Satellite Remote Sensing of Tropospheric Chemistry

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SCIENTIFIC RATIONALE FOR SPACE-BASED OBSERVATIONS OF THE TROPOSPHERE

For more than a decade, it has become evident that the climate and the composition of the Earth's atmosphere are changing at the global scale. Those global changes are driven by complex interactions of chemically and radiatively active species from both natural sources, such as biological activity and volcanoes, and man-made sources, such as industrial emissions and biomass burning. A better knowledge of the global composition and dynamics of the atmosphere and of their long-term evolution, are urgently needed to improve our understanding of the mechanisms that control tropospheric chemistry, and to assess current and future changes. Among others, it is critical to determine the spatial distributions of tropospheric ozone and its precursors (NO_x, CO, hydrocarbons, water vapour), as well as the magnitudes and spatial variations of global changes. Information on temperature, geopotential heights, clouds, and radiation fields is also needed, as well as the detection of events such as fires and lightnings. Tropospheric ozone chemistry largely influences not only air quality and ecosystems, but also climate change and stratospheric ozone depletion. At the surface level, ozone is toxic to the biosphere. In the upper troposphere, it is a significant greenhouse gas. Tropospheric ozone is also the primary precursor of tropospheric OH and therefore largely controls the oxidising capacity of the atmosphere. Other constituents of considerable environmental importance can be classified into: anthropogenic sources (CFC, HCFC), radicals (e.g., OH, NO, ClO), reservoirs (e.g., HNO_3, HCl), and tracers (e.g., CO for biomass burning). Tropospheric aerosols modify the radiative budget of the Earth and provide surfaces for heterogeneous reactions affecting tropospheric chemistry. At high concentrations, they are also harmful to public health.

Tropospheric investigations require measurements over a variety of scales ranging from localised source regions (e.g., urban emissions, volcanic plumes) to the global atmosphere, over a long enough period to separate long-term trends from natural variability associated with seasons or other cyclic or periodic events (e.g., El Niño/La Niña oscillation, sunspot cycle). Neither in situ, airborne, nor spaceborne experiments provide complete coverage in terms of time and space scales, resolutions, or species type, but synergistic use of all types of measurements is capable of addressing the key issues of tropospheric chemistry. By design, field experiments and airborne remote-sensing campaigns observe with good accuracy the smaller spatial and temporal scales, and consequently remain the preferred approach for many studies that focus on detailed processes, especially in the troposphere. For long-lived gases with comparatively uniform concentrations in the atmosphere, surface sampling at a network of stations provides a cost-effective alternative. Results of process studies relying primarily on aircraft and ground-based measurements require satellite measurements in order to extend them to the global domain. Space-based measurements are expensive, achieve lower spatial resolution, and are limited in sensitivity. In addition, clouds and aerosols alter significantly the radiation field and may perturb the measurement around the tropopause and below. But spaceborne sensors provide unique access to the needed, continuous large-scale mapping of the abundances of trace species. This mapping is critical for understanding sources, sinks, and chemical and dynamical processes controlling species with short atmospheric lifetimes (a few weeks or less) and, hence, large spatial and temporal variability. Space-based remote-sensing measures atmospheric composition at higher altitudes than can be conveniently attained by aircraft experiments. Observations from space can also play an invaluable role in the design of process studies by identifying a specific problem. A good example is the tropospheric ozone maximum over the south Atlantic in spring, which was first identified by analysis of TOMS and SAGE-II satellite measurements (see below) and was later confirmed and interpreted with aircraft observations.

TECHNIQUES FOR TROPOSPHERIC REMOTE-SENSING FROM SPACE

A bunch of atmospheric species can be detected from space by observing their absorption or emission spectral features in the ultraviolet and the visible (e.g., ozone, SO_2, NO_2 , NO_3, HCHO, BrO), the infrared (e.g., ozone, CFCs, CO, N_2O, NO, NO_2, HNO_3, HCl, H_2O, CH_4), and/or the millimetre wave (e.g., ozone, OH, HCl, ClO, H_2O) spectral regions. The variety of spectral regions offers different detection capabilities, not only as to atmospheric species but also as to penetration depth in the atmosphere and as to sensitivity to weather conditions (clouds, humidity,..). While absorption measurements can be relatively limited in space and time due to the need of a light source (Sun, Moon, or stars), measurements of the natural thermal emission of the atmosphere may provide diurnal coverage.

Space-based remote-sensing techniques rely on two main viewing geometries: the limb- and the nadir-viewing. Both geometries provide complementary information. When sounding the limb of the atmosphere, a sensor observes the radiation leaving the atmosphere tangentially. The essential advantages are the enhanced amount of absorber, emitter, or scatterer along the line of sight, and the vertical discrimination capability. The limb mode may provide a high vertical resolution, but a relatively poor horizontal resolution. For measurements in solar occultation mode (that is, in absorption when Sun rises or sets), the enhanced light path of the limb geometry combines with large signals to allow the detection of weak absorbers, but observation constraints yield poor spatial and temporal coverage. In addition, the concentration of many relevant atmospheric constituents changes rapidly during twilight, making their retrieval more difficult and less accurate. Stellar occultation can lead to much better spatial and temporal coverage, but other problems arise, such as scintillation because of variations of atmospheric refraction. The limb-viewing geometry has proven to be of incontestable interest for measurements from the upper troposphere through the stratosphere and mesosphere, but unsuitable at lower altitudes due to a too large optical thickness along the line of sight. When pointing in the nadir direction, a sensor observes radiation emitted, reflected or scattered by the atmosphere and the Earth's surface. This is the most powerful viewing geometry to get a real, direct insight down into the troposphere, and consequently a good measurement of the total column of an atmospheric species. Compared with limb sounding, the method allows higher horizontal resolution, and is less sensitive to inaccuracies in the direction of observation. But the height-resolved information is of limited vertical resolution, and the retrieval of tropospheric abundances may be perturbed by the stratospheric contribution to the measured column amount. At high sun elevation, spectral signatures from weak absorbers or emitters may fall below the detection threshold. A variety of spatial and temporal coverages can be achieved by the adequate combination of the observation strategy and the orbiting geometry: e.g., daily global coverage at constant local time with a sun-synchronous polar-orbiting nadir-viewing sensor, and day-night coverage in one month with a limb sounder in low inclination orbit.

SPACE-BASED OBSERVATION STRATEGY: PAST, PRESENT AND FUTURE

Until recently, space-based measurements of atmospheric chemistry focused on the stratosphere and the mesosphere. Tropospheric chemistry observations were almost limited to qualitative information inferred from nadir measurements, such as the detection of fire smokes and of strong tropospheric emissions of ozone by the NASA Total Ozone Mapping Spectrometer (TOMS). The first quantitative estimations of the tropospheric ozone column were obtained from the differentiation between co-located nadir measurements of the total column from TOMS and limb measurements of the integrated stratospheric column from the Stratospheric Aerosol and Gas Experiment II (SAGE-II) [Fishman *et al.*, 1990].

The recent growing interest of the atmospheric science community in spaceborne instruments partly or fully dedicated to tropospheric remote-sensing, is reflected in the current international Earth Observation programmes. Three new instruments have demonstrated the suitability of space-based remote-sensing for quantitative measurements of tropospheric chemistry: the ESA Global Ozone Monitoring Experiment (GOME), the Japanese Interferometric Monitor for Greenhouse Gases (IMG), and the NASA MOnitoring of Pollution In The Troposphere (MOPITT). GOME has been designed to extend the continuous, global mapping of total ozone started with the TOMS series in 1978 to other relevant trace species, some of them of tropospheric interest. A first GOME-1 was launched by ESA on 21 April 1995, onboard its ERS-2 environmental satellite. It since provides routinely the global picture of ozone and NO_2 and detects other relevant trace species, such as BrO, CH_2O, SO_2, and OClO. The GOME series will be continued with the Scanning Imaging Absorption Mapping spectrometer for Atmospheric CHartographY (SCIAMACHY), to be launched in March 2002 onboard the ESA ENVISAT-1 platform. Improved versions of GOME are programmed after 2003: Ozone Monitoring Instrument (OMI) onboard EOS-Aura at the end of 2003, and three GOME-2 instruments as part of the Eumetsat METOP (Meteorological Operational Programmes) missions planned from 2005 onward. IMG is an infrared Fourier transform spectrometer designed to monitor the earth's radiation balance, the temperature profile of the atmosphere, the temperature of the earth's surface, and physical properties of clouds. Atmospheric concentrations of water vapor, ozone, and other greenhouse gases (CO_2, CH_4, N_2O) are derived from IMG spectra. Launched in August 1996 onboard the Japanese Advanced Earth Observing Satellite (ADEOS), a first IMG operated successfully until the spacecraft failure on June 30, 1997. The MOPITT instrument, performing on the NASA EOS Terra satellite since March 2001, measures the upwelling infrared radiance. Using the retrieval technique of gas filter correlation radiometry, MOPITT provides information on the distribution, transport, sources and sinks of CO and CH_4 in the troposphere. The correlation spectroscopy technique was successfully used for tropospheric CO measurements during the two space shuttle flights of the MAPS experiment (Measurement of Air Pollution from Satellites) in 1994.

A number of new-generation instruments are now scheduled for launch over the next decade, within the framework of the International Earth Observing System (IEOS). This interagency programme combines a space-, air- and surface-based measurement strategy to provide the scientific basis to address global change, including tropospheric measurements. The space-based segment of IEOS consists of the EOS/MTPE satellite missions co-ordinated by NASA (Meteor-3M, Aura, Aqua, and Terra), and the polar-orbiting and mid-inclination platforms from Europe (ERS, ENVISAT, METOP series), Japan (ADEOS I and II, TRMM, HIROS), and the U.S. NOAA (POES series). Additional instruments drawing from emerging technology are presently in the planning stage. One may cite the Geostationary Tropospheric Pollution Explorer (GeoTROPE), designed to investigate the changing tropospheric composition. GeoTROPE will combine a ultraviolet/visible/near-infrared spectrometer built up on the GOME heritage, and a Fourier Transform infrared spectrometer which capabilities may be compared to those of IMG. A major advantage of the geostationary operation is that the quasi-permanent viewing of the same scene link diurnal with seasonal and annual time scales and regional with continental space scales.

THE GLOBAL OZONE MONITORING EXPERIMENT

The GOME instrument consists of a UV-visible grating spectrometer observing the solar irradiance and the solar radiation backscattered from both the atmosphere and the Earth’s surface, between 240 and 790 nm [Burrows *et al.*, 1998]. Operated in nadir-viewing geometry, a mirror scans 3 ground pixels of 320 x 40 km, yielding global coverage after three days at the equator and less at higher latitudes. The so-called Differential Optical Absorption Spectroscopy retrieval technique is used to derive column abundances of ozone, NO_2, BrO, CH_2O, SO_2, and OClO from GOME spectra. Height-resolved ozone is obtained by inversion of the radiometric measurements between 240 and 400 nm, using advanced methods inspired from those developed for the NASA BUV stratospheric instruments [Barthia *et al.*, 1996]. The better spectral resolution and the wider, continuous spectral range of GOME allow to extend this technique down through the troposphere. Information on cloud cover and cloud-top height is derived from GOME measurements in the O_2 A-band at 760 nm, while radiometric measurements at several wavelengths from the ultraviolet to the infrared are used to infer information on aerosols.

After three years of successful operation onboard ERS-2, GOME-1 has already demonstrated its ability to provide valuable experimental support to tropospheric chemistry studies [Hahne, 1997]. Enhanced tropospheric emissions of NO_2, CH_2O and ozone are observed over major industrial areas, and in the Tropics during the season of biomass burning. Large-scale forest fires in Indonesia (1996) and Mexico (1998) were monitored by GOME-1 through the detection of abundant emissions of NO_2, CH_2O, smokes, and absorbing aerosols. Measured SO_2 amounts were useful to follow the time evolution of SO_2 plumes released by volcanic eruptions, such as those of the Nyamuragira (Zaire) in December 1996 and the Popocatepetl (Mexico) in July 1997. SO_2 associated with intense anthropogenic pollution events might also be detectable. Enhanced tropospheric BrO abundances were observed by GOME-1 at many occasions in polar regions, giving new insight into the problem of tropospheric ozone depletion. Using GOME-1 measurements of the Earth radiance, solar irradiance, and total ozone, the ultraviolet radiation field at various stratospheric and tropospheric altitude levels and the UV index at the surface are calculated. Thereby GOME provides valuable support to studies of the tropospheric oxidation capacity and related photolysis rates.

Relying on the same concept, the German/Dutch/Belgian SCIAMACHY instrument will observe the atmosphere under different, complementary viewing geometries. It combines the nadir-viewing strategy of GOME with limb sounding of the same air mass. This interleaved strategy will provide an accurate measurement of the vertical distribution and of the stratospheric, tropospheric and total columns of several trace constituents. The extension of the SCIAMACHY spectral range through the mid-infrared (2400 nm) will allow the detection of additional molecules of both tropospheric and stratospheric interests (N_2O, CO, CH_4), and a better retrieval of cloud and aerosol information. Five years of SCIAMACHY operation are planned onboard the ESA ENVISAT-1 polar platform.

FUTURE TROPOSPHERIC CHEMISTRY SENSORS

In the framework of IEOS, a variety of atmospheric chemistry experiments are planned from 2002 onward. The GOME series (GOME-1, SCIAMACHY, OMI, GOME-2) has already been mentioned. Hereinafter, we present a non exhaustive list of other instruments which are expected to provide valuable information for addressing tropospheric chemistry issues.

In addition to OMI, several atmospheric chemistry instruments are planned to fly on the EOS Aura platform. The Tropospheric Emission Spectrometer (TES) is a high-resolution infrared Fourier transform spectrometer, equipped with linear array detectors. TES has the capability to make both limb and nadir observations, covering the lower troposphere up to middle stratosphere (33 km) with a 3-4 km vertical resolution. Both the natural thermal emission of the surface and atmosphere and reflected sunlight are measured, providing day-night coverage. TES is a pointable instrument and can access any target within 45 degrees of the local vertical, or produce regional transects up to 1700 km length without any gaps in coverage. Observations by TES will improve our understanding of long-term variations in the quantity, distribution, and mixing of trace gases in the troposphere, including ozone, NO, NO_2, HNO_3, CO, CH_4, and H_2O. The High Resolution Dynamics Limb Sounder (HIRDLS) is an infrared limb-scanning radiometer designed to sound the upper troposphere, stratosphere, and mesosphere. It will measure temperature, the concentrations of O_3, H_2O, CH_4, N_2O, NO_x, HNO_3, N_2O_5, CFC11, CFC12, $ClONO_2$, and aerosols, and the locations of polar stratospheric clouds and cloud tops. The Microwave Limb Sounder (MLS) will measure lower stratospheric temperature and concentrations of H_2O, O_3, ClO, HCl, OH, HNO_3, and N_2O, for their effects on transformations of greenhouse gases, radiative forcing of climate change, and ozone depletion. MLS will measure upper tropospheric H_2O and O_3 for their effects on radiative forcing of climate change and diagnoses of exchange between the troposphere and stratosphere. MLS will also measure SO_2 and other gases mentioned above in volcanic plumes, to investigate the effects of volcanic injections into the atmosphere.

The Infrared Atmospheric Sounding Interferometer (IASI) is scheduled for launch on the first mission of the EUMETSAT METOP (METOP-1, early 2002). It is a Michelson interferometer in the thermal infrared designed for atmospheric emission measurements at nadir. Its objectives comply with the METOP global objectives as it focuses on operational meteorology and climate aspects. It however has an additional scientific research objective with a focus on climate and tropospheric chemistry research. In the latter field, its particular objectives are the sounding of temperature through the measurement of CO_2 lines, the retrieval of total column amounts of H_2O, CO, O_3, CH_4, N_2O, and possibly of CFCs, and the measurement of atmospheric radiation. The retrieval of height-resolved ozone looks feasible.

INVOLVEMENT OF THE BELGIAN INSTITUTE FOR SPACE AERONOMY AND COLLABORATING INSTITUTES

For several decades, the Belgian Institute for Space Aeronomy (IASB-BIRA) and collaborating institutes have been involved in space-based atmospheric remote-sensing, through: the design of instruments; the development of retrieval algorithms; their geophysical validation; the generation of value-added data products; and the scientific interpretation of satellite data. In collaboration with the universities of Brussels (ULB/LCPM), Liège (ULG) and Reims (GSMA, France), IASB-BIRA performs laboratory measurements and spectroscopic studies of relevant atmospheric species (e.g., CFCs, NO_2, SO_2, H_2O, O_2/O_4). IASB-BIRA contributes to the development, maturation and/or interpretation of geophysical products from the GOME and TOMS series, the ENVISAT instruments, IASI, MOPITT, and SAGE. In particular, prototype algorithms are being designed to derive UV fields and UV index at the surface, solar Mg-II index, and BrO column abundances from GOME and SCIAMACHY measurements. Advanced tools for value-added data processing have been developed to convert satellite observations acquired asynoptically along sparse orbits, into a more suitable format for scientific end-users: e.g., maps of measured atmospheric species, aerosols, and UV radiation field. In collaboration with the Royal Meteorological Institute (IRM-KMI), ULG, the Service d'Aéronomie du CNRS (France), and two dozen of research centres world-wide, IASB-BIRA participates to the geophysical validation of a number of instruments through international research projects involving ground-based monitoring networks, balloons and satellites. A number of ongoing projects aim at investigating the distribution, transport, sources and sinks of tropospheric species through the use of satellite-derived data records.

FOR MORE INFORMATION

Part of the reported work is funded by OSTC Grant MO/35/006. Further information on space-based tropospheric data products and their usability can be found on the web site of the Eurotrac-2 TROPOSAT project at http://crusoe.iup.uni-heidelberg.de/luftchem/troposat/ . A comprehensive list of links to satellite atmospheric chemistry missions and related Earth Observation Programmes is maintained at http://www.ndsc.ws .

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