while CO was higher by up to 80 ppb in October and November, and about 25 ppb in December. These differences were caused by high fire emissions from Indonesia in 2006 associated with the lowest rainfall since 1997, reduced convection during the moderate El Nino, and reduced photochemical loss because of lower H_2O (Logan et al. 2008; Rinsland et al. 2008). Space-based CO measurements and O_3 sondes have been used to look at the spatial and temporal variation of biomass burning over South Africa and South America (Bremer et al. 2004).

The global impact of biomass burning is very much dependent on the injection height and temporal nature of the fires. Satellite CO and surface measurements have been used to examine the injection properties of boreal forest fires (Hyer et al. 2007a) as well as the effects of source temporal resolution on transport simulations of the emissions (Hyer et al. 2007b).

Biomass burning is impacting the lower stratospheric composition. CO in the lower stratosphere (LS) observed with the Aura Microwave Limb Sounder (MLS) shows an annual oscillation in its composition that results from the interaction of an annual oscillation in slow ascent from the TTL to the LS and seasonal variations in sources, including a semi-annual oscillation in CO from biomass burning (Duncan et al. 2007).

e Model Performance

CO measurements retrieved from satellite instruments have been used extensively to assess model performance (Arellano et al. 2007; Bousserez et al. 2007). Twenty six state-of-the-art atmospheric chemistry models have been run to study future air quality and climate change. They were compared to near-global satellite observations from the MOPITT instrument and local surface measurements for CO. The models show large underestimates of northern hemisphere extratropical CO, while typically performing reasonably well elsewhere. The results suggested that year-round emissions, probably from fossil fuel burning in eastern Asia and seasonal biomass burning emissions in southern and central Africa, are greatly underestimated in the current

emission inventories. Variability among models was large, resulting primarily from intermodel differences in representations and emissions of nonmethane volatile organic compounds and in hydrologic cycles, which affect OH and soluble hydrocarbon intermediates (Shindell et al. 2006).

A combination of satellite measurements and models have been used to make a comparison of the chemical nature of air pollution in eastern China and the eastern United States (Tie et al. 2006) highlighting the difference in influence of biogenic and anthropogenic hydrocarbons in both regions. More detailed studies in the same vein have looked at Eastern China (Zhao et al. 2006) showing that during summer, local emissions produce about 50–70% of the O_3 concentration in eastern China. Kar et al. (2008) have used multiple satellite measurement to map the air pollution over the Indo-Gangetic basin.

8.2.4 Formaldehyde, HCHO

HCHO is an important intermediate formed in the oxidation of CH_4 and many other hydrocarbons. The two major loss processes for HCHO, namely photolysis and reaction with OH are relatively fast giving an atmospheric lifetime of typically a few hours. Owing to its high water solubility HCHO can also be removed by wet deposition.

Much of the focus of HCHO measurements from space has been in using it as a proxy measure for isoprene emissions (Palmer et al. 2003). Isoprene can be the dominant precursor of HCHO in regions with strong biogenic emissions and its relatively short lifetime ($<$1 h) means that emissions of the precursor can be localised. The approach has been used to look at seasonal and annual variability in isoprene emissions over the USA (Abbot et al. 2003; Chance et al. 2000; Millet et al. 2008; Palmer et al. 2006), Asia (Fu et al. 2007), Africa (Meyer-Arnek et al. 2005a), Europe (Dufour et al. 2009), tropical regions (Palmer et al. 2007) and globally (Shim et al. 2005). In Asia, the approach was also extended to constrain emissions of alkenes and xylenes as well as the influence of biomass burning (Fu et al. 2007). Work in Europe (Dufour et al. 2009) has demonstrated that the methodology can be extended to regions without "strong" emissions of isoprene.

In combination with other tracers HCHO has been used to trace pollution over the Mediterranean (Ladstatter-Weissenmayer et al. 2003), detect biomass burning over south-eastern Asia (Thomas et al. 1998), quantify the influence of boreal forest fires (Spichtinger et al. 2004), estimate O_3 production sensitivities (Martin et al. 2004a), quantify biogenic and biomass burning budget contributions over Africa (Meyer-Arnek et al. 2005a), constrain tropical tropospheric O_3 (Sauvage et al. 2007) and correlate with ground-based data in the south-eastern USA (Martin et al. 2004b). Space-based HCHO measurements have recently been combined with glyoxal (see Section 8.2.5) measurements (Wittrock et al. 2006). Marbach et al. (2009) to report on enhanced HCHO column density over a ship track in the Indian Ocean. De Smedt et al. (2008) (Fig. 8.5) have derived a 12 year dataset of HCHO from GOME and SCIAMACHY.

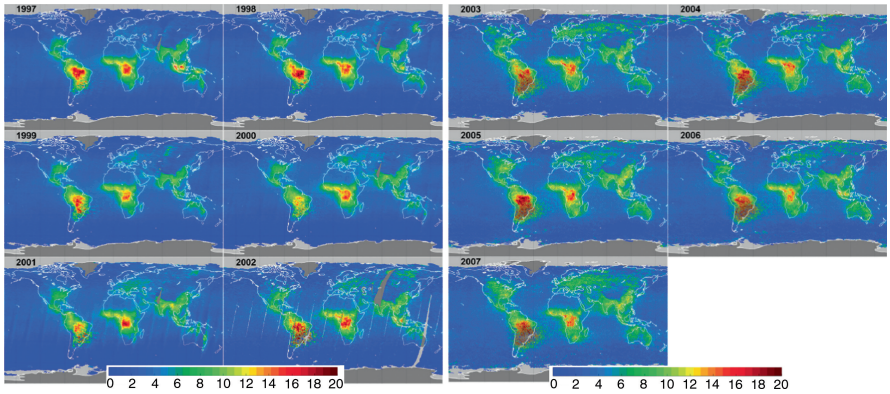

Fig. 8.5 Yearly averaged GOME HCHO columns from 1997 to 2002 (units are 10^{15} molecule/cm^2) (*left*) and SCIAMACHY 2003 to 2007 (*right*) (De Smedt et al. 2008).

HCHO measurements have also been made in the upper troposphere from ACE-FTS and MIPAS (Coheur et al. 2007; Steck et al. 2008). Upper tropospheric HCHO in combination with a range of other trace organics has been used to characterise biomass burning plumes (Coheur et al. 2007).

8.2.5 Glyoxal, CHOCHO

CHOCHO is the smallest α-dicarbonyl, an oxidation product of numerous VOC (Volkamer et al. 2001). Direct and time resolved CHOCHO measurements can provide a useful indicator to constrain VOC oxidation processes (Volkamer et al. 2005) owing to the mixture of anthropogenic and biogenic sources. The atmospheric residence time of CHOCHO is limited by rapid photolysis and reaction with OH radicals, and is about 1.3 h for overhead sun conditions (Volkamer et al. 2005). There are some indications that CHOCHO possibly contributes to secondary organic aerosol (SOA) formation (Liggio et al. 2005a; 2005b; Volkamer et al. 2007). However, the atmospheric relevance of CHOCHO uptake on aerosols is presently not clear. Global observations of CHOCHO from space offer the potential of identifying photochemical hot spots in the Earth's atmosphere, and, coupled with observations of HCHO, improve the understanding of biogenic emissions, biomass burning, and urban pollution.

Wittrock et al. (2006) demonstrated the first measurements of CHOCHO from space. The global pattern of CHOCHO columns was found to be similar to that of HCHO, indicating common atmospheric sources, in particular (biogenic) isoprene. The ratio between CHOCHO and HCHO was found to be about 0.05 in source regions such as the tropical rain forests. At some locations, larger ratios are found and this is attributed to unidentified additional sources of CHOCHO. Large

CHOCHO columns are found primarily over areas having strong biogenic emissions in the tropics which appear to be the dominant global source. During strong biomass burning events, CHOCHO was clearly observed from fires in Alaska (Fig. 8.6)(Wittrock et al. 2006).

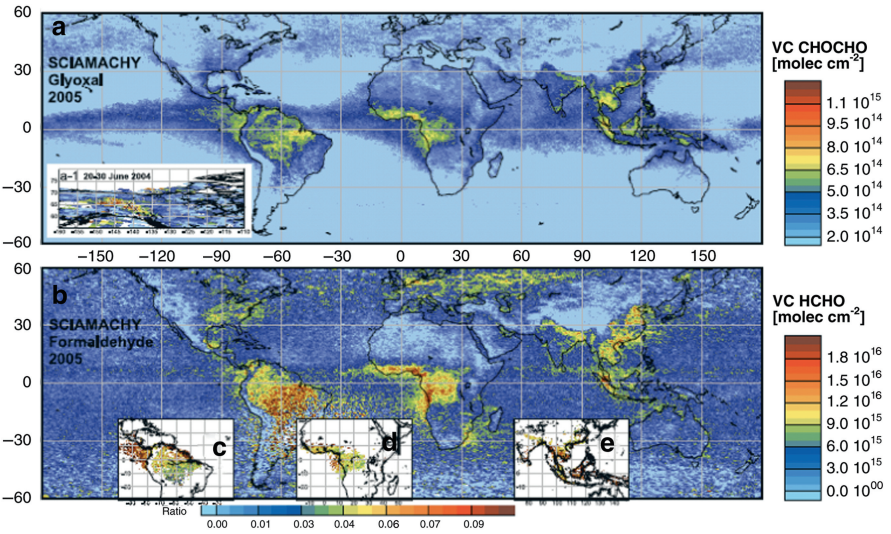

Fig. 8.6 Yearly mean for (**a**) glyoxal and (**b**) formadelyde derived from SCIAMACHY observations in 2005. The sub figures (**c**–**e**) show the ration between glyoxal and formaldehyde (Wittrock et al. 2006).

Five years worth of data has been compiled (Vrekoussis et al. 2009) and the largest columns are seen in tropical and sub-tropical regions associated with high biological activity and the plumes from vegetation fires. The majority of the identified hot spots are characterized by a well-defined seasonality: the highest values being observed during the warm and dry periods as a result of the enhanced biogenic, primarily isoprene, emissions and/or biomass burning from natural or man-made fires. The regions influenced by anthropogenic pollution also encounter enhanced amounts of CHOCHO. There is growing evidence for a potential marine source of CHOCHO (Fu et al. 2008).

8.2.6 Sulfur Dioxide, SO_2

Sulfur chemistry is an integral part of life, owing to its role in plant and human metabolism. Sulfur compounds have both natural and anthropogenic sources in the atmosphere. In modern times, the atmospheric sulfur budget has been dominated by anthropogenic emissions, particularly from fossil fuel burning. It is estimated that 75% of the total sulfur emission budget (ca. 102 Tg S per year) is dominated by

anthropogenic sources with 90% of it occurring in the northern hemisphere. The natural sources include volcanoes, plants, soil and biogenic activity in the oceans.

The oxidation of sulfur compounds in the atmosphere causes a number of different environmental problems such as acidification, climate balance and the formation of a sulfate layer in the stratosphere, the so-called Junge layer. By far the largest sulfur component emitted into the atmosphere is SO_2. Coal and oil combustion contribute up to 80% of the global budget of SO_2 while volcanoes contribute around 10% (Bates et al. 1992).

Satellite measurements of tropospheric SO_2 (and ash) have seen extensive application to identification and quantification of volcanic emissions (Table 8.2). Most of the early measurements (Krueger 1983; Krueger et al. 1995) came from the TOMS instrument whose functionality has been recently extended to OMI (Yang et al. 2007). Authors have demonstrated that the combination of different satellite sensors can give a more holistic chemical and physical picture of volcanic eruptions (Eckhardt et al. 2008; Prata et al. 2007). In particular the use of instruments with sensitivity to the mid-/upper-troposphere have been demonstrated as

Table 8.2 Application of tropospheric satellite SO_2 measurements to volcanic emissions

Volcano	Year(s) of Eruption(s)	Satellite	References
Anatahan	2003	TOMS/ASTER	Wright et al. (2005)
Anatahan	2003–2004	TOMS	Guffanti et al. (2005)
Anatahan	2003	TOMS	Pallister et al. (2005)
Etna	2001	GOME	Zerefos et al. (2006)
Etna	2001 & 2002	GOME	Thomas et al. (2005)
Etna	2002	AIRS	Carn et al. (2005)
El Chichon	1982	TOMS	Seftor et al. (1997)
Galunggung	1982–1983	TOMS	Bluth et al. (1994)
Hekla	1980, 2000	TOMS	Sharma et al. (2004)
Karthala	2005	SEVERI	Prata and Kerkmann, (2007)
Jebel at Tair	2007	IASI	Clarisse et al. (2008)
Jebel at Tair	2007	AIRS/OMI	Eckhardt et al. (2008)
Krafla	1984	TOMS	Sharma et al. (2004)
Manam	2005	OMI/TES	Clerbaux et al. (2008a)
Mauna Loa	1984	TOMS	Sharma et al. (2004)
Miyakejima	2000	ASTER	Urai (2004)
Multiple (20)	1996–2002	GOME	Khokhar et al. (2005)
Nyamuragira	1978–2002	TOMS	Carn and Bluth (2003)
Nyamuragira	1996	GOME	Eisinger and Burrows (1998)
Nyamuragira	2006	OMI; TES	Clerbaux et al. (2008a)
Mt. Spurr	1992	TOMS	Rose et al. (2001)
Mt. St. Helens	1991	TOMS	Bluth et al. (1992)
Pinatubo	1991	TOMS/TOVS	Guo et al. (2004)
Popocatepetl	1996	GOME	Eisinger and Burrows (1998)
Popocatepetl	2000–2001	MODIS	Novak et al. (2008)
Rabual	2006	TES	Clerbaux et al. (2008a)
Redoubt	1989–1990	TOMS	Schnetzler et al. (1994)
Soufriére Hills	2006	OMI/AIRS	Prata et al. (2007)

a way of ascertaining injection height (Ackerman et al. 2008; Prata and Bernardo 2007). The emission of bromine compounds from volcanoes is dealt with in Section 8.2.12. There are practical uses of satellite imagery for protecting international airways from volcanic ash (Tupper et al. 2004).

Owing to the relatively low sensitivity of SO_2 measurements from space there are few reports on anthropogenic sources. Eisinger and Burrows detected widespread SO_2 (Eisinger and Burrows 1998) over south-eastern Europe that they attributed to lignite coal-burning in power plants and these data have been used by Zerefos et al. (2000) to attribute the different sources of SO_2 over Greece. Strong point source emissions such as Peruvian copper smelters (Carn et al. 2007) and a fire at an Iraqi sulfur plant has been identified (Carn et al. 2004). Lee et al. (2008) have used a combination of *in-situ*, ground-based remote sensing and satellite data to follow and attribute the long-range transport of SO_2 from China to Korea, while Krotkov et al. (2008) have used a combination of aircraft and OMI data to validate SO_2 pollution over north-eastern China. Khokhar et al. (2008) have analysed time series of SO_2 from GOME over non-ferrous metal smelters in Peru and Russia.

8.2.7 Ammonia, NH_3

The global emission of NH_3 is about 54 Mt Ny^{-1}. The major global sources are excreta from domestic animals and fertilizers, but oceans, biomass burning and crops are also important (Asman et al. 1998; Schlesinger and Hartley 1992). About 60% of the global NH_3 emission is estimated to come from anthropogenic sources. Boundary layer NH_3 concentrations can vary widely as NH_3 is readily absorbed by surfaces, and reacts with OH and acidic aerosols leading to a rather short atmospheric lifetime in the order of a few hours (Dentener and Crutzen 1994).

The first tropospheric NH_3 measurements from space were made by Beer et al. (2008) using data from TES. NH_3 concentrations over China ranged from 5 to almost 25 ppb, while over North America they were consistently less than 5 ppb. Significant spatial variations were observed over China. The authors note that that the next step is to globalise these observations to understand regional and temporal variations. Recent results from IASI (Clarisse et al. 2009) have demonstrated the first global pictures of NH_3 from space (Fig. 8.7). The strong fires that have occurred in the Mediterranean Basin, and particularly Greece in August 2007, and those in southern Siberia and eastern Mongolia in the early spring of 2008 have shown strong NH_3 biomass burning signatures (Coheur et al. 2009).

8.2.8 Carbon Dioxide, CO_2

CO_2 is the ultimate form of oxidised carbon in the Earth's atmosphere. Much focus in recent times has been on the anthropogenic driven growth in CO_2 concentration and as a source of carbon for plants. In the modern epoch the concentration of CO_2

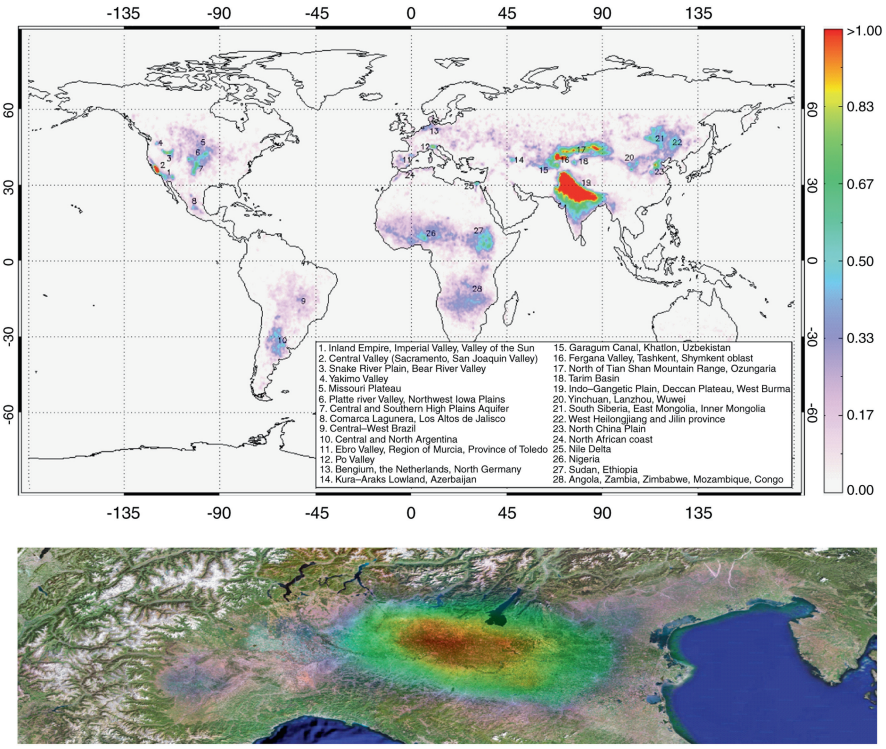

Fig. 8.7 (*Top*) Yearly average total columns of $NH_3(mg/m^2)$ in 2008 retrieved from IASI measurements on a 0.25° by 0.25° grid. (*Bottom*) NH_3 concentrations derived from IASI observations above the Po Valley. The aerial photographs are ©2008 Google – Imagery ©2008 Terrametrics (http://maps.google.com/). Both adapted from Clarisse et al. (2009) (Reprinted by permission of Macmillan Publishers Ltd).

has increased from pre-industrial values of about 280 ppm to values in excess of 380 ppm today. The current concentrations far exceed values inferred from ice cores for the last 650,000 years. The main sinks for atmospheric CO_2 are oceanic and uptake by the terrestrial biosphere.

The first measurements of CO_2 from space (Chedin et al. 2002; 2003; 2005; 2008; Peylin et al. 2007) were mid-tropospheric measurements made in the thermal infrared from TIROS-N/TOVS on NOAA-10. The data were collected for 4 years (1987–1991) and have been used to estimate the influence of tropical biomass burning on mid-tropospheric CO_2 (Chedin et al. 2005; 2008).

AIRS is a thermal infrared spectrometer/radiometer (see Chapters 1 and 3) (Aumann et al. 2003) the data from which a number of authors have used to demonstrate and validate methods for retrieving mid-tropospheric CO_2 (Aumann et al. 2005; Chahine et al. 2005; Crevoisier et al. 2004; Engelen et al. 2004; Engelen and McNally 2005; Maddy et al. 2008; Tiwari et al. 2006).

Buchwitz et al. (2005a; 2005b; 2006) retrieved full tropospheric columns of CO_2 from SCIAMACHY in the short-wave infrared for the first time giving a view of surface CO_2 owing to the enhanced sensitivities at these wavelengths. The data have been used to map out the increasing trend in global CO_2 from space (Buchwitz et al. 2007b; Schneising et al. 2008). Barkley et al. (2006a; 2006b; 2006c; 2007) have extended the Buchwitz methodology looking at assessing validity with a combination of surface, aircraft and model data. Further, they have demonstrated spatial and temporal correlations between SCIAMACHY data more sensitive to surface CO_2 and mid tropospheric data from AIRS over North America (Barkley et al. 2006b) and interesting correlations of CO_2 with surface and vegetation type (Barkley et al. 2006c, 2007) (Fig. 8.8).

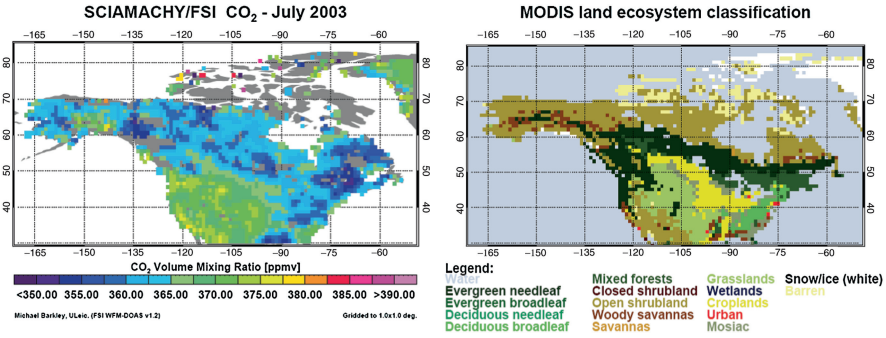

Fig. 8.8 SCIAMACHY CO_2 observations over North America for July 2003 (*left*) with a map of land vegetation cover over this scene (*right*) (Barkley et al. 2006c). The transition from low CO_2 VMRs along the Canadian shield and the eastern coast to the higher values over the mid-western US corresponds to a change in the vegetation type from evergreen needle leaf, mixed and deciduous broadleaf forests to land covered by crops and large grass plains.

Much of the effort in measurement of tropospheric CO_2 from space has been focussed on the precision and accuracy of the measurements, owing to the overwhelming need to use these measurements in an inverse mode to assess emissions. The measurements are being supplemented by new ones from specially designed sensors *i.e*. GOSAT (Hamazaki et al. 2004) and from the second version of OCO (the first instrument was lost in launch) (Crisp et al. 2004).

8.2.9 Methane, CH_4

CH_4 is the second most important anthropogenic greenhouse gas. It also has an indirect effect on climate through chemical feedbacks. More than 50% of present-day global CH_4 emissions are anthropogenic, the largest contributors being fossil fuel production, ruminants, rice cultivation, and waste handling (Bergamaschi et al. 2007). CH_4 levels have almost doubled since preindustrial times. The natural source

strength of CH_4, mainly constituted by emission from wetlands, is particularly uncertain, because these emissions vary considerably in time and space. The dominant atmospheric sink for CH_4 is reaction with OH. Available ground-based measurements are sparse, albeit precise, and limitedly representative at larger scales (Walter et al. 2001). Better knowledge of CH_4 distribution and emissions is indispensable for a correct assessment of its impact on global change.

Measurements of CH_4 from SCIAMACHY show the expected latitudinal gradient (NH 10% > SH) with large-scale patterns of anthropogenic and natural CH_4 emissions (Buchwitz et al. 2005a; 2005b; Frankenberg et al. 2005a; 2006; Schneising et al. 2009; Straume et al. 2005).

Frankenberg et al. (2005a; 2006) using SCIAMACHY data observed pronounced enhancements of CH_4 in the Red Basin of China and in large areas of Asia in general, followed by northern parts of South America and central Africa. In particular, high mixing ratios in Columbia and Venezuela are larger than given by the CH_4 inventories. The largest seasonal variations are caused by rice emissions in Asia, which are very intense during a relatively short time period. The measurements indicate that these emissions already start towards the end of July and decline sharply in November, which is earlier than predicted by the model based inventories. Strong deviations between observed and modelled CH_4 abundances in tropical rainforest regions are observed, hinting at underestimated tropical emissions (Frankenberg et al. 2005a; 2006). This observation has been much debated in the context of the occurrence and role of plants in the production of CH_4 (Dueck and van der Werf 2008). Frankenberg et al. (2008) have recently reinvestigated the relevant CH_4 spectroscopy which may have introduced some biases into the initial retrievals. Separate data from Schneising et al. (2009) still show higher CH_4 over the tropics compared to a model. Upper tropospheric CH_4 profiles have been retrieved from a number of satellites including most recently ACE-FTS and MIPAS (De Maziere et al. 2008).

8.2.10 Water, H_2O

H_2O and the hydrological cycle are key elements of the earth system. As well as being key to life on Earth it is a major part of the climate system. More than 99% of the atmospheric H_2O is found and exists in all three phases in the troposphere. H_2O enters the atmosphere by evaporation mostly over the oceans, but also by transpiration from plants over the continents. The evaporation flux is balanced by the return of H_2O to the surface by various forms of precipitation. In the vapour phase, H_2O can be transported several thousands of kilometres before condensing which leads to greater precipitation than evaporation over the continents, the net balance being provided by surface flow of H_2O. The lifetime of a H_2O molecule in the atmosphere is estimated to be about 10 days.

Noel et al. (1999; 2002) have retrieved total H_2O column measurements from GOME-1 using DOAS in the 700 nm region and Maurellis et al. (2000) in the

585–600 nm region. Such column data have been validated against microwave measurements from SSM/I. Wagner et al. (2005; 2006) have used a similar approach to look at trends in total column precipitable water over the period 1996–2003. In general, the trends showed a strong correlation with near surface temperature though not always over land in the northern hemisphere (Wagner et al. 2006). Mieruch et al. (2008) have observed increasing water trends in GOME-1 data for Greenland, eastern Europe, Siberia and Oceania, whereas decreasing trends have been observed for the northwest USA, central America, Amazonia, central Africa and the Arabian Peninsula. Lang et al. (2007) have evaluated the GOME water climatology against independent *in-situ* measurements from the operational WMO radio-sonde network, against high spatial resolution H_2O vapour columns from MERIS and with ERA40 model results.

The GOME-1 H_2O column series from UV/VIS data have been extended to SCIAMACHY (Noel et al. 2004; 2005) and more recently the operational GOME-2 instrument (Noel et al. 2008). Recently, nadir measurements from TES have been used to retrieve water profiles (Shephard et al. 2008). A study of the 2006 El Nino (Logan et al. 2008) showed a movement in H_2O vapour owing to eastward movement of convection during El Nino leading to higher H_2O over eastern Africa and the western Indian Ocean. Similar results have been found by Wagner et al. (2005) for H_2O column anomalies for the El Nino 1997/1998 in GOME data.

Isotopologue measurements of H_2O are becoming available *e.g.* HDO (Herbin et al. 2007; Steinwagner et al. 2007; Zakharov et al. 2004; Frankenberg et al. 2009) (see Section 8.2.16). The use of such measurements are explained in Monks et al. (2009).

Microwave retrievals of H_2O vapour are dealt with in Chapter 4.

8.2.11 Bromine Monoxide, BrO

In comparison to the chemistry taking place in the stratosphere where halogen chemistry is well known and characterised, there has been much debate as to the role of halogen species in the oxidative chemistry of the troposphere (Platt and Hönninger 2003). There is growing experimental evidence about the prevalence of halogen chemistry as part of tropospheric photochemistry (Platt and Hönninger 2003; Read et al. 2008; Saiz-Lopez et al. 2007b; Simpson et al. 2007). Much of the proposed halogen chemistry is propagated through the reactions of a series of halogen atoms and radicals (Monks 2005).

BrO species can be formed in the polar boundary layer (Barrie et al. 1988; Fan and Jacob 1992; Saiz-Lopez et al. 2007b), mid-latitude marine boundary layer (Saiz-Lopez et al. 2004; 2006), salt pans (Hebestreit et al. 1999) as well as in volcanic emissions (Bobrowski et al. 2003). There are claims that BrO is omnipresent in the atmosphere (Hegels et al. 1998).

The major source of gas-phase bromine in the lower troposphere is thought to be the release of species such as IBr, ICl, Br_2 and BrCl from sea-salt aerosol, following the uptake from the gas-phase and subsequent reactions of hypohalous acids (HO_X,

where X = Br, Cl, I) (Vogt et al. 1996). The halogen release mechanism is autocatalytic (Fan and Jacob 1992) and has become known as the "bromine explosion" (Platt and Lehrer 1997). The initiation of the BrO chemistry is relatively simple:

$$\mathrm{BrX} + \mathrm{h}\nu \rightarrow \mathrm{Br} + X \tag{8.1}$$

$$\mathrm{Br} + \mathrm{O_3} \rightarrow \mathrm{BrO} + \mathrm{O_2} \tag{8.2}$$

The chemistry of halogen radicals has been recently reviewed by Monks (Monks 2005) and the impact of BrO chemistry by Simpson et al. (2007) and von Glasow et al. (2004).

The space-based measurements have clearly illustrated the spatial extent of the BrO in the Antarctic and Arctic boundary layer in spring (Richter et al. 1998; Wagner et al. 2001; Wagner and Platt 1998) (See Fig. 8.9). Various authors using the space-based data have investigated trends (Hollwedel et al. 2004), long-range transport of BrO_x (Ridley et al. 2007), the correlation with surface remote sensing measurements (Theys et al. 2007; Wagner et al. 2007), assessment with hydrocarbon-loss methods (Zeng et al. 2006), the role of the marginal ice zone (Jacobi et al. 2006) and total columns over Arrival Heights (Antarctica) during sunrise (Schofield et al. 2006).

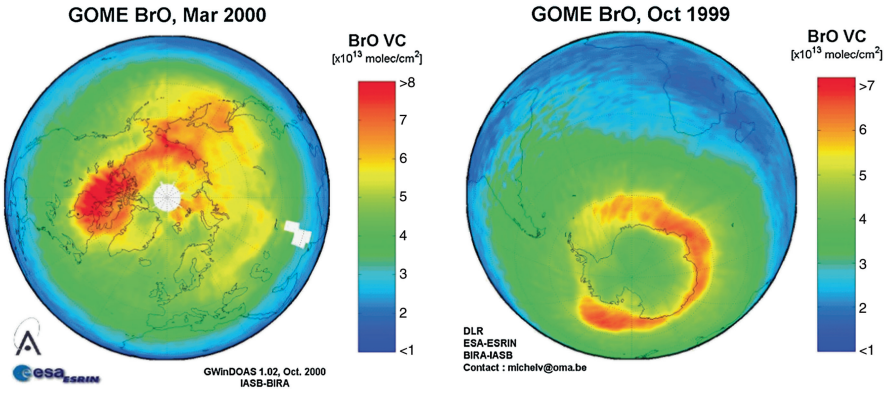

Fig. 8.9 BrO in the Antarctic and Arctic during spring derived from GOME satellite measurements (Van Roozendael et al. 2002).

BrO has been detected, probably in the free troposphere, outside polar regions such as over the Maldives (Ladstatter-Weissenmayer et al. 2007a), mid-latitudes (Van Roozendael et al. 2002) and globally (Hegels et al. 1998). The impact of these observations on, for example, the O_3 budget of the troposphere (von Glasow et al. 2004) remains an open question.

Space based measurements have been used to survey the BrO emissions from volcanoes (Afe et al. 2004). There is some debate as to whether there is an

expectation that the BrO emissions should be correlated with SO_2 emissions (Afe et al. 2004). An unambiguous detection of volcanic BrO was possible for the 2008 eruption of the Kasatochi volcano in Alaska (Theys et al. 2009).

8.2.12 Iodine Monoxide, IO

IO has been detected by ground-based measurements both in the marine boundary layer (Alicke et al. 1999; Allan et al. 2000), the Dead sea (Zingler and Platt 2005) and the Antarctic boundary layer (Friess et al. 2001; Saiz-Lopez et al. 2007b). The major sources of iodine into the troposphere are thought to be from macroalgal sources releasing either molecular iodine and/or organoiodine compounds (Carpenter 2003). Photolysis of I_2 and the organo-iodine compounds releases the iodine.

$$\mathrm{RI} + \mathrm{h}\nu \rightarrow \mathrm{R} + \mathrm{I} \tag{8.3}$$

$$\mathrm{I} + \mathrm{O_3} \rightarrow \mathrm{IO} + \mathrm{O_2} \tag{8.4}$$

During daylight hours IO exists in a fast photochemical equilibrium with iodine viz:

$$\mathrm{IO} + \mathrm{h}\nu \rightarrow \mathrm{I} + \mathrm{O} \tag{8.5}$$

The aerosol "explosion" mechanism, previously described for bromine, acts effectively to recycle the iodine back to the gas-phase (Platt and Hönninger 2003). The potential impact of this chemistry has been demonstrated by Read et al. (2008) who have shown evidence for widespread destruction of tropical O_3 by bromine and iodine monoxides.

Saiz-Lopez et al. (2007a) and Schönhardt et al. (2007) first derived IO from SCIAMACHY measurements. There are significant differences between the two retrievals with Saiz-Lopez et al. observing IO above cloudy regions whereas Schönhardt et al. see negligible IO. Schönhardt et al. (2008) have produced the first global pictures of IO. Figure 8.13 shows the global measurements of IO and Fig. 8.10 shows measurements over Antarctica.

From space, the largest amounts of IO have been detected in springtime over the Antarctic. The seasonal variation of IO in Antarctica showed high values in springtime, slightly less IO in the summer period and again larger amounts in autumn (Schönhardt et al. 2008). In winter, no elevated IO levels were found in the areas accessible to the satellite measurements. The observed satellite seasonal cycle is in good agreement with recent ground-based measurements in Antarctica (Friess et al. 2001; Saiz-Lopez et al. 2007b). Conversely, in the Arctic region, no elevated IO levels were detected in the period analysed. Schönhardt et al. (2008) suggest that this observation is evidence for different conditions with respect to iodine release in the two polar regions.

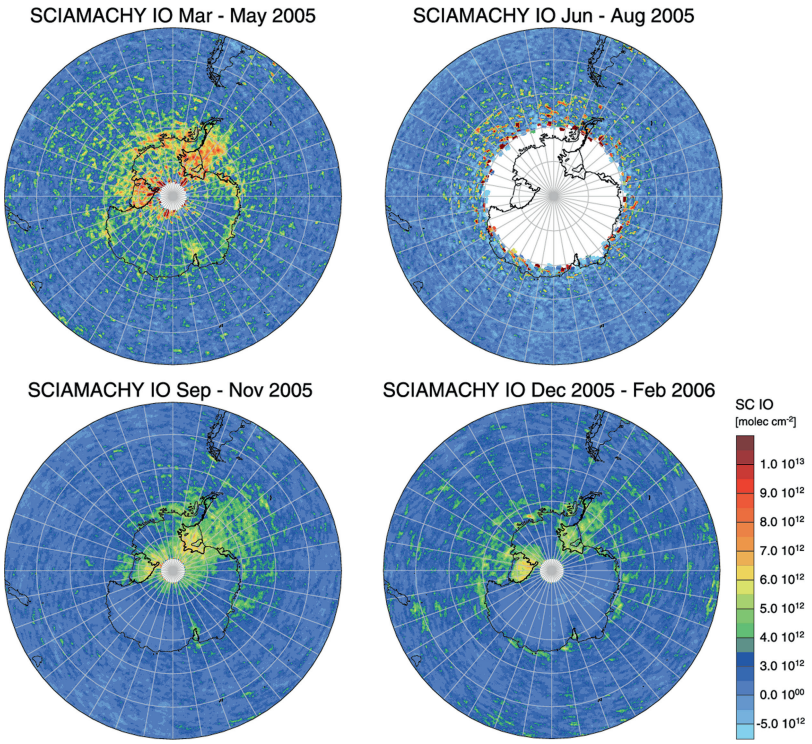

Fig. 8.10 Seasonally averaged slant column amounts of IO above the southern hemisphere from Antarctic autumn to summer (Schönhardt et al. 2008). Maxima of IO columns occur over the Weddell Sea, the Ross Sea and along the coast especially in spring and in autumn with levels remaining positive at some places throughout the summer.

8.2.13 Methanol, CH_3OH

CH_3OH is the most abundant oxygenated hydrocarbon gas in the atmosphere and is therefore a major contributor to non-methane volatile organic compounds, NMVOC (Singh et al. 1995). The primary source of atmospheric CH_3OH is plant growth and decay, the second largest source is atmospheric production with minor sources from biomass burning and anthropogenic emissions (Jacob et al. 2005). There is considerable uncertainty in the atmospheric CH_3OH budget. In the remote troposphere, CH_3OH concentrations are 0.1–1 ppb (Singh et al. 1995) while the concentrations in the continental boundary layer are an order of magnitude larger. The atmospheric lifetime is about 16 days in the free troposphere owing, primarily, to OH oxidation to produce HCHO (Singh et al. 1995).

The first lower troposphere measurements of CH_3OH have been demonstrated by Beer et al. (2008) from TES. Upper tropospheric measurement of CH_3OH have been made by IR occultation from ACE-FTS (see Fig. 8.11) (Dufour et al. 2006; 2007). Dufour have shown the utility of the limb measurements of CH_3OH measurements

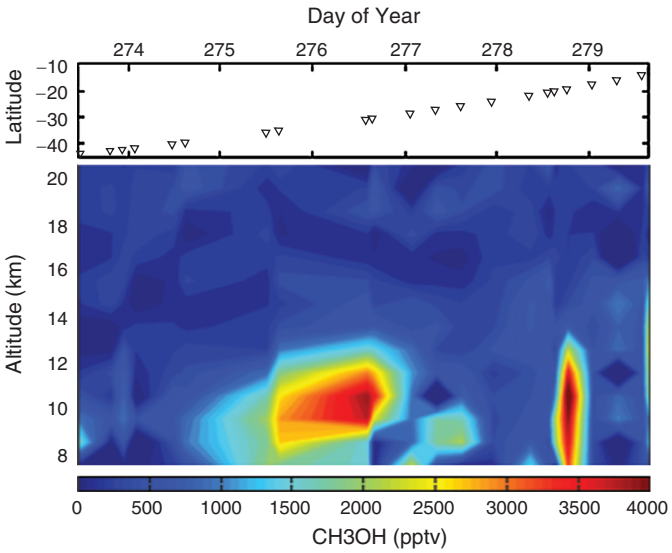

Fig. 8.11 Time series of ACE-FTS measurements of methanol for sunsets between 30th September and 6 October 2004 (Dufour et al. 2006). The latitude and time of each individual measurement are shown at the *top* of the panel.

in the upper troposphere for quantifying surface budgets and the influence of biomass burning (Dufour et al. 2007).

8.2.14 Nitrous Oxide, N_2O

N_2O is the fourth largest single contributor to positive radiative forcing and serves as a long-lived marker of the anthropogenic influence of the N-cycle. Atmospheric concentrations have risen by 16% since pre-industrial times to a value of around 319 ppbV. N_2O is also the main source of NO_x to the stratosphere. N_2O is mainly produced by microbial nitrification and denitrification processes in soils and water. Owing to its long atmospheric lifetime (*ca.* 1 century) the mixing ratio of N_2O shows very little spatial variation, $<1\%$, throughout the troposphere.

There have been relatively limited measurements of full tropospheric column N_2O from space. Estimates in validation exercises suggest precisions in the order of 20% (Piters et al. 2006). Comparisons of early products with FTIR measurements showed agreement within 13% (Dils et al. 2006). Measurements on a ship-borne FTIR campaign showed a strong deviation (*ca.* 30%) between the satellite and ship based data (Warneke et al. 2005). All of these studies make the point that the datasets are not large enough to draw any statistical significance and that there is room for further improvement in the retrievals.

8.2.15 *Nitric Acid,* HNO_3

HNO_3 is the end point of NO_x chemistry in the atmosphere (see also Section 8.2.2) (Monks 2005). The dominant HNO_3 sink is wet removal (contributing to acid deposition) and dry deposition.

First global distributions of HNO_3 in the troposphere and the stratosphere derived from IR satellite measurements have been measured recently from IMG-ADEOS (Wespes et al. 2007) (Fig. 8.12).

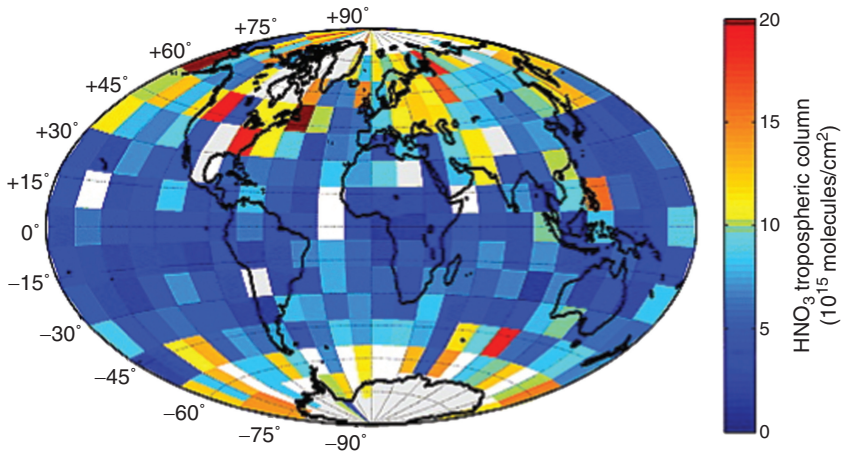

Fig. 8.12 Global distribution of HNO_3 in the troposphere (0–10 km), in 10^{15} molecules/cm^2. Data are averaged on a 15° × 12° grid (Wespes et al. 2007).

There are also several upper tropospheric measurements (Table 8.3). A number of satellite measurements including those of HNO_3 have been used for space-based constraints on the production of NO_2 by lightning (Martin et al. 2007).

8.2.16 *Other Trace Species*

There are a plethora of organic compounds in the atmosphere. The atmospheric budget is controlled by a combination of anthropogenic and natural (frequently biogenic) emissions tensioned against atmospheric chemical loss processes (photolysis or reaction with atmospheric oxidants) or physical loss (mainly heterogeneous removal). Many of these organic compounds are intrinsically linked to the chemistry that controls the global oxidising ability. A range of mainly small lightweight hydrocarbons have been measured in the upper troposphere such as C_2H_2 (Park et al. 2008; Rinsland et al. 2005; 2007b), C_3H_4 (Coheur et al. 2007) and C_2H_6 (Clarmann et al. 2007; Park et al. 2008; Rinsland et al. 2005; 2007b). The oxygenated compounds measured in the upper troposphere have been acetone

Table 8.3 Tropospheric trace gases measured from space. A full list of tropospheric satellite instruments and their characteristics is given in Appendix A

Gas	Instrument	Reference
Ozone	ACE-FTS[a]	Hegglin et al. (2008)
	GOME-1	Del Frate et al. (2005), Hansen et al. (2003), Hoogen et al. (1999), Iapaolo et al. (2007), Ladstatter-Weissenmayer et al. (2004, 2007c), Lamsal et al. (2004), Liu et al. (2006b, 2007), Meijer et al. (2006), Meyer-Arnek et al. (2005b), Muller et al. (2003), Munro et al. (1998), Tellmann et al. (2004), Valks et al. (2003), van der A et al. (2002)
	HIRDLS[a]	Nardi et al. (2008)
	IMG	Coheur et al. (2005)
	OMI	Choi et al. (2008), Martin et al. (2007), Schoeberl et al. (2007), Stajner et al. (2008), Ziemke et al. (2006)
	MIPAS[a]	Clarmann et al. (2007), Fischer et al. (2008), Raspollini et al. (2006)
	MLS[a]	Froidevaux et al. (2008), Ziemke et al. (2006)
	SCIAMACHY	Geer et al. (2006), Rozanov et al. (2007)
	TES	Choi et al. (2008), Eremenko et al. (2008), Jourdain et al. (2007), Logan et al. (2008), Nassar et al. (2008), Osterman et al. (2008), Richards et al. (2008), Worden et al. (2007a, b), Bowman et al. (2009)
	TOMS	Ahn et al. (2003), Browell et al. (1996), Chandra et al. (1998, 1999, 2002, 2003, 2004), Chatfield et al. (2004), Creilson et al. (2003, 2005), Cros et al. (1992), Edwards et al. (2003), Fishman, (1991a, b), Fishman and Balok (1999), Fishman and Brackett, (1997), Fishman et al. (1987, 1990, 1991, 1992, 2002, 2003, 2005), Hudson et al. (1995), Hudson and Thompson, (1998), Kim et al. (1996), Morris et al. (2006), Nganga et al. (1996), Olson et al. (1996), Peters et al. (2002, 2004), Portmann et al. (1997), Richardson et al. (1991), Thompson and Hudson (1999), Thompson et al. (1993, 1996a, 1996b, 2000, 2001, 2003), Tie et al. (2007), Vukovich et al. (1996, 1997), Watson et al. (1990), Ziemke and Chandra, (1999, 2003a, b), Ziemke et al. (1998, 2000, 2001, 2003, 2005)
NO_2	GOME-1	Beirle et al. (2003, 2004a, b, c, 2006), Blond et al. (2007), Boersma et al. (2005), Choi et al. (2005), Chun'e and Baoning (2008), Damoah et al. (2004), Edwards et al. (2003), Franke et al. (2009), Guerova et al. (2006), Han et al. (2009), Hayn et al. (2009), Hild et al. (2002), Ionov et al. (2006), Irie et al. (2005), Jaegle et al. (2004, 2005), Konovalov et al. (2005), Kunhikrishnan et al. (2004a, b), Ladstatter-Weissenmayer et al. (2003, 2007c), Lauer et al. (2002), Leue et al. (2001), Ma et al. (2006), Martin et al. (2002, 2003, 2004b), Meyer-Arnek et al. (2005a), Muller and Stavrakou (2005), Ordonez et al. (2006), Poberovskii et al. (2007), Richter and Burrows, (2002), Richter et al. (2005), Sauvage et al. (2007), Savage et al. (2004),

		Schaub et al. (2005), (2007), Spichtinger et al. (2001, 2004), Stohl et al. (2003), Thomas et al. (1998, 2003), Timofeev et al. (2000), Toenges-Schuller et al. (2006), Uno et al. (2007), van der A et al. (2006, 2008), van Noije et al. (2006), Velders et al. (2001), Wang et al. (2007a), Wenig et al. (2003), Zhang et al. (2007), Zhao et al. (2006)
	MIPAS[a]	Fischer et al. (2008), Raspollini et al. (2006)
	OMI	Boersma et al. (2007, 2008a, 2009), Bucsela et al. (2006, 2008), Celarier et al. (2008), Choi et al. (2008), Kim et al. (2009), Kramer et al. (2008), Lamsal et al. (2008), Mijling et al. (2009), Wang et al. (2007b), Wenig et al. (2008)
	OSIRIS[a]	Sioris et al. (2007)
	SCIAMACHY	Bertram et al. (2005), Boersma et al. (2008b), Bousserez et al. (2007), Franke et al. (2009), He et al. (2007), Heue et al. (2005), Irie et al. (2005), Kaynak et al. (2009), Kim et al. (2006, 2009), Konovalov et al. (2006, 2008), Martin et al. (2007), Richter et al. (2004, 2005), Stavrakou et al. (2008), van der A et al. (2008), Zhang et al. (2007)
	GOME-2	Franke et al. (2009)
N_2O	MIPAS[a]	Fischer et al. (2008), Raspollini et al. (2006)
	SCIAMACHY	Dils et al. (2006), Warneke et al. (2005)
NH_3	MIPAS[a]	Burgess et al. (2006a)
	TES	Beer et al. (2008)
	IASI	Clarisse et al. (2009)
CO	ACE[a]	Clerbaux et al. (2005, 2008c), Folkins et al. (2006a), Hegglin et al. (2008), Rinsland et al. (2005, 2007b, 2008), Turquety et al. (2008)
	AIRS[a]	McMillan et al. (2005), Stohl et al. (2007), Warner et al. (2007), Yurganov et al. (2008), Zhang et al. (2008)
	IASI	Clerbaux et al. (2009)
	IMG	Barret et al. (2005), Turquety et al. (2004)
	MAPS	Connors et al. (1999), Lamarque et al. (1999), Newell et al. (1999)
	MLS[a]	Barret et al. (2008), Duncan et al. (2007), Filipiak et al. (2005), Livesey et al. (2008), Park et al. (2007), Pumphrey et al. (2007)
	MOPITT	Allen et al. (2004), Arellano et al. (2004, 2006, 2007), Bhattacharjee et al. (2007), Bousserez et al. (2007), Bowman (2006), Bremer et al. (2004), Choi and Chang, (2006a, b), Choi et al. (2005), Clerbaux et al. (2008b, c), Crawford et al. (2004), Deeter et al. (2003, 2004, 2007a, b), Edwards et al. (2003, 2004, 2006a, b), Emmons et al. (2004, 2007), Generoso et al. (2007),

(*continued*)

Table 8.3 (continued)

Gas	Instrument	Reference
		Gros et al. (2004), Guerova et al. (2006), Heald et al. (2003, 2004), Ho et al. (2005), Hyer et al. (2007a, b), Ito et al. (2007), Jacob et al. (2003), Kampe and Sokolik (2007), Kar et al. (2004, 2006, 2008), Kim et al. (2005), Lamarque et al. (2003), Lee et al. (2005), Li et al. (2005), Liang et al. (2007), Lin et al. (2007), Liu et al. (2005, 2006a), Luo et al. (2007b), Massie et al. (2006), Peng and Zhao (2007), Petron et al. (2004), Pfister et al. (2004, 2005), Pradier et al. (2006), Rinsland et al. (2006b), Shindell et al. (2005, 2006), Singh et al. (2006), Stavrakou and Muller (2006), Tanimoto et al. (2008), Tie et al. (2006), Turquety et al. (2007, 2008), Velazco et al. (2005), Wang et al. (2006), Warner et al. (2007), Yudin et al. (2004), Yumimoto and Uno (2006), Yurganov et al. (2005, 2008), Zhao et al. (2006, 2007), Zheng et al. (2004)
	SCIAMACHY	Buchwitz et al. (2004, 2005b, 2006, 2007a), de Laat et al. (2006, 2007), Dils et al. (2006), Frankenberg et al. (2005b), Gloudemans et al. (2005, 2006, 2008), Khlystova et al. (2009), Straume et al. (2005), Turquety et al. (2008), Warneke et al. (2005)
	TES	Bowman et al. (2009), Logan et al. (2008), Luo et al. (2007a, b), Rinsland et al. (2006b, 2008), Warner et al. (2007), Zhang et al. (2008)
CO_2	AIRS[a]	Aumann et al. (2005), Barkley et al. (2006b), Chahine et al. (2005), Crevoisier et al. (2004), Engelen et al. (2004), Engelen and McNally (2005), Maddy et al. (2008)
	SCIAMACHY	Barkley et al. (2006a, b, c, 2007), Bosch et al. (2006), Buchwitz et al. (2005a, b, 2007b), Dils et al. (2006)
	NOAA-10 [TIROS-N, TOVS][a]	Chedin et al. (2002, 2003, 2005, 2008), Peylin et al. (2007)
CH_4	ACE[a]	De Maziere et al. (2008), Rinsland et al. (2007b)
	AIRS[a]	Xiong et al. (2008)
	IMG	Clerbaux et al. (2003), Turquety et al. (2004)
	MIPAS[a]	De Maziere et al. (2008), Fischer et al. (2008), Raspollini et al. (2006)
	SCIAMACHY	Bergamaschi et al. (2007), Buchwitz et al. (2005a, b, 2006), Dils et al. (2006), Frankenberg et al. (2005a, 2006), Gloudemans et al. (2005, 2008), Straume et al. (2005), Bloom et al. (2010)
methanol (CH_3OH)	ACE[a]	Coheur et al. (2007), Dufour et al. (2006, 2007), Rinsland et al. (2007b)
	TES	Beer et al. (2008)

HCHO	ACE[a]	Coheur et al. (2007)
	GOME	Abbot et al. (2003), Barkley et al. (2008b), Chance et al. (2000), De Smedt et al. (2008), Fu et al. (2007), Ladstatter-Weissenmayer et al. (2003, 2007c), Marbach et al. (2009), Martin et al. (2004a, b), Meyer-Arnek et al. (2005b), Millet et al. (2006), Palmer et al. (2001, 2003, 2006, 2007), Sauvage et al. (2007), Shim et al. (2005), Spichtinger et al. (2004), Thomas et al. (1998)
	MIPAS[a]	Steck et al. (2008)
	OMI	Millet et al. (2008)
	SCIAMACHY	De Smedt et al. (2008), Dufour et al. (2009), Wittrock et al. (2006)
glyoxal (CHOCHO)	SCIAMACHY	Fu et al. (2008), Vrekoussis et al. (2009), Wittrock et al. (2006)
H_2O	ACE-FTS[a]	Hegglin et al. (2008)
	GOME-1	Lang et al. (2002, 2007), Maurellis et al. (2000), Mieruch et al. (2008), Noel et al. (1999, 2002), Wagner et al. (2005, 2006)
	GOME-2	Noel et al. (2008)
	MIPAS[a]	Milz et al. (2005)
	MLS[a]	Read et al. (2007)
	SCIAMACHY	Mieruch et al. (2008), Noel et al. (2004, 2005)
	TES	Logan et al. (2008), Shephard et al. (2008)
SO_2	AIRS[a]	Carn et al. (2005), Eckhardt et al. (2008), Prata and Bernardo (2007), Prata et al. (2007)
	ASTER[a]	Urai (2004)
	GOES/HIRS[a]	Ackerman et al. (2008)
	GOME-1	Eisinger and Burrows (1998), Khokhar et al. (2005), Thomas et al. (2005), Zerefos et al. (2000, 2006)
	IASI	Clarisse et al. (2008)
	OMI	Carn et al. (2007), Eckhardt et al. (2008), Krotkov et al. (2006, 2008), Prata et al. (2007), Yang et al. (2007)
	SCIAMACHY	Lee et al. (2008)
	SEVERI	Eckhardt et al. (2008), Prata and Kerkmann (2007)
	TES	Clerbaux et al. (2008a)
	TOMS	Bluth et al. (1992, 1994), Carn and Bluth (2003), Carn et al. (2004), Guffanti et al. (2005), Guo et al. (2004), Gurevich and Krueger (1997), Krotkov et al. (1997), Krueger et al. (1995),

(*continued*)

Table 8.3 (continued)

Gas	Instrument	Reference
		Massie et al. (2004), Novak et al. (2008), Pallister et al. (2005), Rose et al. (2001), Schnetzler et al. (1994), Seftor et al. (1997), Sharma et al. (2004), Tupper et al. (2004), Wright et al. (2005)
	TOVS	Guo et al. (2004)
BrO	GOME	Afe et al. (2004), Chance (1998), Hegels et al. (1998), Hollwedel et al. (2004), Ladstatter-Weissenmayer et al. (2007b), Richter et al. (1998), Ridley et al. (2007), Schofield et al. (2006), Van Roozendael et al. (2002), Wagner et al. (2001, 2007), Wagner and Platt (1998), Zeng et al. (2006)
	SCIAMACHY	Afe et al. (2004), Jacobi et al. (2006), Theys et al. (2009), Theys et al. (2007)
IO	SCIAMACHY	Saiz-Lopez et al. (2007a), Schönhardt et al. (2008)
OCS	ACE-FTS[a]	Barkley et al. (2008a), Rinsland et al. (2007b)
acetone	ACE-FTS[a]	Coheur et al. (2007)
PAN	ACE-FTS[a]	Coheur et al. (2007)
	MIPAS[a]	Glatthor et al. (2007)
acetylene (C_2H_2)	ACE-FTS[a]	Park et al. (2008), Rinsland et al. (2005, 2007b), Turquety et al. (2008)
ethane ($C_2H_{6)}$	ACE-FTS[a]	Coheur et al. (2007), Park et al. (2008), Rinsland et al. (2005, 2007b), Senten et al. (2008), Turquety et al. (2008)
	MIPAS[a]	Clarmann et al. (2007)
ethene (C_2H_4)	ACE-FTS[a]	Coheur et al. (2007), Herbin et al. (2009)
SF_6	ACE-FTS[a]	Rinsland et al. (2007b)
	MIPAS[a]	Burgess et al. (2004, 2006b)
CFC-11	ACE-FTS[a]	Mahieu et al. (2008)
	IMG	Coheur et al. (2003)
	MIPAS[a]	Coheur et al. (2003), Hoffmann et al. (2005, 2008)

CFC-12	ACE-FTS[a]	Mahieu et al. (2008)
	IMG	Coheur et al. (2003)
	MIPAS[a]	Hoffmann et al. (2005)
CFC-113	ACE-FTS[a]	Dufour et al. (2005)
HCFC-142b	ACE-FTS[a]	Dufour et al. (2005)
HCFC-22	MIPAS[a]	Moore and Remedios (2008)
	IMG	Coheur et al. (2003)
methyl chloride (CH_3Cl)	ACE-FTS[a]	Rinsland et al. (2007b)
formic acid (HCOOH)	ACE-FTS[a]	Rinsland et al. (2006a, 2007b)
HDO	IMG	Herbin et al. (2007), Zakharov et al. (2004)
	MIPAS[a]	Steinwagner et al. (2007)
	SCIAMACHY	Frankenberg et al. (2009)
HNO_3	ACE-FTS[a]	Martin et al. (2007), Wolff et al. (2008)
	HIRDLS[a]	Kinnison et al. (2008)
	IMG	Wespes et al. (2007)
	MIPAS[a]	Fischer et al. (2008), Raspollini et al. (2006)
	MLS[a]	Santee et al. (2007)
peroxy nitric acid (HO_2NO_2)	MIPAS[a]	Stiller et al. (2007)
HCN	ACE-FTS[a]	Park et al. (2008), Rinsland et al. (2005, 2007b), Turquety et al. (2008)
HCl	ACE-FTS[a]	Mahieu et al. (2008), Rinsland et al. (2007b), Senten et al. (2008)
H_2O_2	ACE-FTS[a]	Rinsland et al. (2007a)

[a]upper troposphere only

(CH_3COCH_3) (Coheur et al. 2007), formic acid (HCOOH) (Rinsland et al. 2006a; 2007b) and H_2O_2 (Rinsland et al. 2007a). Oxidised and reduced nitrogen compounds measured in the upper troposphere are peroxyacetylnitrate (PAN) (Coheur et al. 2007; Glatthor et al. 2007), HO_2NO_2 (Stiller et al. 2007), HCN (Park et al. 2008; Rinsland et al. 2005; 2007b). Upper tropospheric inorganic compounds such as OCS (Barkley et al. 2008a; Rinsland et al. 2007b), SF_6 (Rinsland et al. 2007b), HCl (Mahieu et al. 2008; Senten et al. 2008) and HDO (Herbin et al. 2007; Steinwagner et al. 2007; Zakharov et al. 2004) have also been measured.

There are a range of CFC and HCFC compounds that have been measured in the upper troposphere, namely, CFC-11 (Coheur et al. 2003; Hoffmann et al. 2005, 2008; Mahieu et al. 2008), CFC-12 (Coheur et al. 2003; Hoffmann et al. 2005; Mahieu et al. 2008), CFC-113 (Coheur et al. 2003; Dufour et al. 2005), HCFC-142b (Dufour et al. 2005) and HCFC-22 (Coheur et al. 2003; Moore and Remedios 2008). Moore and Remedios (2008) used a combination of two satellite systems to derive a mean northern hemisphere mid-latitude (20–50°N) HCFC-22 growth rate between 1994 and 2003 of 5.4 $\pm$ 0.7 pptv per year and a mean southern hemisphere growth rate (60–80°S) of 6.0 $\pm$ 0.7 pptv per year in the same period.

There have been important applications of upper tropospheric trace organics to mapping the age and composition of biomass burning plumes (Coheur et al. 2007; Dufour et al. 2006; Rinsland et al. 2005). H_2O_2 has been detected in young biomass burning plumes in the tropics (Rinsland et al. 2007a). A HCOOH emission factor relative to CO of 1.99 $\pm$ 1.34 g/kg during 2004 in upper tropospheric biomass burning plumes is inferred from a comparison with lower mixing ratios measured during the same time period (Rinsland et al. 2006a). Upper tropospheric CO, C_2H_6, HCN, CH_3Cl, CH_4, C_2H_2, CH_3OH, HCOOH, and OCS measurements show enhancement in biomass burning plumes of up to 185 ppbv for CO, 1.36 ppbv for C_2H_6, 755 pptv for HCN, 1.12 ppbv for CH_3Cl, 0.178 ppbv for C_2H_2, 3.89 ppbv for CH_3OH, 0.843 ppbv for HCOOH, and 0.48 ppbv for OCS in western Canada and Alaska at 50°–68°N latitude between 29 June and 23 July 2004 (Rinsland et al. 2007b).

Enhancements of C_2H_6 and O_3 observed in the southern hemisphere have been attributed to a biomass burning plume, which covers wide parts of the southern hemisphere, from South America, the Atlantic Ocean, Africa, the Indian Ocean to Australia. The chemical composition of the part of the plume-like pollution belt associated with South American fires, where rainforest burning is predominant, appears to differ from the part of the plume associated with southern African savannah burning. In particular, African savannah fires lead to a larger O_3 enhancement than equatorial American fires (Clarmann et al. 2007). At the end of the biomass burning season in South America and South and East Africa, elevated PAN amounts of 200–700 pptv were measured in a large plume extending from Brazil over the South Atlantic, central and South Africa, the southern Indian Ocean as far as Australia at altitudes between 8 and 16 km (Glatthor et al. 2007).

Park et al. (2008) have used speciated data to investigate the role of the Asian monsoon as a transport barrier from the upper troposphere to the lower stratosphere, the range of chemical tracers being able to tag different air masses origins and quantify photochemical lifetimes.

8.3 Satellite Observations of Tropospheric Composition: What Can We Learn?

Satellite observations have for the first time provided simultaneous global measurements of several important tropospheric trace gases. These datasets open new horizons in atmospheric science. Characteristic spatial and temporal patterns allow the identification, location, and quantification of different sources to study phenomena such as transport or trends.

The comparison of satellite measurements with CTMs/GCMs provides a substantive challenge to the models. The measurements help to constrain the models and the comparisons indicate the shortcomings of both measurements and models. The applications involving models are discussed in Chapter 9. However, in the following sections we highlight the potential of satellite based trace gas measurements to investigate several central questions in tropospheric chemistry research.

8.3.1 Column Density Maps as Proxies for Emissions

Global maps of tropospheric trace gases, as shown in Fig. 8.13, impressively illustrate the power of tropospheric satellite measurements. The spatial patterns of column densities are determined by (1) sources of the respective trace gas, (2) transport and (3) the lifetime. Enhanced column densities indicate source regions, where spatial patterns are most distinct for short lifetimes.

Thus, to start with a straightforward application, column densities serve as a first-order proxy of emissions. For example, this is particularly distinct in the case of NO_2, owing to the sensitivity of UV/VIS satellite measurements to the lower troposphere and the short lifetime of NO_x, of only a few hours, in the boundary layer, leading to high spatial gradients (Fig. 8.13b). As a consequence, besides the regions of high population density paired with industrial activity, like the US East coast, western Europe, or eastern China, where column densities are generally high, Megacities like Los Angeles, Mexico City, Moscow, Seoul, or Tokyo, clearly show up as “hot spots”. Even emissions from ships can be recognized for the highly frequented route between Sri Lanka and Indonesia (Beirle et al. 2004a; Richter et al. 2004). The emissions from other NO_x sources, *i.e.* biomass burning, soil emissions, and lightning, are less localized and the respective columns are therefore smeared out over large areas in a multi-annual mean.

Similarly, different sources can be identified in the global maps for other trace gases as well. Anthropogenic emissions can also clearly be seen in the maps for CO (Fig. 8.13c) and SO_2 (Fig. 8.13f), particularly over eastern Asia, though the lifetimes of these gases are generally higher than for NO_x, and signals are thus less localized. Nevertheless, maps of mean SO_2 show sharp peaks over copper smelters in Peru (Fig. 8.14). Also recent maps of NH_3 (see Fig. 8.7) show global distributions in relation to strong source regions.

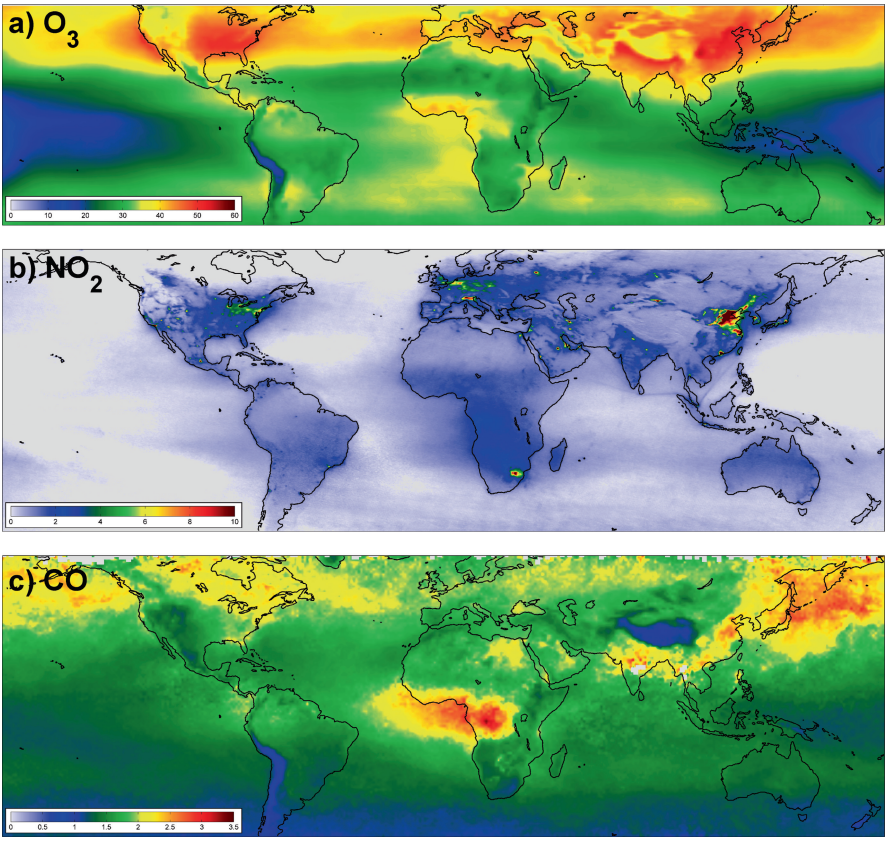

Fig. 8.13 Atlas of tropospheric species observed from space. (**a**) Mean summer column density of tropospheric O_3 (DU) from TOMS measurements 1979–2005. Data provided by Jack Fishman (Fishman et al. 2003). http://asd-www.larc.nasa.gov/TOR/TOR_Data_and_Images.html. (**b**) Mean tropospheric column density of NO_2 (10^{15} molecules/cm^2) derived from SCIAMACHY measurements 2003–2007. Data provided by Steffen Beirle. (**c**) Mean column density of CO (10^{18} molecules/cm^2) derived from nighttime IASI measurements July 2008. Data provided by Matthieu Pommier/Cathy Clerbaux.

There are particularly strong enhancements of SO_2 after volcanic eruptions (Khokhar et al. 2005; Thomas et al. 2005; Yang et al. 2007), as illustrated in Fig. 8.14b (note the change in colour scale compared to a). Satellite measurements can be used to monitor volcanoes remotely (see http://www.gse-promote.org/) and give information on the extent and location of plumes, this information being important for air traffic routing.

Analogous highly sporadic enhancements can be observed for strong biomass burning episodes in a wide range of tracers such as CO, NO_2, HCHO, and CHOCHO (Thomas et al. 1998; Edwards et al. 2006; Wittrock et al. 2006). For example, Fig. 8.15 shows the enhancement of CO from the Alaskan forest fires in July 2004 (see also Section 8.2.3).

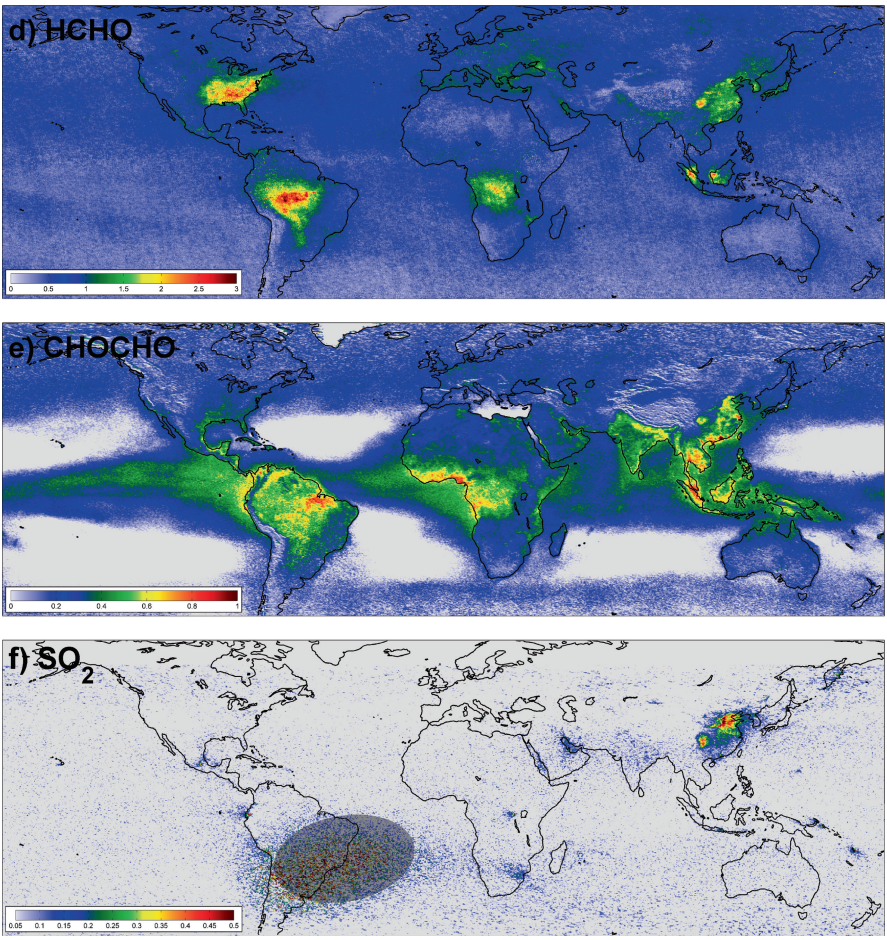

Fig. 8.13 (continued) Atlas of tropospheric species observed from space. (**d**) Mean column density of HCHO (10^{16} molecules/cm^2) derived from OMI August 2006. Data provided by Thomas Kurosu. (**e**) Mean glyoxal (CHOCHO) column density (10^{15} molecules/cm^2) from SCIAMACHY measurements 2003–2007. Data provided by Mihalis Vrekoussis (Vrekoussis et al. 2009). (**f**) Mean column density of SO_2 (DU) from SCIAMACHY measurements 2007. The shaded area over South America/South Atlantic masks improper fit results owing to the South Atlantic Anomaly (SAA). Data provided by Andreas Richter.

The origin, amount and impact of halogen oxides on tropospheric composition is still highly debated (Platt and Hönninger 2003; von Glasow et al. 2004; Monks et al. 2009). The detection of tropospheric BrO (Wagner and Platt 1998; Richter et al. 1998) and IO (Saiz-Lopez et al. 2007a; Schönhardt et al. 2008) from space were milestones for polar tropospheric chemistry research, proving the existence of halogen oxides over extended areas in polar spring (Fig. 8.9) and indicating where and when *in-situ* measurements should be performed for in-depth analysis of the basic chemical mechanisms.

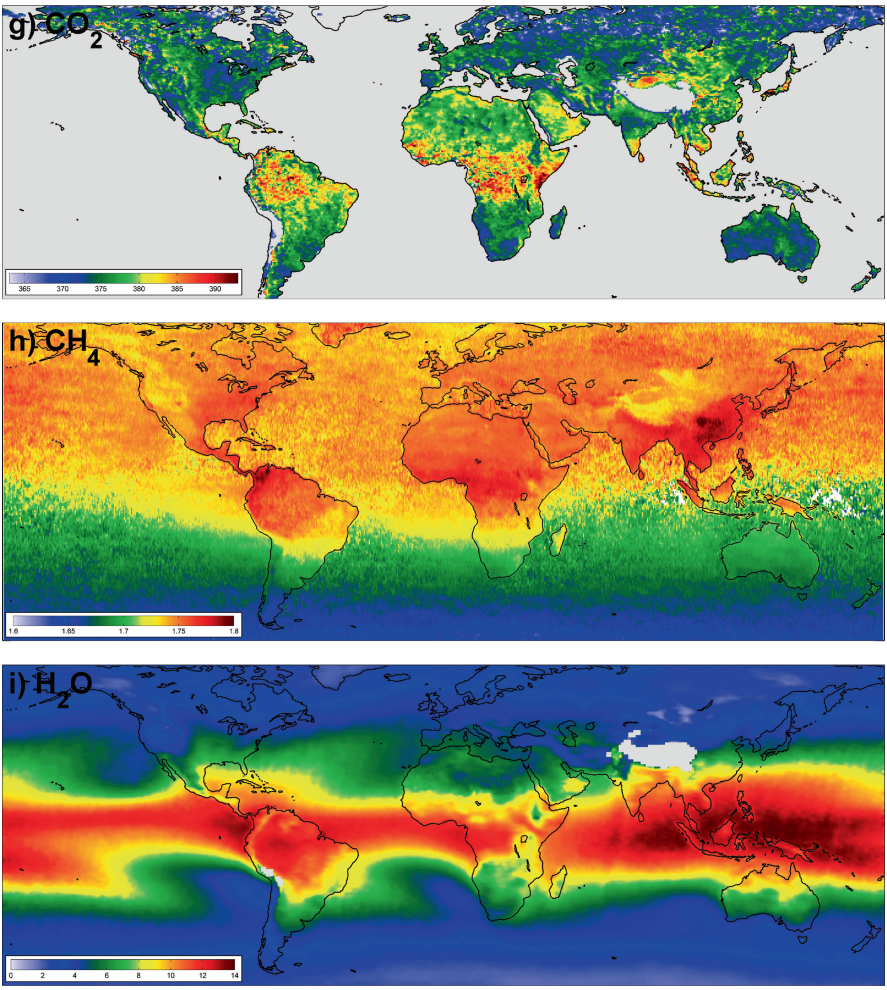

Fig. 8.13 (continued) Atlas of tropospheric species observed from space. (**g**) Mean tropospheric mixing ratio (ppm) of CO_2 derived from SCIAMACHY measurements in May 2003. Data provided by Michael Buchwitz (Buchwitz et al. 2007b). (**h**) Mean tropospheric mixing ratio (ppm) of CH_4 derived from SCIAMACHY measurements 2004. Data provided by Christian Frankenberg (Frankenberg et al. 2008). (**i**) Mean tropospheric column density of H_2O (10^{22} molecules/cm^2) derived from GOME measurements 1996–2004. Data provided by Thomas Wagner (Wagner et al. 2006).

In the cases of some VOCs observable from space, in particular HCHO and CHOCHO, (Fig. 8.13d, e), the secondary production from photochemical degradation exceeds direct emissions. Enhanced column measurements thus serve as proxy for the emissions of precursors and indicators of photochemistry (Millet et al. 2008; Palmer et al. 2003; 2006). For example, enhanced HCHO columns over south-eastern US are used as a quantitative proxy for biogenic isoprene emissions (Palmer et al. 2006).

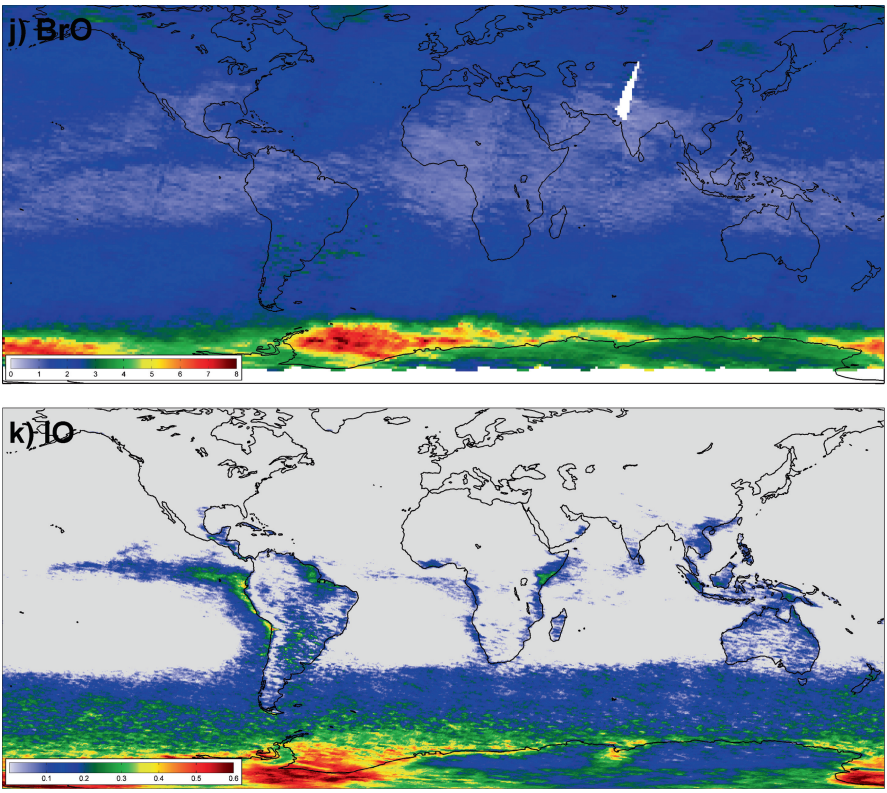

Fig. 8.13 (continued) Atlas of tropospheric species observed from space. (**j**) Mean tropospheric column density of BrO (10^{13} molecules/cm^2) derived from GOME measurements September 1997. Data provided by Nicolas Theys/Michel van Roozendael. (**k**) Mean tropospheric column density of IO (10^{13} molecules/cm^2) derived from SCIAMACHY measurements Sep–Nov 2005. Data provided by Anja Schönhardt (Schönhardt et al. 2008).

The global pattern of tropospheric O_3 shown in Fig. 8.13a reflects the availability of O_3 precursors, *i.e.* NO_x and VOCs. Highest columns being found over eastern Asia and the US East coast. The enhancement over the western Atlantic from Africa is because of the presence of the compounds (see CO, HCHO, CHOCHO), NO_x from biomass burning and NO_x from lightning (Martin et al. 2007).

From the spatial distribution alone as measured from space, on individual days or temporally averaged, it is possible to learn about sources of different trace gases. Consequently, satellite measurements have been used to estimate or constrain emissions in several studies (Arellano et al. 2004 (CO); Bergamaschi et al. 2007 (CH_4); Leue et al. 2001; Martin et al. 2003; Jaegle et al. 2005 (NO_2)) on a global scale, as well as in numerous studies focusing on particular regions and/or source types. For such inversion studies, knowledge of lifetimes and transport is needed, which is provided by chemical transport or general circulation models, to link emissions to columns. The link of satellite observations to chemistry models is

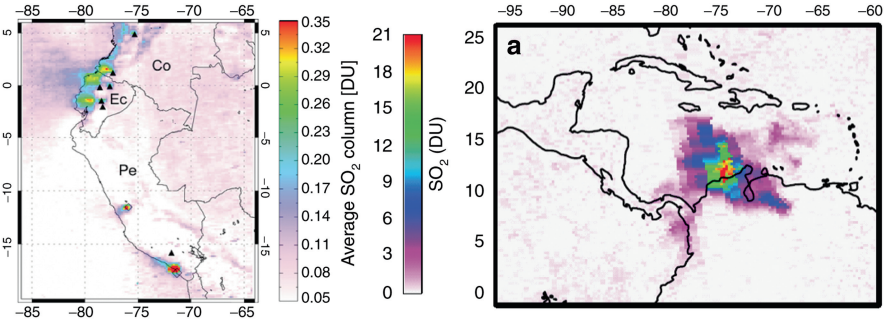

Fig. 8.14 (**a**) Average SO_2 column amounts measured by OMI over southern Colombia (Co), Ecuador (Ec), and Peru (Pe) between 1st September 2004 and 30th June 2005. Triangles mark Volcanoes, while Peruvian copper smelters are indicated by diamonds (Carn et al. 2007). (**b**) Observed OMI SO_2 column over the volcanic plume emitted from Soufriere Hills Volcano (Montserrat; 16.72°N, 62.18°W) on 21st May 2006 (Yang et al. 2007).

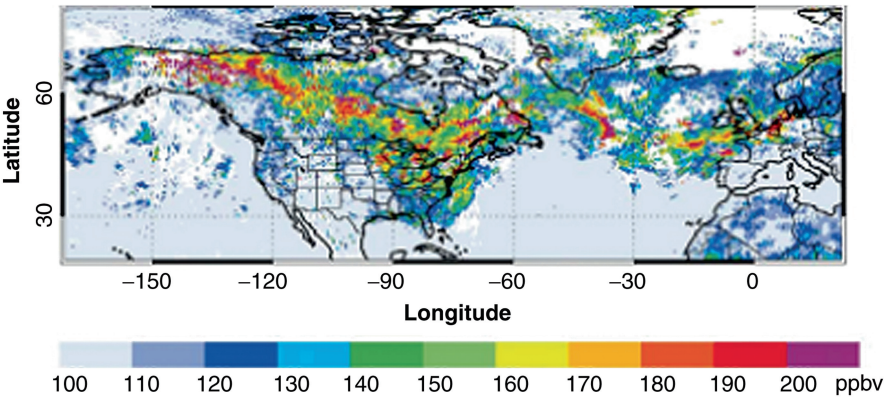

Fig. 8.15 MOPITT 700-hPa CO mixing ratio for 15–23 July 2004 (Fishman et al. 2008).

discussed in Chapter 9. However, independent lifetime information can also be gained from the satellite measurements themselves from spatial patterns downwind the sources (Beirle et al. 2004a).

8.3.2 Monitoring Transport and Circulation

The availability of temporally consecutive global measurements allows an investigation of transport of various trace gases for individual episodes as well as in terms of predominant transport patterns. Satellite observations not only reveal the location and strength of sources, but also the fate of the different trace gases visible from space. In addition, transport patterns (in particular of trace gases with lifetimes of weeks to months) serve as tracers for the validation of transport models. In

particular, CO is a good marker for long range transport (LRT) owing to its lifetime of a few months (see Section 8.2.3).

Local emissions from industry or biomass burning can contribute appreciably to levels in remote regions. However, even close to sources, the question arises as to what fraction is due to these local emissions, and how much is due to long-range/intercontinental transport. For instance, Liang et al. (2007) reported that the Asian influence on pollution levels observed in the free troposphere over North America in summertime contributes about 7%. Heald et al. (2003) and Zhang et al. (2008) report on Trans-Pacific transport events of CO (Fig. 8.16), suggesting that similar LRT episodes affect North American levels of O_3. Gloudemans et al. (2006) found evidence for long-range transport of CO from biomass burning in the southern hemisphere using SCIAMACHY measurements, concluding that South American biomass burning emissions contribute up to 30–35% of the CO levels over Australian biomass burning regions.

The transport of SO_2 plumes from volcanic eruptions can be investigated from a time series of satellite maps (Prata et al. 2007) (Fig. 8.27). Such volcanic plumes can travel thousands of km over several days. The heavy eruption of the Kasatochi volcano in Alaska on 7th August 2008 led to the first detection of volcanic BrO in satellite spectra. The BrO plume could be tracked over a time period of about 6 days (Theys et al. 2009) (Fig. 8.17).

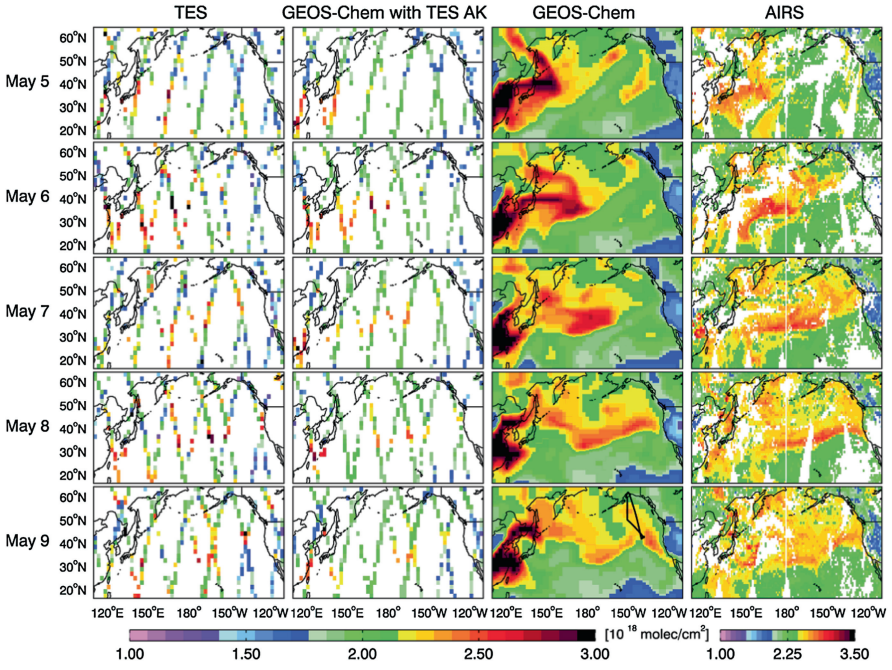

Fig. 8.16 CO columns from AIRS, TES and the GEOS-Chem model during the transpacific Asian pollution event, 5th–9th May, observed by the INTEX-B aircraft (Zhang et al. 2008).

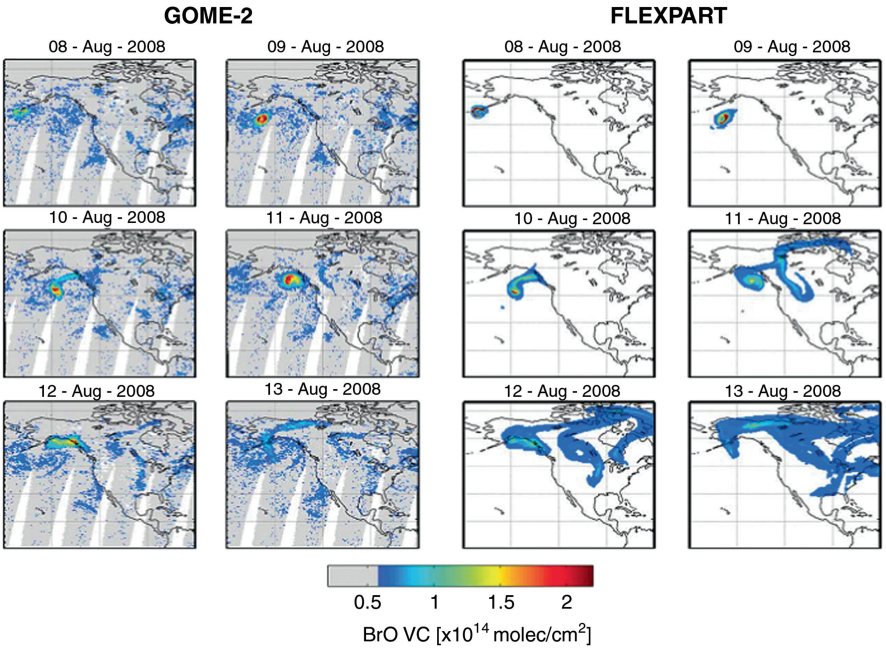

Fig. 8.17 GOME-2 measurements of BrO total columns after the eruption of the Kasatochi volcano and corresponding FLEXPART column simulations (Theys et al. 2009).

In the case of NO_2, LRT is unusual owing to the short lifetime of NO_x in the boundary layer, but it occurs occasionally when the boundary layer pollution is lifted to the upper troposphere, where the NO_x lifetime can reach several days (Wenig et al. 2003). In addition, PAN acts as reservoir for NO_x and plays an important role for the LRT of nitrogen oxides (Singh et al. 1992).

A prominent example of intercontinental transport of an anthropogenic NO_x plume within a meteorological "bomb" from the US East coast to Europe within five days was reported by Stohl et al. (2003) (Fig. 8.18).

A statistical analysis of transport patterns of NO_x is given in Eckhardt et al. (2003) where NO_2 distributions for high and low NAO (North Atlantic Oscillation) index are compared. For high NAO, NO_2 levels over northern Europe are significantly higher than for low NAO in winter, owing to the changes in wind patterns. An increase in NAO as reported by Hurrell (1995) implies an increase in O_3 precursors in the Arctic. Similarly, Creilson et al. (2003) report a positive correlation of NAO indices and tropospheric O_3 columns over Europe due to transport of O_3 precursors from North America to Europe.

Thus, analysis of range, frequency and significance of transport events permits us to verify our knowledge about atmospheric processes, in particular the lifetimes of different trace gases. Satellite measurements are also a powerful tool to investigate the question of how far regional pollution levels are local or impacted by LRT (Keating and Zuber 2007).

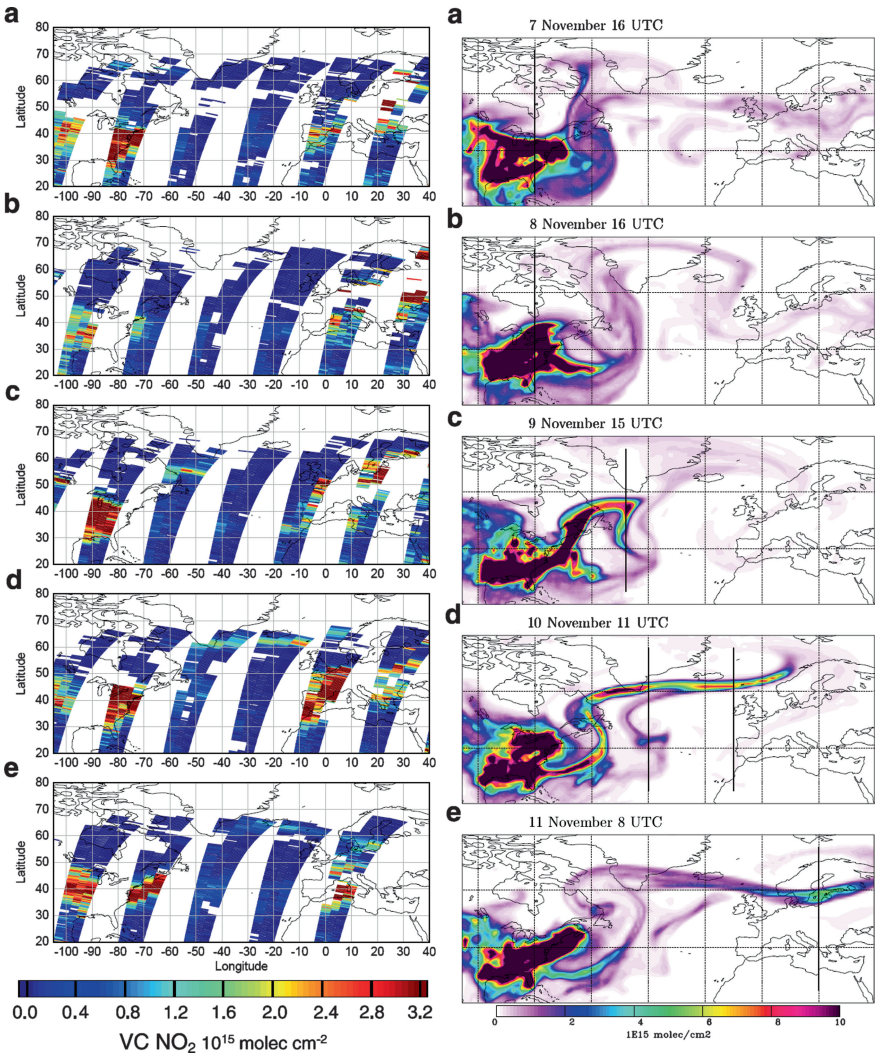

Fig. 8.18 Transport event of NO_2 from North America to Europe in November 2001 (Stohl et al. 2003). *Left*: TVCDs of NO_2 (GOME). *Right*: Total columns of the FLEXPART NO_x tracer.

8.3.3 Trends

There is now more nearly 15 years worth of tropospheric column measurements particularly in the UV/Vis spectral range. Using these data, studies on long-term temporal changes or trends of a range of different trace gases are becoming possible.

Fig. 8.13b shows that China is the region with the highest observed NO_2 columns in the world. There has been a tremendous growth in industrial activity during recent

years. The pattern was different some years ago when it was dominated by the U.S. and Europe (Leue et al. 2001). Richter et al. (2005) first studied the trend of NO_2 columns and found a strong increase of 50% over 8 years in the Chinese region, while NO_2 columns over Europe and parts of the U.S. decreased (Fig. 8.19). Similar trend studies have been performed by Irie et al. (2005), Konovalov et al. (2008), Stavrakou et al. (2008), van der A et al. (2008), Hayn et al. (2009) and Kim et al. (2006) focussed on changes in NO_2 over U.S. power plants and found significant reductions owing to the implementation of pollution controls by utility companies in the eastern U.S.

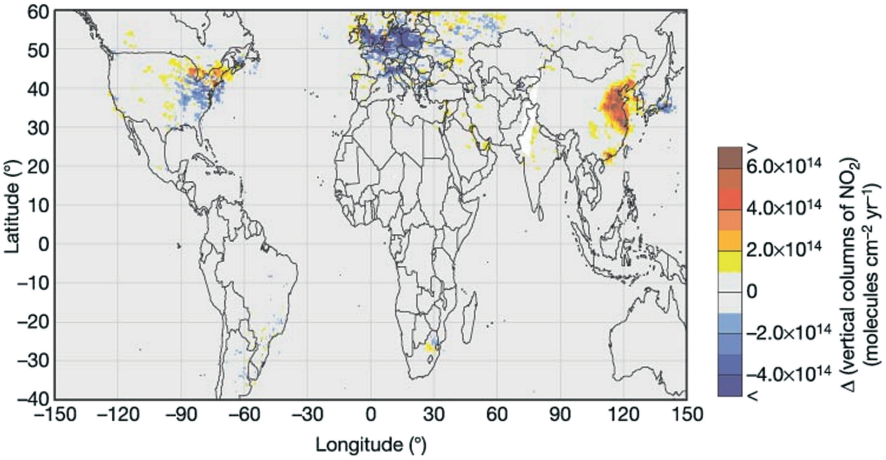

Fig. 8.19 Average annual changes in tropospheric NO_2 as observed by GOME from 1996 to 2002 (Richter et al. 2005 (Reprinted by permission of Macmillan Publishers Ltd)).

The increase of NO_2 columns over China illustrates the need for the updating of the relevant emission inventories on annual basis. Models using outdated emission inventories cannot be expected to simulate atmospheric chemistry realistically.

Similar trend investigations have been performed for CO (Yurganov et al. 2008) and SO_2 (Khokhar et al. 2008). Results are not as clear as with NO_x emissions over China; nevertheless, Yurganov et al. (2008) find an increase of global CO of about 2% per year for the second half of the year from MOPITT measurements between 2000 and 2006, while for the first half of the year, no significant change was detected. Ongoing measurements and an increasing number of CO sensors will allow more detailed regional studies to be performed in the near future. Khokhar et al. (2008) report on a decrease of SO_2 columns of 25% from 1996 to 2002 over the copper smelter Ilo in Peru, derived from GOME measurements.

For O_3, the time-series are available from 1979. However, the determination of trends of tropospheric O_3 is difficult owing to the dominant stratospheric column that also changes. Nevertheless, Ziemke et al. (2005) report an increase of tropospheric O_3 of 2 to 3 DU per decade for mid-latitudes for both hemispheres, while for

the tropics, no significant change could be found, as also reported by Thompson and Hudson (1999).

The combustion of fossil fuels has lead to a large increase in atmospheric CO_2 levels, which is overlaid by the seasonal variation imprinted by the uptake and release of CO_2 by plants. This is impressively documented in the long-time series of CO_2 mixing ratios over Mauna Loa, Hawaii (Keeling et al. 1976). Buchwitz et al. (2007b) present a good match for the Mauna Loa time-series with SCIAMACHY northern hemispheric mean columns, and show an appreciable increase in CO_2 even for the relatively short time-series available from SCIAMACHY (Fig. 8.20).

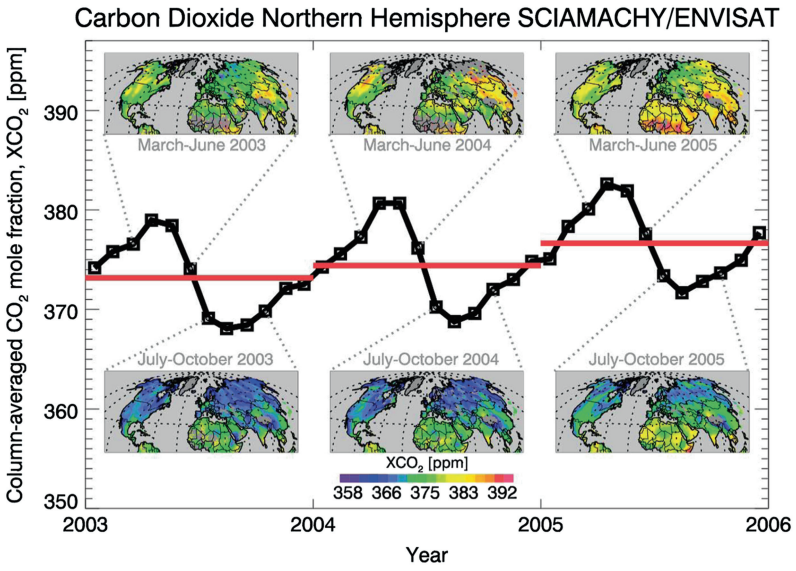

Fig. 8.20 Atmospheric CO_2 over the northern hemisphere from 2003 to 2005 as retrieved from SCIAMACHY satellite measurements (Buchwitz et al. 2007b).

As well as trace gases that are directly affected by anthropogenic emissions, water vapour, a natural greenhouse gas, is subject to changes owing to climate change. Since water vapour is strongly coupled to temperature, global warming should enhance water vapour columns, establishing a positive feedback mechanism. From GOME measurements, an increase of total column precipitable water of about 3% per year is reported by Wagner et al. (2006), in parallel with the increase in surface temperature, and hence indicates a strong positive climate feedback for water vapour. Similar results are found by Mieruch et al. (2008).

A continuous time series of satellite measurements will allow us to monitor future changes of various trace gases, and so to check the efficiency of measures taken to reduce air pollution. Satellite measurements have the potential to verify compliance of international climate protocols such as the Kyoto Treaty.

8.3.4 Periodical Temporal Patterns

As well as longer-term trends, periodical temporal patterns can be analyzed. Annual cycles are the usual dominant patterns within a time series of atmospheric trace gases owing to changes in emissions, chemistry and/or viewing conditions of the same periodicity.

The annual cycle of CO reflects the emissions during tropical biomass burning seasons (Edwards et al. 2006a; 2006b; Frankenberg et al. 2005b). Similar response of biomass burning emissions can be found in measurements of NO_2, HCHO or CHOCHO (Jaegle et al. 2005; Stavrakou et al. 2009; Myriokefalitakis et al. 2008). Palmer et al. (2006) investigated seasonal variability of isoprene emissions over North America using satellite measurements of HCHO. On shorter time scales, the clear coincidence of the onset of precipitation with enhanced NO_2 column densities was used to identify and estimate soil emissions of NO_x (Jaegle et al. 2004; Bertram et al. 2005).

Tropospheric BrO in polar regions shows an annual peak in hemispheric spring (Fig. 8.9), which is an important clue to its origin (Monks 2005). The specific annual cycle indicates that newly-formed sea ice plays an important role in the heterogeneous release of bromine leading to the bromine explosion. This could either be directly or indirectly due to the formation of highly saline frost flowers or aerosols resulting from frost flowers (Kaleschke et al. 2004).

Van der A et al. (2008) determined maps of the month with highest NO_2 column density and, with simple assumptions on yearly cycles of different NO_x sources, derived maps of the dominant NO_x source (Fig. 8.21).

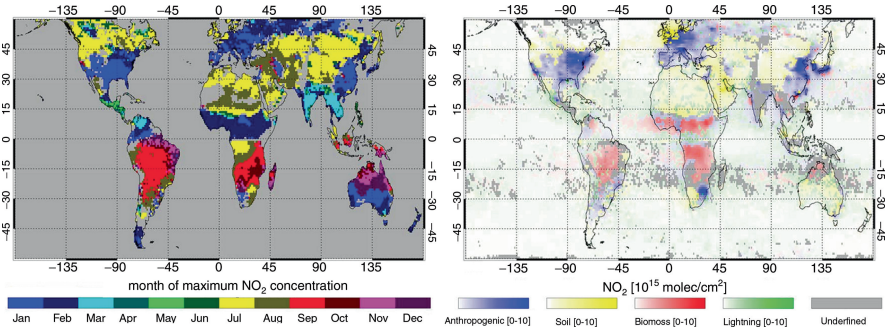

Fig. 8.21 (*Left*) Month of maximum NO_2; (*Right*) Dominant NO_x source identification (van der A et al. 2008).

Anthropogenic emissions in most industrialized countries follow a weekly cycle with emission reductions on Sundays. This is reflected in the weekly pattern of NO_2 column densities (Fig. 8.22): A clear reduction of NO_2 columns on Sundays can be found for the United States, Europe, South Korea and Japan, while columns reach a minimum on Friday in the Middle East and Saturday in Israel. In China, no weekly cycle is observed.

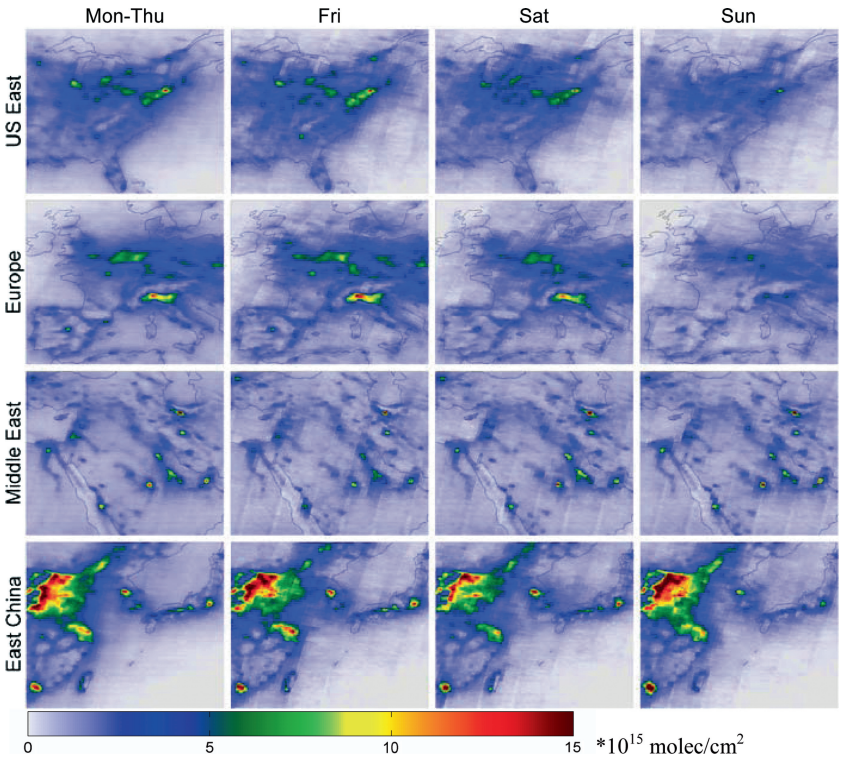

Fig. 8.22 Weekly cycle of NO_2 column densities for the United States, Europe, the Middle East, and eastern Asia (SCIAMACHY measurements 2003–2007).

The observation of a weekly cycle helps to discriminate between different NO_x sources that show different reductions, such as traffic and power generation (Beirle et al. 2003; Kaynak et al. 2009). In addition, analysis of the complete weekly pattern holds information on the NO_x lifetime. For example, Monday levels of NO_2 over Germany are significantly lower than Tuesday levels in winter, since Monday "inherits" comparably clean Sunday air owing to the longer lifetime of about 1 day (Beirle et al. 2003).

8.3.5 Synergistic Use of Different Measurements

Several years of satellite measurements and a growing number of instruments in space have resulted in extensive datasets providing information on many atmospheric trace gases. At the same time, there is considerable additional information on clouds, aerosols, or ground albedo which is of importance for the quantitative interpretation of column measurements from space. Furthermore, satellite measurements of other

quantities such as lightning flashes, fires, night-time light pollution or vegetation indices, can serve as proxies for various emission sources such as lightning NO_x, biomass burning emissions of CO, VOC and NO_x, or anthropogenic versus biogenic activity.

The possibilities offered by comparisons between the different datasets and their combined use are manifold, e.g., combining different species from one sensor, the same species from different sensors, different species from different sensors, or trace gases with auxiliary data like lightning etc. The potential of synergistic use of the available information has not yet fully been exploited, but recent studies have begun to demonstrate the insights that can be gained from the combined use of various datasets. Here follows a short discussion on some aspects of different synergistic applications and their potential.

a Improving Retrievals

Additional measurements are required for the retrieval of tropospheric column densities. The sensitivity of satellite measurements for tropospheric trace gases is affected by aerosols and clouds as well as the spectral ground albedo. Information about these parameters can be gained from the spectral measurements themselves (Chapters 5 and 6), with the important advantage that, for example, cloud properties directly match the time and location of the column measurements. In addition, imaging spectrometers like MODIS or MERIS, and space-born LIDARs (CALIPSO), provide detailed information on clouds and aerosols with high spatial resolution.

Satellite observations designed for stratospheric trace gas observations can be used to extract tropospheric columns from total column measurements. This is of particular importance for the retrieval of tropospheric O_3, since the total O_3 column is dominated by the stratosphere. For instance, satellite SBUV measurements have been used to derive tropospheric O_3 from TOMS measurements (Fishman and Balok 1999). For a range of tropospheric gases, for example, NO_2 or BrO, the stratospheric column has to be known to retrieve the residual tropospheric column. SCIAMACHY operates in an alternating limb-nadir mode to allow the direct retrieval of stratospheric columns for the correction of nadir column measurements (Sierk et al. 2006; Beirle et al. 2009).

b Identifying Sources

Trace gas columns from satellite measurements can be compared to independent measurements of different parameters like fire or flash counts, which are proxies for different trace gas sources. For instance, fire counts from the satellite instruments ATSR or MODIS, are indicators of biomass burning and have been compared to CO (Edwards et al. 2006), HCHO and NO_2 (Spichtinger et al. 2004) and CHOCHO (Wittrock et al. 2006). Flash counts from the Lightning Imaging Sensor (LIS) have been compared to NO_2 columns over Australia to estimate NO_x production by lightning (Beirle et al. 2004c) (Fig. 8.23).

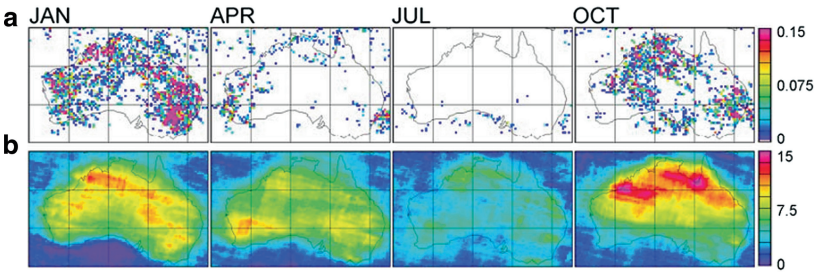

Fig. 8.23 Monthly mean LIS flash counts (upper row, flashes per day and km^2) and GOME tropospheric NO_2 column (lower row, 10^{14} molecules/cm^2) in Australia for several months in1999 (Beirle et al. 2004c).

Toenges-Schuller et al. (2006) used light pollution at night, as measured from DMSP, as proxy for anthropogenic emissions; it shows a good spatial correlation with NO_2 columns.

c Learning About Atmospheric Chemistry

From the UV/vis spectral measurements, several trace gases (*e.g.* NO_2, SO_2, HCHO, CHOCHO, CO, O_3) can be derived simultaneously, giving information on different sources, such as anthropogenic versus biomass burning, and allows insights into their chemistry and lifetimes. For instance, Martin et al. (2003) have differentiated NO_x saturated from NO_x sensitive regions from the observed ratio of HCHO to NO_2 columns (Fig. 8.24).

Results from different sensors for the same trace gas can be used synergistically. Currently, four nadir spectrometers in the UV/vis (GOME, SCIAMACHY, OMI and GOME-2) are in operation simultaneously. Comparisons between different instruments can provide consistency checks. In addition, the different overpass times provide information on the diurnal variations of trace gas columns. Boersma et al. (2008a) compared monthly mean NO_2 columns from SCIAMACHY (overpass time: 10:00) and OMI (13:30) (Fig. 8.25). OMI columns are lower than SCIAMACHY columns over fossil fuel combustion regions, mainly because of higher OH concentrations, and thus shorter lifetimes, in the early afternoon. Over biomass burning regions, OMI columns are higher owing to the diurnal cycle of fire activity, which is not considered in emission inventories.

d Learning About Profiles

Measuring one trace gas at different wavelengths provides additional profile information owing to different altitude sensitivities. For instance, MOPITT (4.7 μm) is more sensitive to CO in the free troposphere while CO columns in the NIR spectra

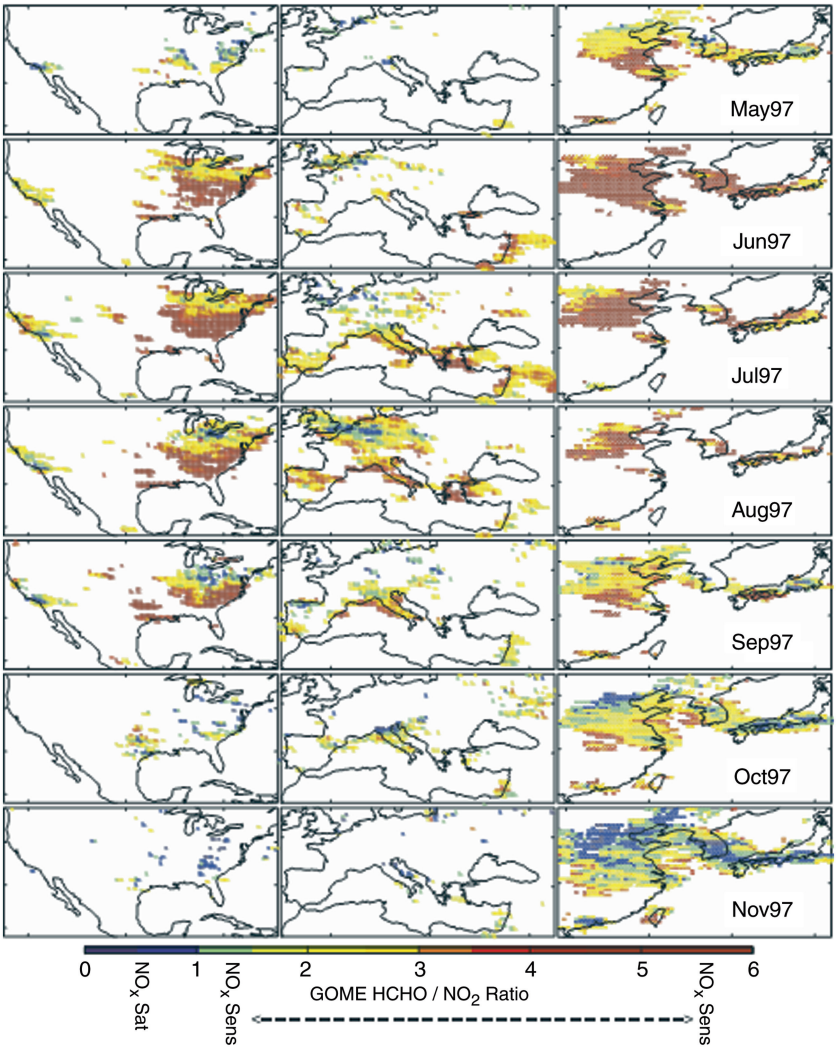

Fig. 8.24 Monthly mean tropospheric HCHO/NO_2 column ratio from GOME (Martin et al. 2004a).

from SCIAMACHY (2.3 μm) reach towards the ground; the difference allows the boundary layer concentration of CO to be estimated (Fig. 8.26) (Turquety et al. 2008).

e Multi-Platform Observations

Similar comparisons of trace gas columns from different sensors have been performed and are in progress for many trace gases. For CO, there are several new

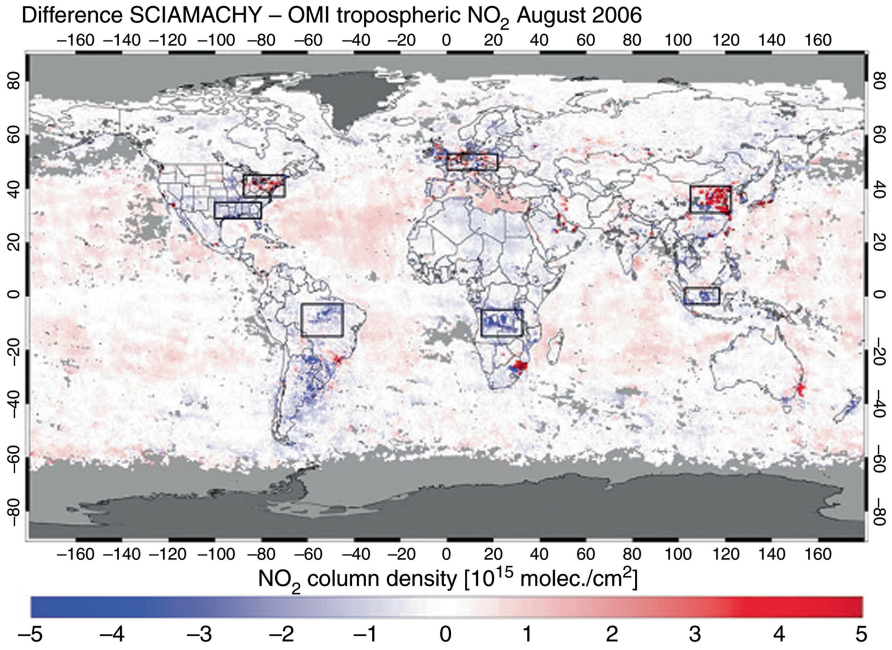

Fig. 8.25 Absolute difference between monthly mean SCIAMACHY and OMI tropospheric columns for August 2006 (Boersma et al. 2008a).

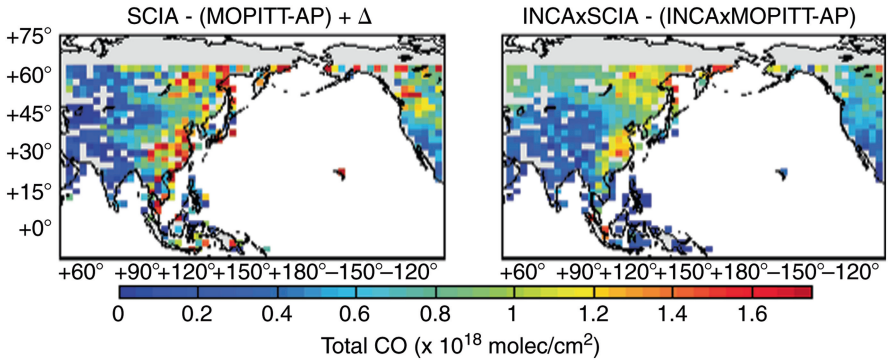

Fig. 8.26 Boundary layer residual of CO derived from the difference of SCIAMACHY and MOPITT columns of CO (Turquety et al. 2008).

instruments, so that now five (N)IR instruments (MOPITT, SCIAMACHY, AIRS, TES, IASI) are available for synergistic use. For instance, Prata et al. (2007) used measurements from AIRS (IR), SEVIRI (IR), MLS (MW, Limb), and OMI (UV/vis), to track the early evolution and long range transport of a volcanic cloud from Soufrière Hills volcano, Montserrat (Fig. 8.27) in order to estimate the total

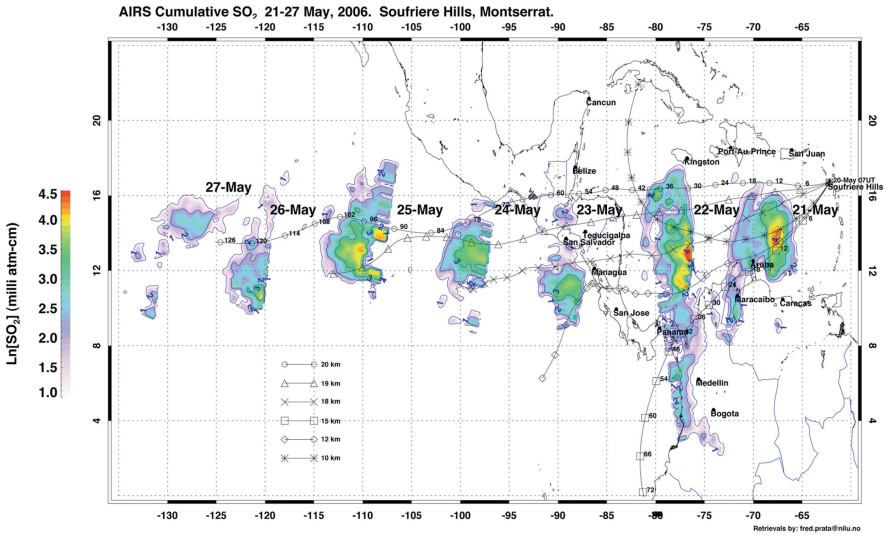

Fig. 8.27 AIRS SO_2 total column retrievals for 21st–26th May 2006 from the Soufriere Hills volcano, Montserrat (Prata et al. 2007).

SO_2 emissions. Such comparisons of the results from several satellite measurements, having different measurement times, wavelengths (and thus averaging kernels) etc. will further improve our understanding of tropospheric composition and chemistry.

8.3.6 Operational Use

Most of the tropospheric composition data collected from satellites so far has been from "research" instruments on individual science missions. However, both TOMS-OMI series and GOME-SCIMACHY missions have yielded long term measurements. As has been shown previously in Section 8.3.2 there is much to gain from long-time series of satellite data in mapping change in the earth-system.

However time series and monitoring, requiring consistent long-term measurements delivered in a timely fashion require operational measurements: measurements made with satellite instruments which are intended to deliver the data for the foreseeable future. An example is provided by the satellites used by the meteorological services.

IGACO is a strategy for bringing together ground-based, aircraft and satellite observations of 13 chemical species in the atmosphere. IGACO will be implemented as a strategic element of the GAW programme of the WMO.

Using the current generation of satellites, pilot operational services are being developed, *e.g.* PROMOTE/GMES (http://www.gse-promote.org/), or TEMIS (www.temis.nl) that provide information on air quality, UV and climate gases.

Long-term operational tropospheric chemical measurements from satellites have been initiated in concert with the meteorological community of MetOp (Klaes et al. 2007), providing measurements in the UV/vis (GOME-2) and the IR (IASI). In a parallel area, data fusion of various satellite sensors has been used to generate an operational aerosol prediction (Al-Saadi et al. 2005).

Despite the uncertainties of tropospheric trace gas columns derived from satellite measurements, particularly due to clouds and trace gas profiles, the improving spatial resolution and the growing number of different sensors available will allow us to plan different applications. For instance, future satellite instruments with footprints of some km^2 would allow the monitoring of air quality, in particular NO_x, on urban scale. A number of groups have indentified the utility of geostationary observations, not only for air quality applications where high spatial and temporal measurements are required, but also for operational assimilation into predictive models (Bovensmann et al. 2002, 2004; Burrows et al. 2004; Fishman et al. 2008; Munzenmayer et al. 2008). In particular, the existing satellite instruments on sun-synchronous orbits are not capable of resolving the diurnal chemistry cycle.

There has been some discussion on the application and use of so-called chemical weather (Lawrence et al. 2005), a direct analogy to meteorological weather, and the role satellites would play in delivery of operational data.

In future, satellite measurements could also play an important role in monitoring emission policies by measuring levels and trends of pollutants (NO_x, CO, SO_2), O_3, and greenhouse gases (CO_2 and CH_4).

8.4 Summary and Outlook

Satellite observations of tropospheric trace gases are a new and powerful tool to study tropospheric composition. The spatial and temporal information obtained globally provides unique information on sources, transport, and sinks for a range of gas-phase and particulate species. Tropospheric composition satellite data is beginning to make the transition from observation to quantification.

Looking to the future, a new view will be afforded with the long term continuous measurements from new satellites. There are now a number of programs to make the supply of high quality tropospheric composition data operational. In many senses, the satellites that have given so much insight into the troposphere were designed for research purposes and we await data from a generation of satellites designed for that purpose. A significant challenge still exists to merge the satellite data to the *in situ* observing systems in a meaningful and consistent way.

There is a clear need for improved spatial resolution, spatial cover and increased temporal coverage. Geostationary observations have a lot to offer (Fishman et al. 2008; Munzenmayer et al. 2008) for greater regional insights into tropospheric chemistry from space. There are scientific challenges across the globe, such as the continuing challenge of urbanisation and Megacities (Molina and Molina 2004), and a trans-national observing system for the tropics, that will require a new generation

of satellite measurements. Synergistic use of multiple instruments has just started and there is much data still to be analysed; there are many further applications ahead. Tropospheric satellite observations have to be ready to meet the challenges of climate change as we look to the future and this will put great demands on facilities, data, analysis and understanding.

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