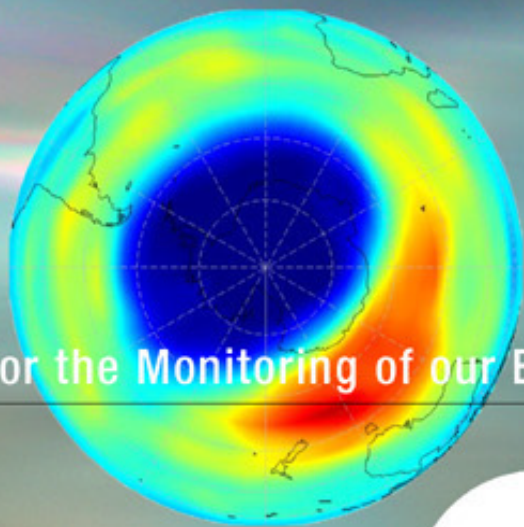


IGACO

THE INTEGRATED GLOBAL
ATMOSPHERIC CHEMISTRY
OBSERVATIONS THEME



For the Monitoring of our Environment from Space and from Earth

IGOS
Integrated Global Observing Strategy



September 2004

An international partnership for
cooperation in Earth observations

ESA SP-1282, September 2004
Report GAW No. 159 (WMO TD No. 1235)

THE CHANGING ATMOSPHERE

**AN INTEGRATED GLOBAL
ATMOSPHERIC CHEMISTRY OBSERVATION
THEME FOR THE IGOS PARTNERSHIP**

**Report of the Integrated Global Atmospheric Chemistry Observation
Theme Team**

September 2004

Figures on the cover courtesy of E. Hilsenrath (NASA), P.C. Simon (IASB), NILU.

The Integrated Global Observing Strategy (IGOS) is a partnership of international organisations that are concerned with global environmental-change issues. It links research, long-term monitoring and operational programmes, bringing together the producers of global observations and the users that require them, to identify products needed, gaps in observations and mechanisms and to respond to needs in the science and policy communities. Its principal objectives are to integrate satellite, airborne and *in situ* observation systems.

The IGOS partners comprise the Global Observing Systems (GOS), the international organisations that sponsor the Global Observing Systems, the Committee on Earth Observation Satellites (CEOS) and International Global Change Research programmes.

The IGOS Partners recognise that a comprehensive global Earth observing system is best achieved through a step-wise process focused on practical results. The IGOS themes allow for the definition and development of a global strategy for the observation of selected environmental issues that are of common interest to the IGOS partners and to user groups. Examples of such themes include the *Oceans, Coral Reefs, the Carbon and Water Cycles, Coastal Zone and Atmospheric Chemistry*.

The IGOS Atmospheric Chemistry theme was approved by IGOS in 2001. After six working group meetings, and several interim reviews by IGOS partners, the present theme report was approved for implementation by the IGOS partnership in May 2004.

In 2003, the participants at the Earth Observation Summit launched the intergovernmental *ad hoc* Group on Earth Observations (GEO), with the goal of furthering the creation of a comprehensive, coordinated, and sustained Earth observing system or systems. IGACO strives to serve the GEO aims for atmospheric composition.

Further information on IGOS can be obtained from: <http://www.igospartners.org>.

The IGACO theme report is available from: www.wmo.int/web/arep/gaw/gawreports.html

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Long-term measurements have clearly shown that human activities are changing the composition of the Earth's atmosphere. Research has demonstrated the important consequences of such changes for climate, human health, the balance of ecosystems and the ability of the atmosphere to cleanse itself of harmful pollutants and greenhouse gases. The awareness that chemical species in the atmosphere are key elements of the Earth system, and public concern about the impact of man, has led international organisations, such as WMO, UNEP and ICSU, to support national and international research programmes and assessments. These in turn triggered international conventions (e.g. Vienna Convention on Ozone Depletion, United Nations Framework Convention on Climate Change) and national directives.

In a declaration published on July 31, 2003, the International Earth Observation Summit emphasised the vital importance of Earth observation activities, encouraging the development of a comprehensive, coordinated, and sustained Earth observation system.

The 'Integrated Global Atmospheric Chemistry Observations' (IGACO) theme is a component of the Integrated Global Observing Strategy (IGOS) partnership; it was approved as the fourth theme of IGOS in June 2001, following those for the Ocean, the Global Water Cycle, and the Global Carbon Cycle. A panel of international scientists were convened by the WMO and ESA to produce this theme report; the panel is grateful for the able assistance of a number of distinguished reviewers, and for many constructive comments from the Secretariat and Executive Committee of the IGOS partnership, as well as the Strategic Implementation Team of CEOS.

The objective of this report is to initiate a process leading towards a globally coordinated development of future observation and integration programmes, whose components are either in place or, with careful planning, can be implemented within the next 10 years. The report

- identifies the current major societal and scientific issues associated with atmospheric composition change;
- establishes the requirements for observations of atmospheric composition and their analysis, integration and utilisation;
- reviews the existing observational systems, including data processing and distribution, and validation programmes *vis-à-vis* these requirements; and
- proposes an implementation plan to adapt the systems to meet the identified requirements.

The emphasis of the report is on the need for long-duration integrated observations and their societal and scientific applications. The focus will therefore be on operational systems providing continuity and reliability, and on setting priorities, in order to establish a technically and programmatically feasible long-term solution. It should be noted that the report addresses the needs for a *global* observation system and the value-added benefit that comes from integration. The schedule for implementation is divided into short- and long-term actions. One thing is clearly evident: with the lead times for deploying satellites and for developing ground stations and routine aircraft programmes, planning for funding and implementation of both stages has to begin immediately if the aims of the report are to be fulfilled.

This report draws upon the WMO/CEOS report on a strategy for integrating satellite and ground-based observations of ozone, which collected world-wide expertise and generated quantitative observation requirements and summaries of various observational systems.

The report starts with an Executive Summary containing the major recommendations. Chapter 1 deals with the environmental context of IGACO. The major challenges in atmospheric chemistry that underlie the proposal are discussed in Chapter 2 where the scientific requirements are specified. Chapter 3 outlines the current state of the three main measurement components of IGACO, ground-based, aircraft and satellite, as well as the associated models needed for integration. In Chapter 4 a target list of chemical species and atmospheric parameters is given together with the criteria used for the selection. The status of the observation system for each species is then discussed. Observational requirements for each of the chosen species are given for use in extending and improving the present observational systems. Finally, in Chapter 5, implementation is discussed. The proposed structure of IGACO is presented with a set of recommendations. An implementation process is suggested.

It is the panel's sincere wish that the strategy proposed in this report commends itself to organisations, funding agencies and research communities around the World which would be involved in implementing an Integrated Global Atmospheric Chemistry Observation system. It is our hope that it will bring us one step closer both to

observing the atmosphere and, with the knowledge gained, to alleviating the current chronic problems in the atmosphere caused by human activity.

Finally as editors, we would like to thank our colleagues, the co-authors and reviewers, whose names and affiliations are set out in Appendix C. It is thanks to their commitment and enthusiasm that this report has been produced.

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The aim of this report is to define a feasible strategy for deploying an Integrated Global Atmospheric Chemistry Observation System (IGACO), by combining ground-based, aircraft and satellite observations with suitable data archives and global models. The purpose of the system is to provide representative, reliable and accurate information about the changing atmosphere to those responsible for environmental policy development and also to prediction centres for weather and the environment. IGACO will also improve our scientific understanding of the changing atmosphere.

Motivation

The atmosphere, like the other components of the Earth system, is affected by the continuous increase in human population and activity, which have resulted in a variety of remarkable changes since the industrial revolution of the 19th century. Among these are:

- the global decrease in stratospheric ozone and the attendant increase in surface ultraviolet radiation, emphasised by the ozone hole appearing over the Antarctic;
- the occurrence of summer smog over most cities in the World, including the developing countries, and the increased ozone background in the northern troposphere;
- the increase in greenhouse gases and aerosols in the atmosphere and associated climate change;
- acid rain and the eutrophication of surface waters and other natural ecosystems by atmospheric deposition;
- enhanced aerosol and photo-oxidant levels due to biomass burning and other agricultural activity;
- the increase in fine particles in regions of industrial development and population growth with an attendant reduction in visibility and an increase in human health effects; and
- the long-range transport of air pollution to regions far from the industrial activity.

Many of these changes in atmospheric composition have socio-economic consequences through adverse effects on human and ecosystem health, on water supply and quality, and on crop growth. A variety of abatement measures have been introduced or considered to reduce the effects. However, continued growth in human activities to expand economies and to alleviate poverty, will ensure that these effects continue to be important for the foreseeable future.

The Grand Challenges in Atmospheric Chemistry

Four grand challenges in atmospheric chemistry underlie the environmental issues above:

- tropospheric air quality;
- the oxidation efficiency of the atmosphere;
- stratospheric chemistry and ozone depletion; and
- chemistry - climate interactions.

The scientific understanding of each challenge requires long-term observation of the atmosphere and points firmly to the need to establish an integrated global observation network.

The Need for IGACO

Over the last half century, a variety of national networks has been installed to monitor atmospheric trace gases of local concern and, as the trans-boundary nature of air pollution was recognised, there has been some limited coordination of network development in Europe, in North America and globally. Most of the activities concern ground-based stations, including balloon sondes and remote sensing. Regular aircraft measurements were started in the 1990s by deploying unattended instrument packages on commercial airliners.

Satellites have added a new dimension by providing a global picture of the total column ozone together with some profile information. Now, it has become possible to measure from space, total and tropospheric columns of a number of atmospheric constituents as well as concentration profiles, albeit limited in resolution, in nadir-viewing geometry, as well as high-resolution profiles in the stratosphere and upper troposphere. At the same time, global

weather and climate models that incorporate chemical cycles are emerging, which are capable of assimilating a variety of observations into a global picture.

Most of the current ground-based activities are primarily regional and are driven by specific scientific or legal objectives. Unfortunately, because of the lack of overall coordination and inherent shortcomings in the individual systems, none of the present monitoring activities achieves the completeness, representativeness and resolution required for obtaining a global view. There are thus large gaps in our picture of the atmosphere and in our understanding of the various influences that drive atmospheric variability and long-term changes.

To obtain a reliable global picture of the changing atmosphere, a coupled observational/modelling system is required, which is built partly on components that are already available, but is designed to obtain and integrate fully the best data that the diverse observation systems provide. The implementation of IGACO will require substantial investment to fill gaps in the existing satellite and other systems, whose current generations will cease operation just at the time when IGACO should go into operation. Clearly, if we want to avoid severe uncertainties in our diagnostic and prognostic capabilities, decisive steps towards the assembly of IGACO must be taken without delay.

The Chemical Species and Ancillary Parameters Required for IGACO

Taking account of several realistic financial and logistic constraints, the chemical species and parameters to be included in IGACO were selected using the following criteria:

- the chemical species or variable plays an important role in one or more of the four key atmospheric chemistry issues, identified in IGACO, and there is added value in its incorporation in an integrated global observation system;
- it is now possible, or likely to be possible, to measure the atmospheric constituent or parameter globally and on a long-term basis so as to achieve a synergism between satellite, ground-based and aircraft observations, and model assimilation systems.

The selected atmospheric constituents and parameters are:

ozone (O ₃)	water vapour (H ₂ O)	carbon dioxide (CO ₂)
carbon monoxide (CO)	nitrogen dioxide (NO ₂)	bromine oxide (BrO)
chlorine monoxide (ClO)	hydrogen chloride (HCl)	nitrous oxide (N ₂ O)
CFC-12 (CF ₂ Cl ₂)	HCFC-22 (CHClF ₃)	chlorine nitrate (ClONO ₂)
methane (CH ₄)	formaldehyde (HCHO)	volatile organic compounds (VOC)
sulfur dioxide (SO ₂)	nitric acid (HNO ₃)	chlorine dioxide (ClO ₂)
nitrogen monoxide (NO)	methyl bromide (CH ₃ Br)	halons (e.g. CF ₃ Br)
<i>J</i> (NO ₂) and <i>J</i> (O ¹ D) (UV radiation at specific wavelengths in the troposphere).		

and the following aerosol optical properties at multiple wavelengths:

optical depth (<i>V_s</i> + <i>R</i>)	extinction coefficient (<i>V_s</i>)	absorption optical depth (<i>V_s</i>)
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Ancillary Parameters

In addition, the following additional parameters are required for atmospheric chemical modelling and retrieval of remote sensing data:

temperature (T)	pressure (p)	fire frequency
cloud-top height	cloud coverage	albedo
solar radiation	lightning flash frequency	wind (u, v, w)

Chapter 4 of the report details the *targets and thresholds for precision, accuracy, temporal and spatial resolution* that must be met for each of the chemical species and parameters to be meaningful within IGACO.

The Essential Components of IGACO

For full functionality the system should include the following four components:

- **Networks of ground-based instrumentation**, including balloon sondes, millimetre wave radiometers, lidars, UV-Visible and FTIR spectrometers, to measure ground concentrations, column densities and vertical profiles of the principal chemical species, aerosols and ancillary parameters, together with UV radiation, on a regular basis; the implication is that the current network needs to be maintained and expanded to fill critical gaps in coverage.
- **Regular aircraft-based measurements** for chemical and aerosol measurements in the troposphere and particularly in the upper troposphere and lower stratosphere (UT/LS), which is sensitive to chemical and climate changes. The current fleet needs to be expanded with respect to instrumentation and global coverage and to include regional aircraft that monitor the lower to middle troposphere. Routine measurements in the lower to middle troposphere using non-commercial light aircraft could be a useful part of the system.
- **Satellite-based instrumentation**, preferably mounted on a combination of geostationary (GEO) and Low Earth Orbit (LEO) satellites to provide measurements with the temporal and spatial resolution needed for sufficient coverage. To obtain the time resolution necessary to encompass tropospheric variations, three to four geostationary satellites would be required to cover all longitudes. An alternative would be several LEO polar-orbiting satellites. Upper tropospheric/lower stratospheric observation requirements would be covered by two polar-orbiting limb-sounding missions.
- **A comprehensive data modelling system** capable of combining the data for the chemical species, aerosols and ancillary parameters, obtained from the three measurement components, into a comprehensive global picture; assimilation techniques for chemical species other than ozone are still in the demonstration phase and need to be developed into operational procedures, which will require the development of physical and chemical parameterisations.

Further essential parts of IGACO are end-to-end quality assurance and quality control, as well as data collection, integration and analysis.

Summary of IGACO Recommendations

The principal recommendations of IGACO, as detailed and discussed in Chapter 5 are:

- | | |
|-----|--|
| GR1 | Establishment: an Integrated Global Atmospheric Chemistry Observation System (IGACO) should be established for a target list of atmospheric chemistry variables and ancillary meteorological data. |
| GR2 | Continuity: the data products from satellite and non-satellite instruments, which are to be integrated into a global picture by IGACO, must have assured long-term continuity. |
| GR3 | Management of IGACO: the responsibility for the co-ordination and implementation of the IGACO should rest with a single international body. International and national agencies responsible for aspects of IGACO should be committed partners and agree on their appropriate responsibilities. |
| GR4 | Gaps in observational coverage: for each target species and variable, the present gaps in the current spatial and temporal coverage should be filled by extending the existing measurement systems. |
| GR5 | Long-term validation of satellite observations: in order to ensure the accuracy and consistency of satellite measurements, sustained quality-assurance measures, over the entire lifetime of satellite sensors, are essential. |
| GR6 | Validation of vertical profile data from satellite observations: a set of high-performance scientific instruments using ground, aircraft and balloon platforms, possibly operated on campaign basis, must be maintained to provide the crucial validation data. |
| GR7 | Comparability: the ability to merge observations of different types must be ensured by insisting that appropriate routine calibration and comparison activities linking diverse measurements together are part of an individual measurement programme. |

- GR8 Distribution of data: universally recognised distribution protocols for exchange of data on atmospheric chemical constituents should be established.
- GR9 Multi-stake holder World Integrated Data Archive Centres (WIDAC) should be established for the targeted chemical variables.
- GR10 Storage for raw data should be established so that they can be re-interpreted as models and understanding improve.
- GR11 The development of comprehensive chemical modules in weather and climate models with appropriate data assimilation should be an integral part of IGACO.
- GR12 Strong coordination with the meteorological services is essential so that the ancillary meteorological data, required by IGACO, is accessible.

Specific recommendations (SR1 to SR7) for the implementation of IGACO are given in Section 5.3 of the report, together with suggestions for the commitment process.

The Implementation of IGACO

Two steps are required to implement IGACO in good time and avoid gaps in data coverage.

IGACO Phase-1: short term (0-10 year, i.e. before 2014)

Specific actions should include:

- Establishment of an IGACO system for the atmospheric constituents, O₃, H₂O, CO₂, CO, NO₂, BrO, ClO, HCl, N₂O, the CFCs, ClONO₂ and aerosol optical properties.
- Development of data harmonisation and quality assurance/quality control for all the aspects of the initial system.
- Upgrading of selected ground-based stations and routine aircraft platforms.
- Development of operational algorithms necessary for satellite measurements in the troposphere for as many parameters as possible.
- Development of modelling and assimilation tools, parameterisations and strategies to produce a reliable overall picture from the various measurement components.
- Planning and initiation of a sustainable *long-term* network for IGACO Phase-2, with emphasis on a coordinated satellite system comprising both LEO and GEO instruments, complemented by non-satellite observational networks.

IGACO Phase-2: long-term (>10 year, i.e. beyond 2014)

As shown by the timelines in Chapter 4, there is a looming gap in the provision of satellite-based instruments for atmospheric chemistry observations since, by the beginning of the second phase, the present "demonstration mode" satellites will have finished their missions. For IGACO, new operational satellites as well as an enhanced non-satellite measurement network will be required.

Specific long-term requirements are:

- to provide an operational network consisting of the appropriate combination of LEO and GEO satellites to give the required coverage and time resolution for the tasks envisaged; *immediate action* is required from the agencies involved to avoid a time gap in global surveillance;
- to provide satellite instruments for measurements of the atmospheric constituents in Phase 1 together with CH₄, HCHO, volatile organic compounds (VOC), SO₂, HNO₃, OCIO, NO, CH₃Br, and the halons, together with *j*(NO₂) and *j*(O¹D);
- to upgrade ground-based stations to measure all the constituents, including aerosol chemistry, and to install new stations in locations to which the assimilation models are particularly sensitive;

- d. to secure routine aircraft measurements, to implement instrumentation for all the constituents, and to add routes to improve global coverage; and
- e. to develop further models and assimilation techniques to provide a reliable picture of the atmospheric variability and long-term changes.

Commitment

IGOS requires that a theme such as IGACO be led by one or more of its partners, and that "unless exceptionally approved, it is expected that one of the four observing programmes will lead future implementation." Considering that the focus of IGACO is on atmospheric composition, the natural lead is the WMO/GAW programme. Other essential players in the implementation are the satellite agencies represented by CEOS, the other components of WMO, and the research community in universities and organisations.

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Chapter 1 outlines the environmental concerns associated with anthropogenic changes to atmospheric composition in the context of the Earth system. It lists the related international conventions and the societal benefits of an IGACO system.

Chapter 2 explains the scientific background of the four major issues: air-quality, oxidising efficiency, stratospheric ozone and climate-chemistry interaction; and highlights the role of the various chemical species.

Chapter 3 provides an overview of the four components of the existing global observation system for atmospheric composition: ground-based, airborne, and satellite data, as well as data assimilation and modelling.

Chapter 4 analyses the current observation system species by species, and establishes detailed observational requirements for a future IGACO system.

Chapter 5 gives the IGACO recommendations for observations and modelling, validation, data distribution and archiving; and makes suggestions for the organisation, management and phasing of the implementation phase.

1.1 Man's Influence on the Atmosphere

The Earth is now firmly entrenched in an 'anthropocene' epoch where the activities of humans on a planetary scale are appreciably altering the processes that control the Earth's system. The system comprises a complex set of biological, physical and chemical processes taking place in and between the oceans, continents and the atmosphere. Our atmosphere has sufficient oxygen to maintain life, and quantities of trace gases, which serve to maintain the UV-protecting stratospheric ozone layer, and a greenhouse effect which maintains habitable temperature conditions at the surface. The system of trace gases is maintained by a complex web of chemical processes that also serve to maintain the atmosphere in a sufficiently 'clean' condition for life to continue.

Historically, human activities have always interacted with the atmosphere, but the growth of population and industrialisation in the 19th and 20th century has led to dramatic changes in the Earth system. The development of large mega cities and the changes in land usage, occurring in the last 50 years, are two outstanding examples. Air pollution, the over-burdening of the atmosphere with emitted substances, has been known from the earliest times, but it was always a local nuisance. In the 20th Century pollution spread to regional and global scales and became a subject of concern both nationally and internationally. Table 1.1 lists some of the noticeable environmental effects attributable to the atmosphere. Although they appear in a roughly chronological order, all are still of current public concern.

Many aspects of air pollution adversely affect human, animal and ecosystem health and have considerable economic consequences. Such effects have been recognised for some time and the public concern about the atmosphere is reflected in a number of international agreements.

- The Convention on Long-Range Transboundary Air Pollution (LRTAP) (1979) with protocols on Sulphur (1985 & 1994), Nitrogen Oxides (1988), Volatile Organic Compounds (VOC) (1991), Heavy Metals (1998), Persistent Organic Compounds (POP) (1998), and Acidification, Nutrification and Ground Level Ozone (1999).
- The Vienna Convention on the Protection of the Ozone Layer (1985) and the Montreal Protocol on Substances that Deplete the Ozone Layer (1987), and the subsequent adjustments and amendments.
- The Framework Convention on Climate Change (1992) and the Kyoto Protocol (1997).

- The Convention to Combat Desertification in Countries Experiencing Serious Drought and/or Desertification, particularly in Africa (1994).

Most agreements call for the monitoring, reporting or assessment of data on environmental parameters. In addition, conventions for limitations of the pollution of the sea require continuing estimates of pollution inputs from the atmosphere since, in some cases, these are as great as from the river inflows. It is to support these various agreements that IGACO, the project described in this report, is principally conceived.

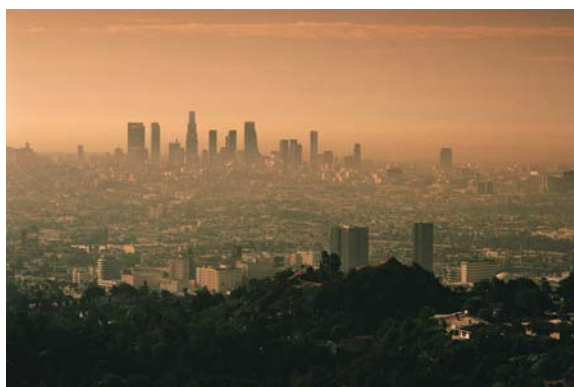


Figure 1.1. Smog over Los Angeles. [Courtesy of E. Hilsenrath, NASA]

1.2 IGACO in the Context of the Earth System

The proposal is to create an observation system for the Earth's atmosphere by extending and improving the current measurement capabilities and integrating them together using modelling techniques to provide pictures of the atmospheric composition on both short and long time scales. The system is referred to in this report as IGACO (Integrated Global Atmospheric Chemistry Observations).

The atmosphere interacts with the land and the ocean, the other components of the Earth system, and is essential to sustaining life on Earth (Fig. 1.2). Its importance is recognised within the International Geosphere Biosphere Programme (IGBP) and, in all three of its programmes concerned with the atmosphere – IGAC (International Global Atmospheric Chemistry), SOLAS (Surface Ocean and Lower Atmosphere Study) and ILEAPS (Integrated Land Ecosystem – Atmosphere Processes Study) atmospheric chemistry is regarded as a primary engine for global climate and environmental change. IGACO will support and interact with the other themes of IGOS (International Global Observation Strategy), providing information on atmospheric composition and deriving information on emissions. It will thus play a major role

Climate change and the greenhouse effect	The likely change in climate due to a continuing increase in the average global temperature. Resulting from the increase in carbon dioxide (e.g. from industrial and vehicular combustion) and other trace gases such as methane, ozone and the HCFCs. Aerosols also play a part, by perturbing the Earth's radiative balance, both directly through the scattering and absorption of sunlight, and indirectly through the influence on clouds and the hydrological cycle. Changes in both tropospheric and stratospheric temperatures are likely to have negative (or amplifying) effects on many issues.
London smog	Visible soot particles and sulphur dioxide, trapped in a static high-pressure weather system. Resulting from domestic heating and industrial activities. Now largely eliminated in the developed countries, but still a problem in the developing World.
Los Angeles smog or 'summer' smog	Products of photo-oxidation, notably ozone and PAN, and other lachrymatory compounds, and particles. Resulting from the photo-oxidation of industrial and vehicular emissions in the presence of sunlight. Endemic under static weather conditions throughout the World, despite countermeasures taken in the developed and developing World.
Acid rain	Acid precipitation, which threatened natural ecosystems and forests over Europe and North America; resulting from industrial emissions of sulphur dioxide, which is photo-oxidised to form sulphate. Although sulphur emissions have been reduced, acidity is still a problem with the growing contribution of nitrogen compounds from industry and vehicles to acidity, and the SO ₂ and NO _x from ships.
Ozone depletion in the stratosphere	Loss of ozone from the stratosphere, with a consequent increase in UV radiation reaching the surface. Resulting from the release of CFCs from industrial processes, which are inert in the troposphere but photolyse in the stratosphere. The ozone hole is a dramatic seasonal demonstration. Although the production of CFCs has been curbed, ozone depletion will remain for a long time because of the slow rate at which the compounds, still mostly present in the troposphere, diffuse to the stratosphere.
Nutrication of coastal waters and freshwater lakes; eutrophication	Change in coastal and freshwater ecosystems with the increased deposition of nitrogen compounds from the air. Resulting from industrial and vehicular emissions. Eutrophication and "tides" of poisonous algae are more obvious manifestations.
The increase in background tropospheric ozone	Observed in the northern hemisphere, the increase is attributed to the intercontinental transport of photo-oxidants. Current background levels are comparable to those known to affect plant growth.
Enhanced aerosol and photo-oxidant levels due to biomass burning	Observed annually over the Atlantic and Indian Oceans, as well as South-East Asia as a result of biomass burning and agricultural activities in Africa, South America and Asia.
Aerosols in and downwind of regions of high population	The recently observed "brown cloud" over the western Indian Ocean, far from land, originating from industrial, agricultural and vehicular activities in the Indian subcontinent and South-East Asia and possibly China.
The intercontinental transport of pollutants and aerosols	Recently observed across the North Atlantic, Pacific and southern Indian Ocean.
Stability of the atmospheric oxidation efficiency	The efficiency of the atmosphere for removing greenhouse gases and pollutants has been called into question.

Table 1.1: Some atmospheric problems attributed to human activities.

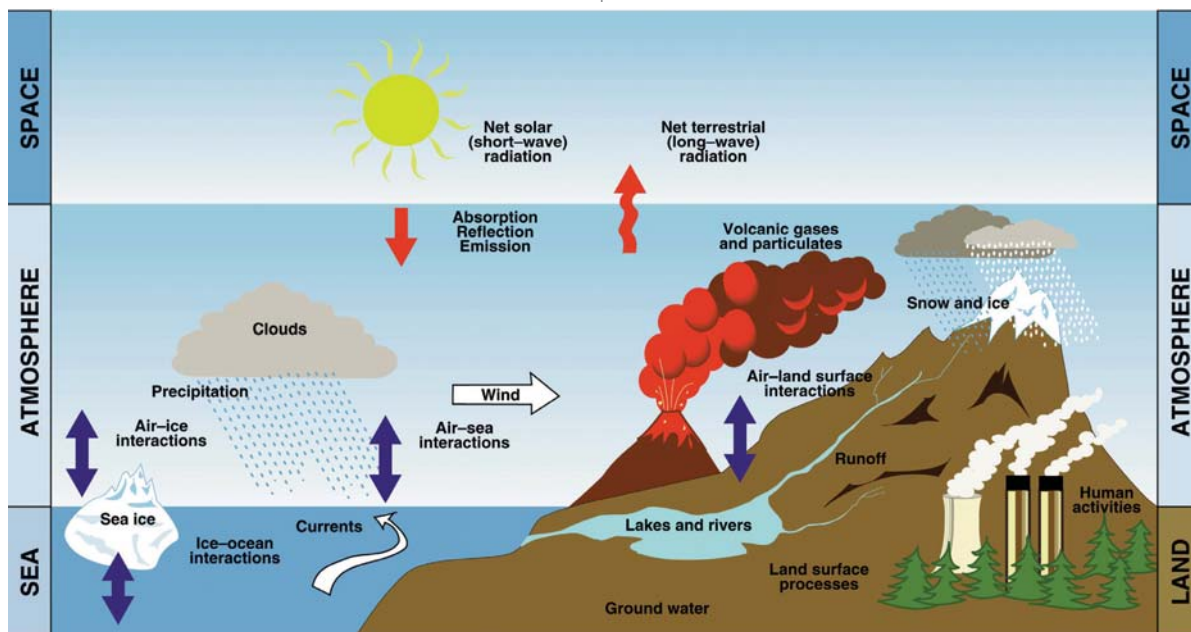


Figure 1.2. A representation of the atmosphere's role in the Earth system [Picture courtesy of WMO].

in determining the rapid changes that are taking place in our environment and so, it is hoped, lay the scientific foundations for the sound policy needed to combat the deleterious effects that our global society is having on the Earth.

1.3 The Goal, the Uses and the Applications of the Results from IGACO

The principal goal of IGACO is to provide the data and information required to quantify the changes taking place in the atmosphere. The purpose of IGACO is to underpin the necessary understanding of the complex interactions within the atmosphere itself and, in the context of IGOS, to provide sound information on the changes taking place within the Earth system as a whole.

The principal uses and application of products expected from the proposed IGACO system include:

1. *detection of long-term man-made trends* in the concentration of greenhouse gases and aerosols related to climate change above natural variability;

2. *better environmental assessments* related to climate, air quality, ozone depletion and the long-range transport of pollution between regions;
3. *better quantification of pollution sources and their atmospheric pathways* to sensitive downwind receptors;
4. *reliable global concentration fields* of the selected chemical variables and aerosols at various altitudes for the study of outstanding problems in atmospheric chemistry;
5. *better predictions of UV intensities* at the Earth's surface both in populated and remote regions;
6. *direct observation of plumes from major events* such as forest fires, dust storms and volcanic eruptions; and
7. *improved regional forecasts of both weather and air quality*, and to provide forecasts in regions where these are unobtainable at the moment.

The reliable and detailed knowledge developed within IGACO will provide the basis for future regular scientific assessments of the state of the atmosphere and of the environment, and of global change.

Entering the 21st century, we are faced with an atmosphere that has changed and is continuing to change. Continued economic growth in the developed and developing world, and the much-needed growth required to alleviate poverty in the under-developed world, will ensure that the effects, particularly in the troposphere, will continue to increase and be of increasing concern for the foreseeable future. Although there appears to be a basic understanding of the major environmental problems, the details and the further effects, which will certainly reveal themselves as the atmosphere continues to change, require monitoring on a global scale.

In order to fulfil efficiently their requirements for protecting the atmosphere, new or revised conventions and protocols must be firmly based on a scientific system analysis, which requires a thorough understanding of how chemical emissions are processed in the atmosphere during transport over the land and sea. The major issues in atmospheric composition and chemistry, which manifest themselves in the problems listed in Table 1.1, are related to *oxidising efficiency* and *air pollution* in the troposphere, *ozone depletion* in the stratosphere and the *coupling between atmospheric chemistry and climate change*. The main objective of IGACO is to provide quantitative and representative information on these well-recognised problems of man's influence on the atmosphere. However, the observing system should also provide the ability for recognising – if not predicting – new and hitherto unknown and potentially harmful developments.

The grand challenge of IGACO itself lies in the prediction of atmospheric chemical change, which is partly disguised by the natural variability of the atmosphere. The observed variability in the concentration of a compound is related to the spatial distribution of its sources and by its atmospheric turnover time, as determined by its bio- and geochemical sinks. The turnover times of atmospheric trace constituents span more than seven orders of magnitude, ranging from less than a second for the most reactive chemicals, such as the hydroxyl radical (OH) which determines the oxidation efficiency of the atmosphere, to several decades for N₂O and CFCs, which are destroyed in the stratosphere. Temporal variability is intrinsically coupled to spatial inhomogeneity by atmospheric dynamics.

The architecture of an efficient integrated observing system must take account of the knowledge of the relevant scales for each compound. For example, it is practically impossible to obtain a representative and accurate estimate of the atmospheric oxidation efficiency by observing OH itself. A sufficiently accurate determination of trends in the concentrations of long-

lived gases, such as CH₄, N₂O or CO₂, on the other hand, can be obtained from a small number of well-placed measurements. Identification of source regions and estimation of source strength for long-lived compounds requires the capacity for detecting small concentration differences on a global scale and with a spatial resolution that allows the resolution of political units and biotopes.

2.1 Air Quality: The Globalisation of Air Pollution

As already indicated in Section 1.1, the quality of air over populated areas is of continuing concern as the degradation of air quality has and will have a major impact on human health, agricultural productivity and natural ecosystems. The photochemical oxidation of hydrocarbons and CO in the presence of NO_x is a major source of tropospheric ozone and other photo-oxidants on both regional and global scales. Abatement applied in several developed countries has reduced the emissions of the precursors, and there are also indications of a corresponding decrease in peak ozone levels. However, the complicating influence of meteorological conditions still clouds the picture.

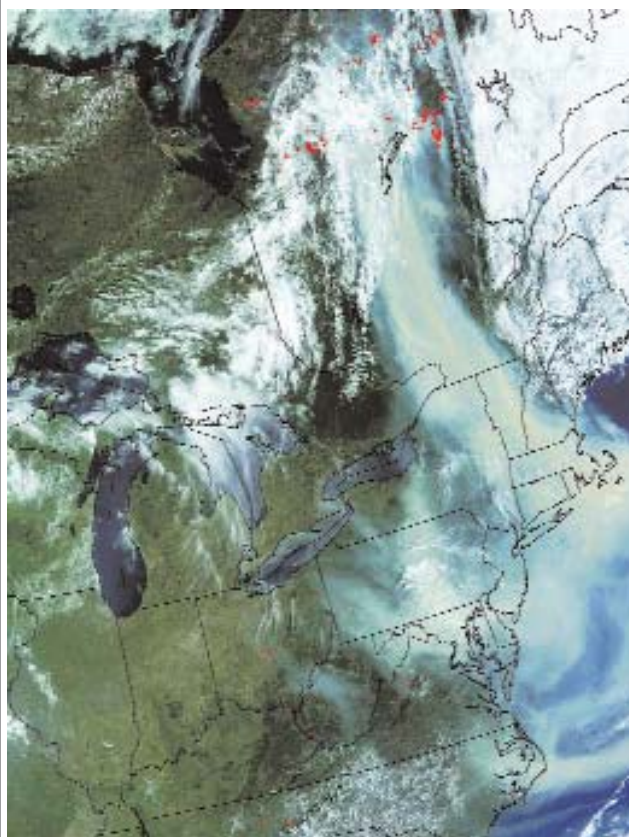


Figure 2.1. Smoke plume from Canadian forest fires as seen by MODIS. [Courtesy of E. Hilsenrath, NASA]

The quantitative understanding of the various sources of aerosols, their chemical composition as a function of size and their sources and sinks is still poor. The regional and inter-continental transport of aerosols, O_3 , CO and NO_2 from biomass burning and from industrial pollutants in both the northern and southern hemispheres has been revealed by satellite and ground-based observations.

While the emissions that cause these various effects are necessarily local, the consequences are regional and global due to atmospheric transport and the chemical processing that takes place during transport. A pollutant can be transported over country boundaries within a day or so and around the hemisphere within a week. The exchange between hemispheres takes of the order of a year in the troposphere. There is growing evidence for a continued increase in the background of ozone in the troposphere, probably due to emissions from both developed and developing countries.

In order to improve existing abatement strategies for air quality laid down by the various international protocols and conventions, it is necessary to obtain information on the local versus regional or inter-continental influence on peak ozone levels, ozone exposure and particle distribution and on the background levels in the atmosphere.

IGACO will contribute to linking local and regional air pollution problems to the issue of global air quality by providing and maintaining the infrastructure for observations (surface observations, profile measurements, satellite data and ancillary meteorological data) that are integrated with a modelling system to produce comprehensive global atmospheric constituent fields. Of similar importance is the subsequent capability for

local and national agencies to feed these global fields into their higher resolution air quality prediction models. The output from these models will, in turn, provide the decision support tools for regulatory enforcement.

From the larger scientific perspective, the upper troposphere and lower stratosphere is a particularly sensitive transition region where the chemical lifetime of ozone is relatively long while the meteorological transport time is short. The large variability in key trace gases and the small scales of important transport processes put a greater demand on the capabilities of the observing system with regard to spatial and temporal resolution. Convection transports pollution from the surface into the upper troposphere, where chemical lifetimes are longer and transport much faster. However, ozone-rich air can also come from the stratosphere. It is presently still not possible to quantify reliably the role of changing surface emissions or the changes in stratosphere-troposphere exchange on the composition of the upper troposphere and hence on climate and air quality. The role of convection is poorly understood and not adequately treated in models. Observational data (cloud parameters for example) are needed that allow parameterisations to be tested.

As new types of chemicals are released, there needs to be a process by which their impact on the global atmosphere can be evaluated. This approach should rest upon accurate observations and a solid scientific understanding of the atmospheric chemical processing system.

Requirement: Global modifications to atmospheric composition must be observed in order to monitor the influence of inter-continental transport on ozone and particles in terms of plumes (dust, ozone, CO, NO_x) and

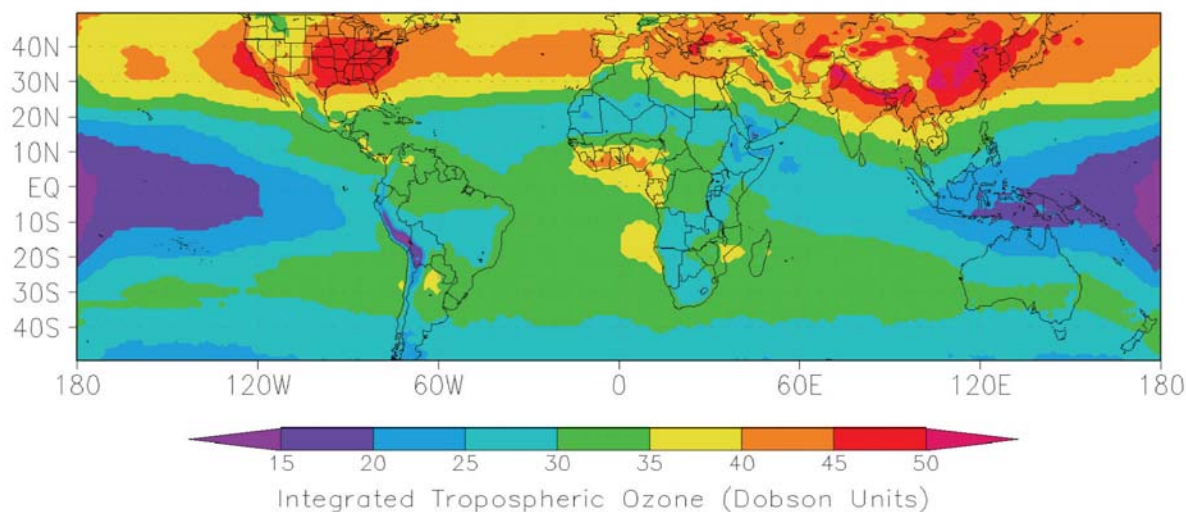


Figure 2.2. Tropospheric ozone, averaged over June, July and August, from combined TOMS and SBUV data, showing major source regions and large-scale transport in the Northern hemisphere. [Courtesy J. Fishman, NASA]

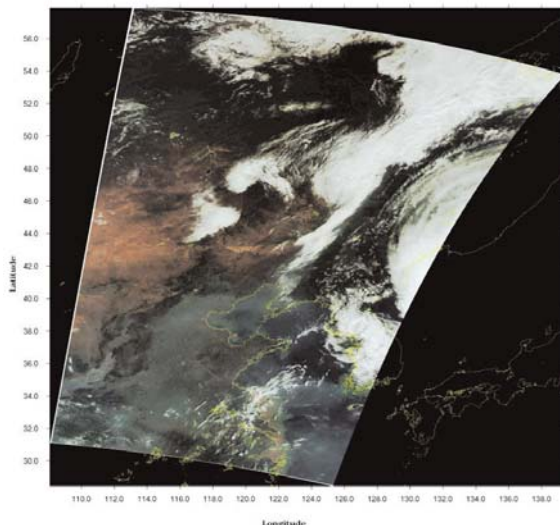


Figure 2.3. The Chinese “brown cloud” as seen by the GLI instrument onboard ADEOS-2. [Courtesy of T. Ogawa, JAXA]

with respect to a gradual change in the global background. High-resolution measurements are required in the upper troposphere and lower stratosphere in order to distinguish the effects due to stratosphere/troposphere exchange and long-range air transportation.

On regional scales, some rationalisation of and a better coordination between the existing ground-based networks (urban, regional and global) is important, including the harmonisation of data archives and data quality. These must include the monitoring of relatively short-lived reactive species (such as VOCs) and meteorological parameters.

This requirement has been emphasised in several recent reports (see for example: US National Academy of Sciences, 2001).

2.2 Oxidising Efficiency: The Atmosphere as a Waste Processor

The efficiency of the atmosphere in functioning as a general disposal system for most natural and anthropogenic emissions has been called into question. In order to maintain its self-cleaning capacity, the atmosphere needs a sufficient concentration of hydroxyl (OH) radicals and we do not know whether these concentrations are sustainable in the face of an ever-increasing burden of emissions, nor do we know when such a limit will be reached.

A much-simplified diagram of the chemical troposphere (Fig. 2.4), illustrates the complexity and inter-connectedness of the atmospheric processes. Photo-oxidation appears in the centre of the diagram; acidification on the right. The species shown in green are greenhouse gases.

Carbon compounds such as CO, CH₄ and other hydrocarbons, together with NO_x, O₃, H₂O and UV radiation, regulate the distribution of the hydroxyl radical (OH). Hydrocarbons and CO deplete OH concentrations in the absence of NO but, on the other hand, enhance OH through catalytic chain reactions in the presence of nitrogen oxides. The strongly non-linear response of the radical balance to changes in chemical composition and UV radiation, and the relatively small spatial and temporal scales at which NO_x

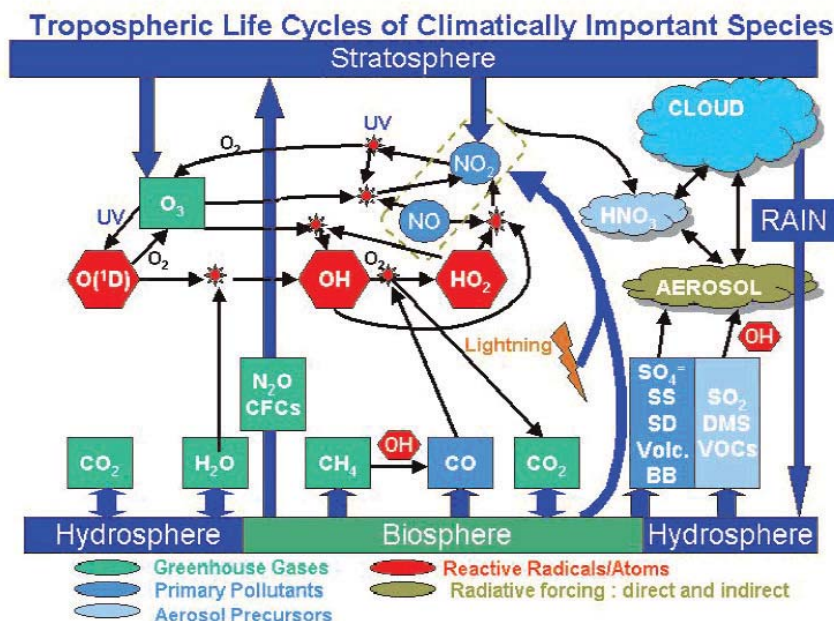


Figure 2.4. A simplified scheme showing the inter-connectedness in the chemistry of the troposphere. [Picture courtesy of G. Brasseur, reworked by L. Barrie, WMO]

and the more reactive VOCs are removed from the atmosphere, represents a real challenge for atmospheric chemistry transport models, if they are to predict accurately the global atmospheric OH field and its response to changing emissions and climate. There are still large uncertainties in the emissions of carbon compounds and NO_x . These concern, in particular, natural emissions from wetlands, vegetation, soils, and forest fires, anthropogenic biomass burning and the NO_x production from lightning. The potential importance of oxygenated organic compounds as sources of free radicals for the upper troposphere was shown only recently. High water-vapour concentrations and high UV-B levels favour the formation of OH in the boundary layer. However, this may be counteracted by low O_3 and NO concentrations in some regions. Recent research has also shown that halogen radicals may also be playing a small but significant role in the oxidising efficiency of the troposphere. Similarly, the nitrate radical, which results in large part from anthropogenic NO_x emissions, can have significant local importance at night.

Changes in the oxidation efficiency are linked to climate and stratospheric O_3 through changes in temperature, water vapour, and UV radiation, as well as lightning, rainfall patterns and changes in atmospheric circulation. Because of the very short lifetimes of OH and the other radicals, the oxidation efficiency of the atmosphere can only be monitored indirectly.

Requirement: Detailed monitoring of the key parameters (NO , H_2O , UV-B, NO_2 , O_3 , CO, CH_4 and other VOCs) that control the concentration of OH on a global scale is required. The emphasis must be on global coverage including remote areas. Particular emphasis must be on the tropics, where most of the oxidation takes place for the longer lived gases, such as CH_4 and CO. Because of the nonlinear nature of the chemical system, vertical profile information between the surface and the tropopause for UV and the short-lived gases is essential for the calculation of the OH concentration. This is currently not possible with existing satellites. The suggestion is therefore to assimilate the tropospheric column information from satellites with local fields from surface networks and aircraft measurements in chemistry-transport models with appropriate chemistry schemes for calculating OH, or to monitor selected trace gases that are removed by reaction with OH and whose sources are accurately enough known.

2.3 The Stratospheric Ozone Shield

One of the lessons taught by stratospheric ozone depletion is that trace amounts of relatively benign chemical compounds can have a large impact on atmospheric chemistry. The ozone hole further demonstrated that the system can spring unpleasant

surprises: ozone depletion at the time was thought to be well understood and the understanding had to be modified to encompass chemical processes and meteorological processes that had hardly been considered before. And while controls on chlorofluorocarbons appear to have stabilised the polar ozone loss, and current models predict that the polar ozone holes will gradually decrease in intensity and duration, it would be foolhardy to believe that the global atmospheric system is incapable of further surprises.

Chemically, the stratosphere as a whole and the ozone layer in particular has undergone major changes within the last half century. Some of these are anthropogenic in origin and are relatively well understood; for others, explanations are still required, despite their unmistakable signal. Anthropogenic chlorine is the primary culprit leading to the thinning of the ozone layer, especially at polar latitudes. However, the frequency of measurements deep in the Arctic vortex remains low. The situation is unsatisfactory given the highly non-linear sensitivity of Arctic stratospheric ozone to cold winters. Over the last two decades in mid-latitudes an average decrease of total ozone of 4% has been found in the northern hemisphere and 6% in the southern. Chemical and dynamical perturbations caused by strong volcanic eruptions make it impossible to derive a linear trend, which highlights the importance of continuous measurements throughout the expected recovery of the ozone layer during the coming decades.

UV irradiance has increased since the early 1980s by 6 to 14% at more than ten sites distributed over the mid and high latitudes of both hemispheres, in agreement with estimates from satellite measurements. The high spatial and temporal variability of the predominant variables affecting ultraviolet radiation at

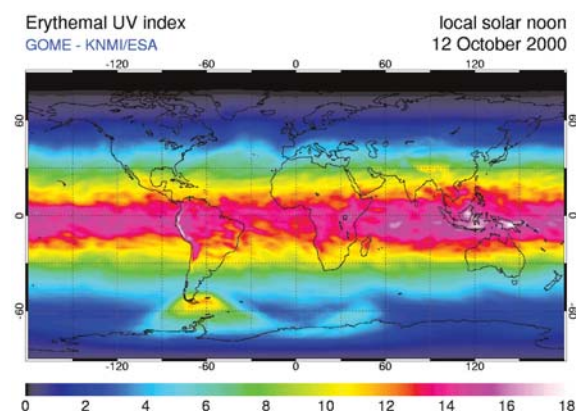


Figure 2.5. Example global map of erythemal ultraviolet radiation index. Note the enhancement at the tip of South America due to the ozone hole. [Courtesy of KNMI]

the Earth's surface and throughout the troposphere (clouds, aerosols, snow/ice cover, total ozone) continue to impede the quantification of surface UV radiation on a global scale. Accurate long-term high-coverage data are required to obtain reliable estimates of the global UV trend.

Continued monitoring of stratospheric chlorine and bromine are essential to assess the recovery of stratospheric ozone to natural levels. However, shorter-lived ozone-depleting substances might have a significant effect on stratospheric ozone when emitted in the tropics, where deep convection may transport these substances rapidly into the lower stratosphere. Estimates of ozone depletion potentials are currently uncertain because of the lack of data on the products of the tropospheric degradation, and also for tropical emissions.

Reactive nitrogen compounds (NO_y) are now attracting attention. Observations and modelling results show that a massive de-nitrification (removal of up to 70% of NO_y) in the Arctic lower stratosphere in the winter of 1999-2000 increased ozone loss by as much as 30% at 20 km altitude in the spring. The origin of lower stratospheric NO_y needs to be better understood. It has been known that some is transported from above, and that it is produced *in situ* at high altitudes by lightning and air traffic. However, there are now also indications that NO_y may be transported into the lower stratosphere from the tropical tropopause region by *quasi*-horizontal, isentropic transport following deep convection from the polluted mid-latitude boundary layer. These recent observations point to the need to measure stratospheric and upper tropospheric NO_x and NO_y both locally and globally.

Stratospheric humidity also seems to have increased appreciably during the last five decades. An average increase in water vapour of 1% per year is estimated to have led to roughly a doubling in stratospheric water vapour at altitudes between 15 and 28 km. Global satellite measurements covering latitudes 60°N- 60°S show a similar trend of 0.6-0.8% per year for altitudes above 25 km, but no significant trend at lower altitudes. The increases in water vapour are substantially larger than can be explained by tropospheric CH_4 trends. Characterisation of stratospheric water vapour trends remains limited by the lack of global long-term measurements.

A long-term stratospheric record of global aerosol coverage has been established from a diverse system of largely uncoordinated ground-based observations covering over two decades and documenting the effect of volcanic eruptions. These observations are augmented by extensive satellite measurements.

However, the issue of whether the background level has ever been reached after volcanic eruptions and whether the known sulphur-containing trace gases are sufficient to account fully for the background level require more comprehensive long-term observations. We require a detailed understanding of the space and time distribution of aerosols, for which no systematic measurement system yet exists.

Because of decreasing temperatures and increasing water vapour, there is a possibility of increased occurrence of polar stratospheric clouds (PSC) in the Arctic stratosphere. A comprehensive measurement system should include PSCs as a synoptic variable, combining lidar observations from the ground and nadir-looking thermal imagery from space.

Current global ground-based observational networks for total column and vertical profiles operated by the WMO Global Atmosphere Watch (GAW) and the Network for Detection of Stratospheric Change (NDSC) have a long history of comprehensive quality observations. However, gaps exist in measurement methodology, network coverage, data archiving and analysis, and these are discussed further in Chapter 4. Sufficient satellite-observation capabilities are available until the end of the lifetime of ENVISAT and EOS-Aura. Continuation of such measurements beyond that time frame is needed for successful monitoring of the recovery of the ozone, and for confirmation of the trend of increasing H_2O .

Requirement: Measurements of vertical profiles are needed for O_3 , water vapour, NO_x , HNO_3 , halogen sources, active halogens and their reservoirs, as well as aerosol properties and methane (CH_4) (the last as a tracer and for investigation of the H_2O budget), together with temperature and ancillary parameters.

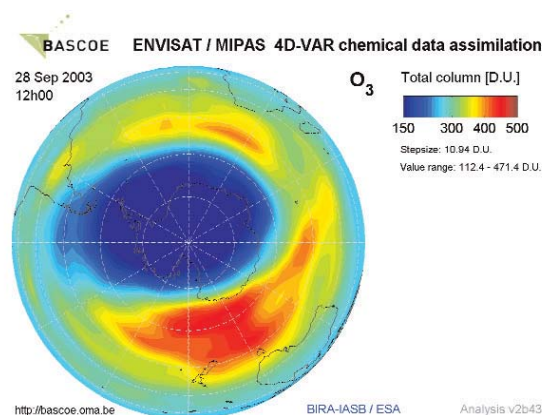


Figure 2.6. The ozone hole, forming each year in early spring over the Antarctic. Assimilated Envisat-MIPAS data. [Courtesy of IASB]

2.4 Coupling Between Atmospheric Chemistry and Climate

The interaction between atmospheric chemistry and climate is one of the most intriguing and important branches of atmospheric research, which scientists are only just beginning to explore. The atmospheric chemistry is driven primarily by the energy of the incoming solar UV radiation, generating atoms and free radicals which initiate catalytic reactions. These reactions are dependent on temperature and transport and hence upon climate. The climate itself depends on the energy input, from greenhouse gases with strong absorption bands in the mid-infrared, which determines the outgoing part of the radiation energy balance of the atmosphere. The distributions and concentrations of greenhouse gases are in turn influenced by atmospheric chemistry.

A number of trace gases produced by human activities add to both the greenhouse effect and to the chemical activity and burden (Fig. 2.8). Ozone plays multiple climate forcing roles, through the absorption of solar ultra-violet radiation and terrestrial infrared radiation. Changes in CH₄ concentration affect climate and the O₃ distribution in the troposphere and in the stratosphere. Aerosols, both natural and anthropogenic, are another important component in the climate system. Tropospheric aerosols produce a direct radiative forcing by virtue of scattering and absorbing solar radiation, and an indirect forcing by changing the radiative properties of clouds. The exact magnitude of the total aerosol forcing, especially the forcing due to black carbon, remains one of the largest unknown factors in climate research.

Evidence is now emerging of the feedbacks between chemistry and climate. The increase in the abundance



Figure 2.7. Polar stratospheric clouds form at very low temperatures. Chemistry within these clouds enables anthropogenic chlorine to establish fast catalytic ozone destruction. [Courtesy: E. Hilsenrath, NASA.]

of greenhouse gases and ozone loss result in lower stratospheric temperatures. Low stratospheric temperatures permit the formation of cloud and aerosol particles, which have implications for both atmospheric chemistry and radiation. Understanding the conditions under which these particles form is important if accurate estimates are to be made for an atmosphere with altered temperature distributions.

It is to be expected that changes in the temperature distribution of the stratosphere will alter the stratospheric circulation and dynamics. These in turn are coupled to the tropospheric circulation and so to the tropospheric temperature distribution and climate.

Climate change may induce changes in the dynamics of the tropopause region, which affect the transport of water vapour and other trace gases from the troposphere into the stratosphere, and thus affect aerosol formation, chemistry, and radiation in the stratosphere. Understanding how stratospheric water vapour might change in the future requires basic understanding of the dynamics of the tropopause region and troposphere-stratosphere exchange mechanisms.

The downward transport of stratospheric ozone has a major effect on ozone concentration in the upper troposphere; in a warmer climate, such transport is expected to increase and hence influence tropospheric chemistry.

The Brewer-Dobson circulation transports tropical air to higher latitudes and influences the composition of the mid- and high-latitude atmosphere. The circulation also affects the structure and temperature of the polar vortex by increasing its temperature and hence affecting polar ozone chemistry. Models predict that in a warmer climate the Brewer-Dobson circulation will be stronger and will affect the composition of both the stratosphere and troposphere.

A variety of chemistry/climate models are being developed to describe and understand the interactions and their implications. The task is challenging because of the time and spatial scales that must be encompassed; from chemical reactions occurring in microseconds to the development of the atmosphere over decades and from metres to tens of thousands of kilometres. Moreover both chemistry and climate are nonlinear and separation of the natural variability of the coupled system and the anthropogenic induced changes is far from trivial.

Understanding the coupling between climate and chemistry is a grand challenge for the next decades, requiring large efforts both from the observational side and from the modelling.

Requirements: a) *The integrated global observation*

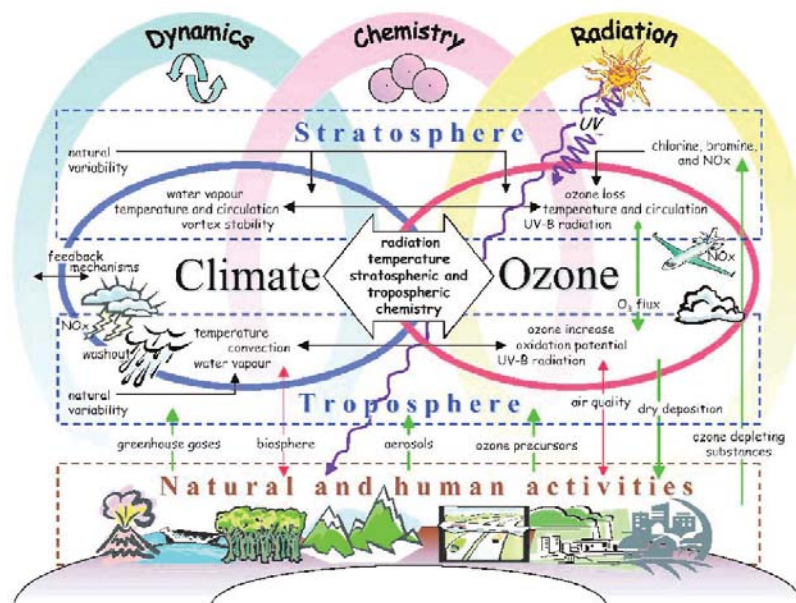


Figure 2.8. Interactions between climate, atmospheric composition, chemical and physical processes and human activities (after Isaksen, 2003).

system for climate-chemistry interaction requires sustained and long-term measurement of the distribution and sources and sinks of greenhouse gases (H_2O , CO_2 , CH_4 , O_3 , N_2O) and related gases (CO , NO_x , NMHC), aerosols (optical properties and size parameter) and meteorological variables such as winds, temperatures

and clouds with appropriate vertical resolution, temporal resolution and global representativeness.

b) Chemistry climate models should be further developed and validated by comparison with these 4-D datasets for atmospheric composition, meteorology and climate.

The global long-term observational system for atmospheric composition comprises four components:

- routine ground-based measurements including *in situ*, remote sensing and balloon instrumentation;
- systematic aircraft measurements;
- satellite measurements with instrumentation viewing in the nadir and limb directions; and
- a data modelling system with assimilation to provide a comprehensive global picture.

The three measurement components, having different strengths and weaknesses, complement each other.

3.1 Ground-based Instrumentation

The last 50 years have seen a revolution in the development of *in situ* techniques for the measurement of atmospheric trace constituents.

Current ground-based networks involve a number of different instrument types:

- measurement instruments operating in modes ranging from continuous to periodic sampling;
- balloon sondes profiling tropospheric and stratospheric concentrations of O₃ and water vapour;
- passive remote sensing spectrometers operating in various wavelength regions providing total column or low resolution vertical profiles of a number of atmospheric gases and aerosols; and

- active remote sensing lidar instruments for high-resolution remote sensing of atmospheric components.

A variety of sophisticated measurement techniques are used, such as emission and absorption spectroscopy, gas chromatography with electron capture, flame ionisation or mass-spectrometric detectors, chemical ionisation mass-spectrometry and chemi-luminescence.

The current role of globally distributed ground stations is to provide high quality observations for:

- detecting long-term trends in atmospheric concentrations;
- monitoring air quality on a regional to global scale;
- understanding the long range transport pathways of pollution;
- evaluating and developing regional to global scale models that include atmospheric chemicals (e.g. local and regional weather and air quality forecast models, long-range transport models and climate models); and
- calibration and validation of satellite observations.

The WMO GAW programme coordinates global ground-based networks measuring greenhouse gases, ozone, UV radiation, aerosols and selected reactive gases. GAW also combines the considerable monitoring activities conducted by member countries with those of global partners such as the Network for Detection of Stratospheric Change (NDSC), NASA

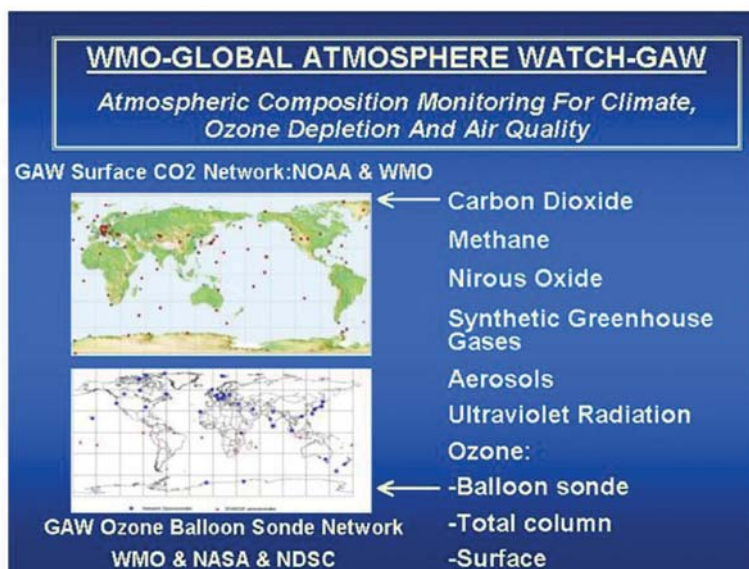


Figure 3.1. Maps of two global ground-based networks for balloon vertical profiling of ozone and carbon dioxide. The networks are collaborations involving WMO members and partners. [Kindly provided by the GAW World Data Centres for Ozone and the UV, and for Greenhouse Gases].



Figure 3.2. The Jungfraujoch laboratory in the Swiss Alps hosts a large number of research projects and contributes to the long-term monitoring of atmospheric composition.

SHADOZ, NASA AERONET, and regional networks such as EMEP, EANET and CAPMoN in order to extend coverage offered for these classes of compounds. The activities since the start with O₃ in 1957 have included the establishment of the WMO member-supported GAW measurement stations, the Central Calibration Laboratories, the Quality Assurance/Science Activity Centres, World Calibration Centres and the five World Data Centre facilities, all under the guidance of scientific advisory groups of internationally recognised experts. However, despite the best efforts, global observations often lack inter-calibration and data harmonisation. Two examples of global ground-based networks maintained through such partnerships are shown in Fig. 3.1.

3.2 Airborne Instrumentation

Aircraft have been used for many years to probe the free troposphere and lower stratosphere on a research basis. The first attempt to utilise commercial aircraft for regular atmospheric observations was the NASA Global Atmospheric Sampling Programme (GASP) in the 1970s. After GASP had been terminated in 1979, the idea was reactivated in the 1990s. In the European programme MOZAIC, automatic instruments for O₃ and H₂O (plus CO and NO/NO_y in 2001) were installed on several commercial aircraft in 1994 and have, since then, provided regular data for the upper troposphere/lower stratosphere (UT/LS) with more than 2000 flights and 4000 tropospheric profiles per year.

Other projects with commercial aircraft include grab sampling packages, flown biweekly, since 1993 between Australia and Japan to provide CO₂, CH₄ and CO data at cruise altitude. The Swiss NOXAR project provided the first regular NO_x data with 500 flights between 1995 and 1996. In CARIBIC, an air freight container with *in situ* instruments for O₃, CO, and aerosol measurements and grab samples for VOCs, CFCs, N₂O and isotopic analysis was deployed during 83 flights between 1997 and 2002. Fig. 3.3 shows a map of the routes covered by the different programmes.

Routine aircraft measurements are currently the most efficient tool for obtaining representative information on the UT/LS and vertical profiles in the troposphere at high resolution and with uniform quality. However, as most ongoing projects are funded under short-term research contracts, they are in continuous danger of being terminated. Continuation is secure

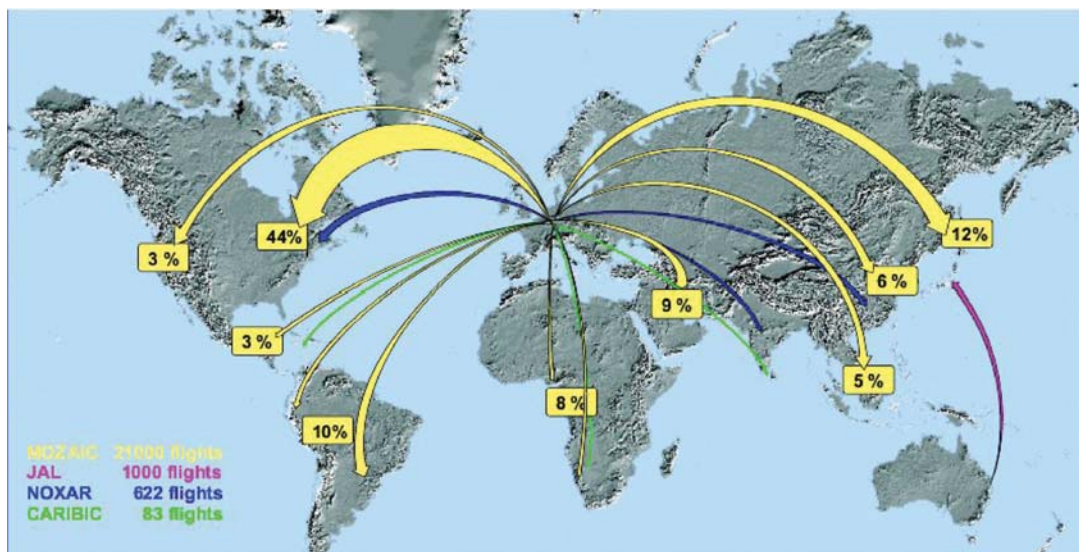


Figure 3.3. Flight routes of MOZAIC (21000 flights 1994-2003, yellow), JAL (ca 1000 flights 1993-2003, purple), NOXAR (622 flights, blue), and CARIBIC (83 flights 1997-2001, green). [Picture courtesy of Andreas Volz-Thomas, Jülich]

for the Japanese grab-sampling programme until 2017 and until 2014 for CARIBIC, which will start operation with an enhanced measurement suite (O_3 , H_2O , CO , NO , NO_2 , NO_y , $HCHO$, aerosol size and composition, BrO , halocarbons, VOCs, CH_4 , CO_2 , Hg , and O_2) in 2004 with 1 to 2 flights per month. These measurements are valuable for IGACO, but cannot provide a truly global picture of the free troposphere and UT/LS. An extension of the MOZAIC-type measurements with many flights per day and extension of the global coverage is urgently needed.

There are also a few initiatives for using small aircraft on a regular basis. Examples are the NOAA/CMDL initiative for collecting vertical profiles of CO_2 and CH_4 , several times per week at a number of sites around the World, the RIALTO initiative of the Canadian Environment service for making regular measurements of O_3 and possibly NO_2 , $HCHO$, SO_2 , BrO by MAXDOAS (multi axial DOAS) in the lower troposphere on regular flights of the Canadian coastal guard, and a few pilot programmes that perform aerosol scattering and aerosol absorption profiling on a routine basis.

3.3 Satellite Instrumentation

Compared to point measurements made by ground-based sensors and one-dimensional routes followed by airborne instrumentation, the advantage of spaceborne sensors is their global three-dimensional coverage and regular repeat cycle. Traditionally data-sparse regions above the oceans, polar zones and developing countries are observed with the same uniform data quality as the industrialised countries. Space data are the natural feed for global atmospheric models.

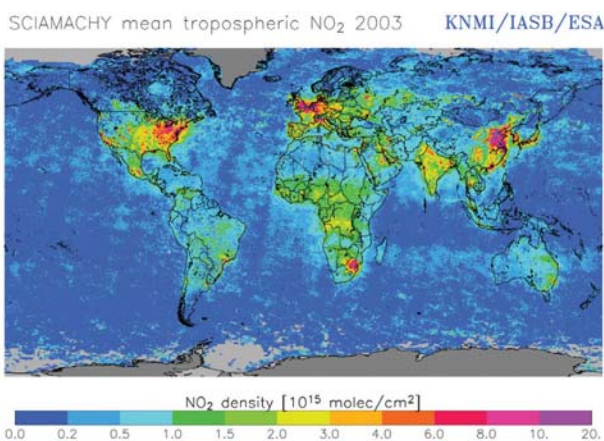


Figure 3.4. Global tropospheric NO_2 . A yearly average of observations taken by Sciamachy in 2003. The industrial sources are clearly visible. [Image courtesy of H. Kelder, KNMI]



Figure 3.5. Earth's limb from Space. Satellite observations in this viewing direction can provide high resolution profile data of the stratosphere and upper troposphere. [Courtesy E. Hilsenrath, NASA]

The first satellite measurements of atmospheric trace gases and aerosols focused primarily on the investigation of stratospheric ozone depletion. At these altitudes, observations in both nadir and limb viewing geometry are not adversely affected by clouds. Stratospheric profile information can best be acquired by viewing in limb geometry. Nadir-viewing instruments provide total column measurements, but some vertical resolution can also be achieved if the response function of the species being measured exhibits an altitude dependence. Research satellites, such as UARS, ENVISAT, AURA, have contributed strongly, and will contribute further, to stratospheric chemistry science. Long-term records have been obtained for total column/low-vertical-resolution ozone (TOMS, GOME), high-resolution stratospheric ozone profiles (SAGE) and a limited set of other species. The transition to operational monitoring has been initiated for low-resolution ozone and some other total column products obtained from nadir-viewing instruments on NPOESS and METOP. There is not yet any provision for a system covering high-resolution stratospheric profiles of ozone and other constituents, such as aerosol and active and reservoir species of the nitrogen and halogen families.

High-resolution profiling of the UT/LS region is essential for monitoring climate–chemistry interactions, and stratosphere–troposphere exchange, and supports the interpretation of total-column information from nadir sounders and the assessment of global pollution issues. A first step in this direction will be made by the exploitation of current research

satellite data (ENVISAT, AURA), but with limitations on vertical resolution, global coverage, and the ability to screen clouds. A long-term monitoring system is yet to be put in place.

Monitoring of the tropospheric composition is needed for climate and air-quality applications and for the assessment of the oxidising efficiency of the atmosphere. The mid and lower troposphere is difficult to access with limb-sounding, due to the opacity of the atmospheric layers above and the high sensitivity of this viewing geometry to cloud and aerosol contamination. Nowadays, information on the troposphere is extracted from nadir-pointing instruments that were primarily designed for the stratosphere (e.g. GOME). Some impressive results have been obtained by exploiting, for example, temperature-induced altitude dependences in the spectral signatures of trace gases. Products include ozone profiles with two to three tropospheric pieces of information and tropospheric column data for species whose stratospheric abundance is either low or can be estimated with models. As an example, Fig. 3.4 shows a global map of the tropospheric column of NO₂ which is of high importance for air quality and oxidising efficiency. However, for tropospheric applications, the existing instruments suffer from several severe limitations, which must be eliminated in the next generation of sensors:

- The horizontal resolution is too low for detailed assessments and local predictions of air pollution; and the pixel size is too large, leading to a loss of much of the data due to cloud contamination.
- Vertical resolution is a persistent challenge for tropospheric measurements from space. To date, no concerted effort has been made to combine all means to obtain more than total columns, such as
 - co-registered measurements of nadir-viewing UV-visible-near-infrared instruments delivering total column data with mid-infrared high resolution spectrometers providing some height information from the mid-troposphere upwards;
 - combination of nadir- and limb-viewing observations, in order to obtain lower/mid tropospheric columns by subtraction of the integrated upper tropospheric and stratospheric limb-sounder profiles from the total columns obtained with nadir sounders;
 - high-spectral-resolution, near-infrared observations with full global coverage to obtain detailed information on climate gases and aerosol.
- The temporal sampling of the existing systems is too low to capture the pronounced diurnal variations, in particular in the boundary layer.

The choice of the orbit depends on the specific data requirements. High-vertical-resolution profiles – only possible in the stratosphere and upper troposphere – are obtained using the limb-sounding technique, which requires a Low Earth Orbit (LEO). All present satellites are in LEO, typically at 800 km, most of them Sun synchronous; i.e. the satellite passes over the same location at the same local time. The repeat cycle depends on the spatial sampling, but daily repeat cycles are routinely achieved globally with a single satellite; two satellites could provide a twelve-hour time sampling.

To obtain the time resolution necessary to encompass variations in tropospheric constituents, the geostationary orbit (GEO) (at ca 36000 km) is another option. The long distance requires the use of larger telescopes, but the continued observation of a given area by a single pixel (the exception being polar regions), allows the use of integration to recover a satisfactory signal-to-noise ratio while still achieving a time resolution of the order of an hour. Thus, diurnal variations resulting from both chemical transformations and regional transport are readily observed. Meteorological satellites have successfully exploited this orbit for tropospheric measurements of clouds and other parameters required for weather prediction. Three to four GEOs would be required for global coverage. An alternative would be several LEO polar-orbiting satellites, their number depending on the required time resolution and the orbit height. The trade-off between the two options is determined by temporal resolution, coverage required, signal-to-noise ratio, size and stability of footprint, and by technical and cost considerations.

One point worth emphasising, however, when considering space observations, is that to realise its potential a space-based system requires an ongoing ground-based calibration measurement system. In the past, this requirement has not been included in satellite missions.

3.4 Modelling and Data Assimilation

Modelling and data assimilation bring together the host of measurements to provide optimal temporal and spatial reconstructions of key atmospheric constituents. Such a synthesis yields invaluable information on the consistency and the quality of measurements so that trends, variability and sources and sinks of these species can be quantified. In turn, improved models will be developed to provide better predictions as well as a better reconstruction of past changes. Eventually, it is envisaged that the synthesis of information will be used by operational agencies, policy makers and the general public as part of a decision-support system.

CTM Models. The basic tool in an Integrated Observation System is a chemical transport model (CTM). Such models are based on the physics of fluid dynamics and transport, input from an emissions module to provide data on man-made and natural biosphere emissions, and a chemical module, which may include hundreds of species and chemical reactions. More complex processes involving aerosols, liquid processes in cloud droplets, and reactions on the surface of solid particles, are usually treated through parameterisation. Such models of the atmospheric dynamics and chemistry have the ability to calculate missing and under-sampled parameters and to generate continuous and self-consistent fields of atmospheric trace constituents.

CCM Models. Long-term descriptions of the atmosphere can be obtained with chemistry-climate models (CCM), where the evolution of atmospheric composition and climate is modelled simultaneously and interactively, taking into account the feedbacks between chemistry and climate. Present models are reasonably successful, although there are weaknesses in the parameterisations, the emission scenarios assumed, and the chemical mechanisms employed.

There is an inherent problem of averaging - using values within a model grid box, which vary markedly within the dimensions of the box. This problem is equivalent to the problem of the representativeness of a particular measuring station for a model grid square, and indeed the representativeness of a satellite column measurement for the average concentration.

Data Assimilation. An integrated observation system will combine the information contained in the measurements and the theoretical model by means of a data assimilation scheme in order to generate data of improved quality. The heritage of data assimilation is the development of weather-forecasting models that minimise the difference between model evolution and measurements. An essential element in the scheme is the quantification of errors that may originate from either measurement or model; the gain in information content is continually assessed. The output will be the best available knowledge of the state of the atmosphere; it must be validated by independent measurements.

Assimilation of satellite measurements of chemical species, notably stratospheric ozone, into an operational weather forecasting system is a relatively

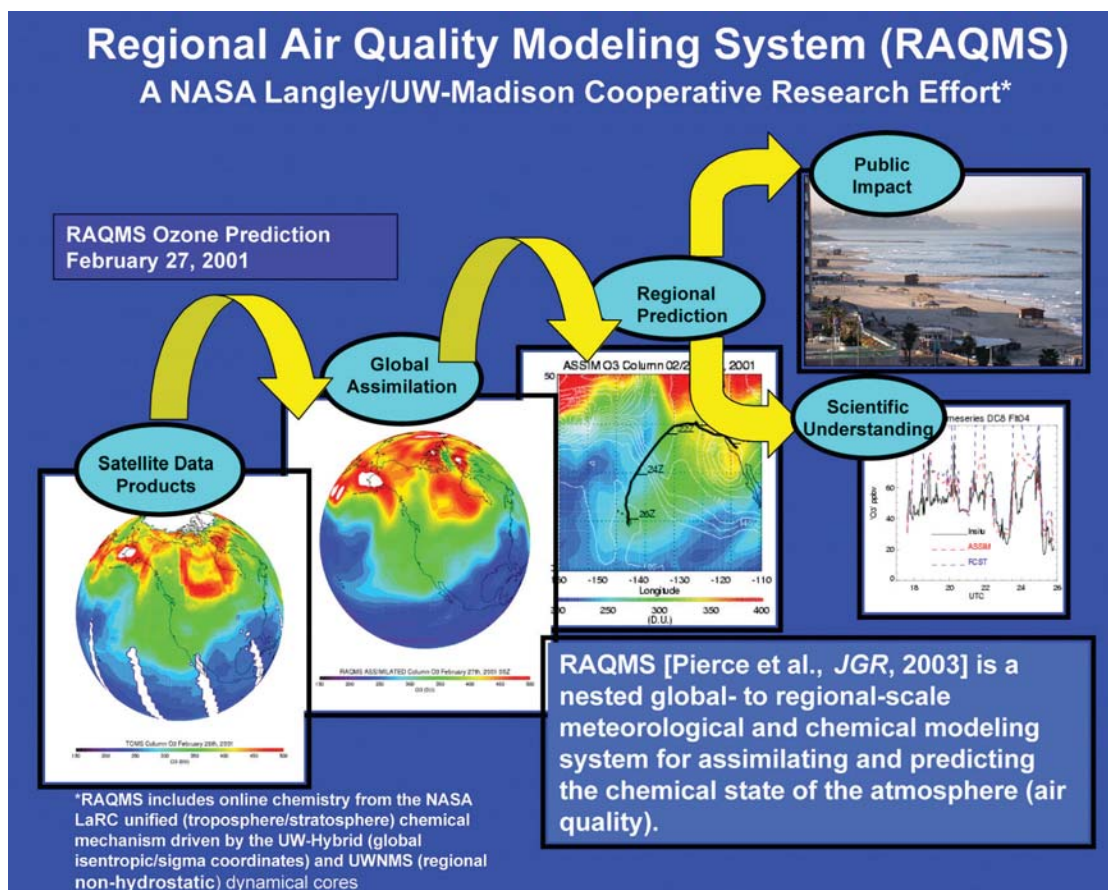


Figure 3.6. An example of a regional air-quality forecast system based, in this case, mainly on satellite measurements.
[Figure courtesy of Jack Fishman, NASA]

recent development. It has been used in numerical weather prediction to improve significantly the assimilation of satellite radiances. It has been successfully applied to forecast the evolution of the ozone hole and to forecast the UV exposure of the Earth's surface. The daily UV forecasts delivered by many meteorological organisations are a visible demonstration of this technique for the general public.

New challenges in data assimilation and CTM model development are posed by the inverse modelling of emissions. An important application is the improvement of the greenhouse gas emission inventories derived from atmospheric concentration distributions. The successful application of this technique had been limited by the sparse data available from ground-based stations. With the advent of space-based measurements much better global coverage can be achieved. However, the improvement of the accuracy of space-based measurements and how such remote observations relate to observed boundary layer and surface observations remain critical scientific challenges.

Air Quality Forecasting. An important application of an integrated observation system is the generation of an air pollution forecast or a chemical weather forecast. Such forecasts would be useful for Environmental Agencies and the general public as potential decision-support tools, which can be used to implement regulatory actions on emissions from sources such as vehicular traffic and electric power generation, and to provide health warnings for polluted areas. An example of a forecast system using mainly satellite data is shown in Fig. 3.6.

Utilisation of air-quality-forecast models requires near-real-time chemical data acquisition, typically within three to six hours. For rapidly varying species, sampling must be frequent enough to capture these variations. For example, species that exhibit significant diurnal variation require measurements at sub-daily and, ideally, hourly intervals. Building an operational system for chemical weather-forecasting imposes challenging requirements on the quality and consistency of the input data, the quality and reliability of the data generation and distribution, as well as the long-term continuity prospect of the entire system.

4.1 Introduction

The problems in atmospheric chemistry have been simplified in Chapter 2 into four issues:

- tropospheric air quality;
- the oxidation efficiency of the atmosphere;
- stratospheric chemistry and ozone depletion; and
- chemistry climate interactions.

It is possible to study and monitor these issues by observing a number of chemical compounds, aerosol properties and other parameters. A list of these variables and chemical species, and the issues in which they are involved is given in Table 4.1. They were chosen on the basis of the following criteria :

- a. the chemical species or variable plays an important role in one or more of the four key atmospheric chemistry issues and there is added value in its incorporation in an integrated global observation system;
- b. it is now possible, or likely to be possible, to measure the atmospheric constituent or parameter globally and on a long-term so as to achieve a synergism between satellite, ground-based and aircraft observations, and model assimilation systems.

The list in Table 4.1, chosen on the grounds of likely feasibility, is far from exhaustive. There are many other desirable variables such as precipitation chemistry and aerosol composition, some of which are already being addressed in the ground-based networks. More detailed information on the choices and on the present and likely future state of the measurements is given below.

4.2 Specific Chemical Species and Other Parameters

In this section, the state of the observational system for most compounds is portrayed as a timeline diagram. Each diagram summarises the past and future state of satellite observations, non-satellite observations and status of assimilation (i.e. smart interpolation) of global chemical transport models based on forecast/climate models. The shading of the timelines indicates a best estimate of how well a particular measurement provides quality global coverage.

For non-satellite measurements, *demonstration mode* means that there is an incomplete network of measurements that are not archived or publicly available, through a data centre; *pre-operational mode* means that an approximation of a global network is available, but the observations do not yet fully meet quality assurance and inter-network comparability

requirements; *operational mode* means that a global network exists in which data is of high quality and available from a central archive.

Satellite missions are mentioned only if the records exceed three contiguous years (for past missions) or if the projected lifetime is at least 3 years (for ongoing and future missions).

Ozone: O₃

O₃ is a key atmospheric constituent in all four air-chemistry issues. In *air quality*, atmospheric boundary layer ozone is of concern for human health and for damage to vegetation, and many environmental agencies set regulatory standards to minimise the effects on human health and vegetation. In the boundary layer, O₃ is produced by the photochemical oxidation of VOCs in the presence of NO_x and sunlight; it adds to O₃ transported down from the stratosphere to the middle troposphere. For the *oxidising efficiency* of the troposphere, ozone plays a role as an indicator of photochemical production, similar to that occurring in the boundary layer close to sources. As an oxidant, it converts SO₂ to sulphate aerosols in clouds; it is the primary source of OH radicals: when photolysed it produces O (¹D) which reacts with water vapour to form OH. For *climate*, O₃ in the middle to upper troposphere is an important greenhouse gas. In the *stratosphere*, it acts as an absorber of ultraviolet solar radiation, providing a UV shield for the Earth, as well as an energy source. The shield is, however, vulnerable to catalytic destruction of O₃ by halogen compounds such as CFCs.

Here, because of differences in scale and observational techniques, timelines are shown for O₃ in the troposphere and in the stratosphere. It must, however, be emphasised that the interface between these two atmospheric regions, the upper troposphere and the lower stratosphere (UT/LS), is of much importance in atmospheric transport and cannot be neglected in planning the IGACO system.

The timelines show that there are numerous satellite stratospheric O₃ measurements of demonstration quality up until 2010. After 2010, two operational quality LEO satellites are available for global ozone characterisation.

There are operational ground-based networks for vertical profiles and total column O₃ operated by GAW. Stratospheric ozone vertical profiles (see Fig. 3.1) are obtained from the global balloon-sonde network maintained by GAW and its contributing partners NASA/SHADOZ and NDSC. It is operational, but needs considerable effort to fill gaps in coverage and to complete the establishment of a formal measurement protocol and quality assurance programme. The GAW

Chemical Species	Air Quality	Oxidation Efficiency	Climate	Stratospheric Ozone Depletion
O ₃	✓	✓	✓	✓
CO	✓	✓	–	–
j(NO ₂)	✓	✓	–	–
j(O ¹ D)	✓	✓	–	–
H ₂ O (water vapour)	✓	✓	✓	✓
HCHO	✓	✓	–	–
VOCs	✓	✓	–	–
Active nitrogen: NO _x = NO+NO ₂	✓	✓	–	✓
Reservoir species: HNO ₃	✓	✓	–	✓
N ₂ O	–	–	✓	✓
SO ₂	✓	–	✓	–
Active halogens: BrO, ClO, OClO	–	–	–	✓
Reservoir species: HCl, ClONO ₂	–	–	–	✓
Sources: CH ₃ Br, CFC-12, HCFC-22, halons	–	–	–	✓
Aerosol optical properties	✓	–	✓	✓
CO ₂	–	–	✓	–
CH ₄	–	✓	✓	✓
Critical Ancillary Parameters				
Temperature	✓	✓	✓	✓
Pressure	✓	✓	✓	✓
Wind speed (u, v, w)	✓	✓	✓	✓
Cloud-top height	✓	✓	✓	✓
Cloud coverage	✓	✓	✓	✓
Albedo	✓	✓	✓	✓
Lightning flash frequency	✓	✓	✓	✓
Fires	✓	✓	✓	–
Solar radiation	✓	✓	✓	✓

Table 4.1. Key atmospheric chemical species and the relevant environmental issues. The table gives a list of the atmospheric constituents to be targeted in IGACO. Tick marks indicate their importance to the four atmospheric challenges. Also included are aerosol optical properties – a broad categorisation which encompasses the scattering and absorption of solar radiation by particles of all sizes.

4 ATMOSPHERIC CHEMICAL OBSERVATIONS FOR TARGETED PARAMETERS

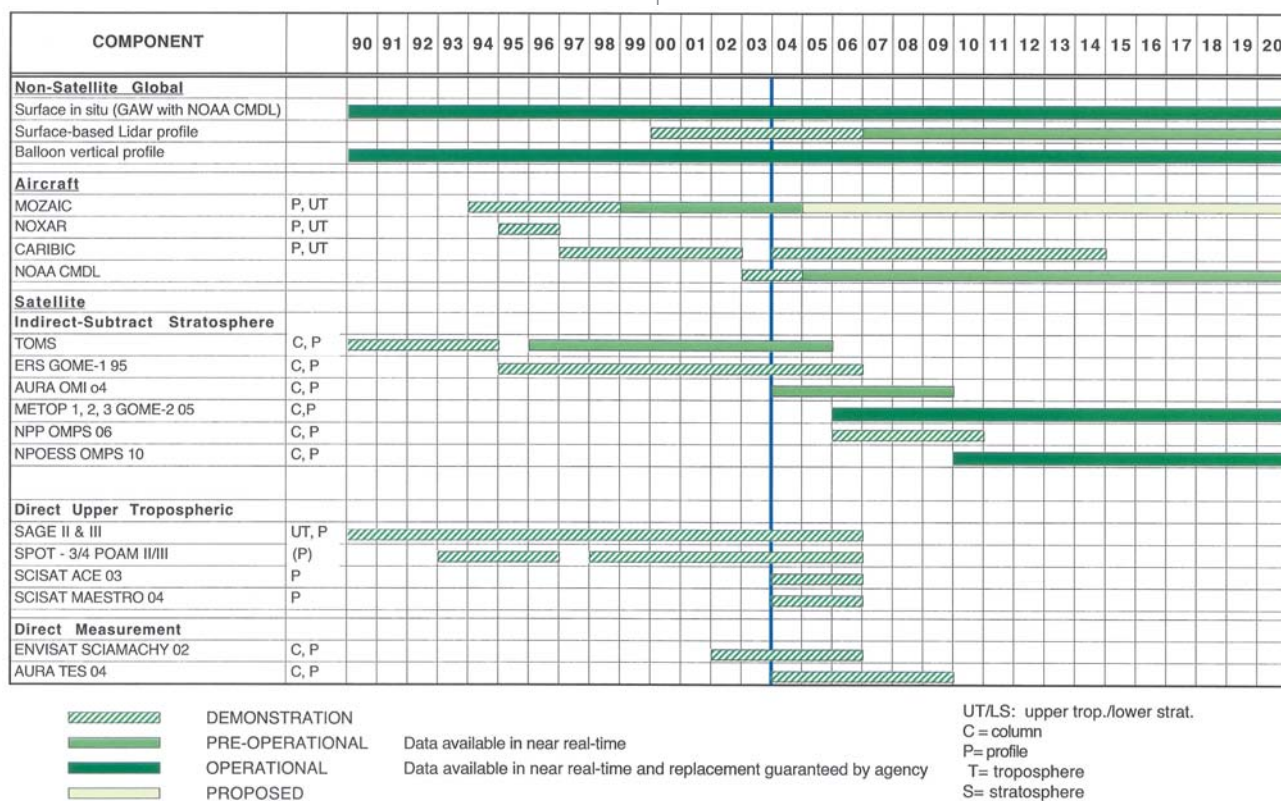


Figure 4.1. An overview of satellite, ground-based and aircraft measurements for tropospheric O₃

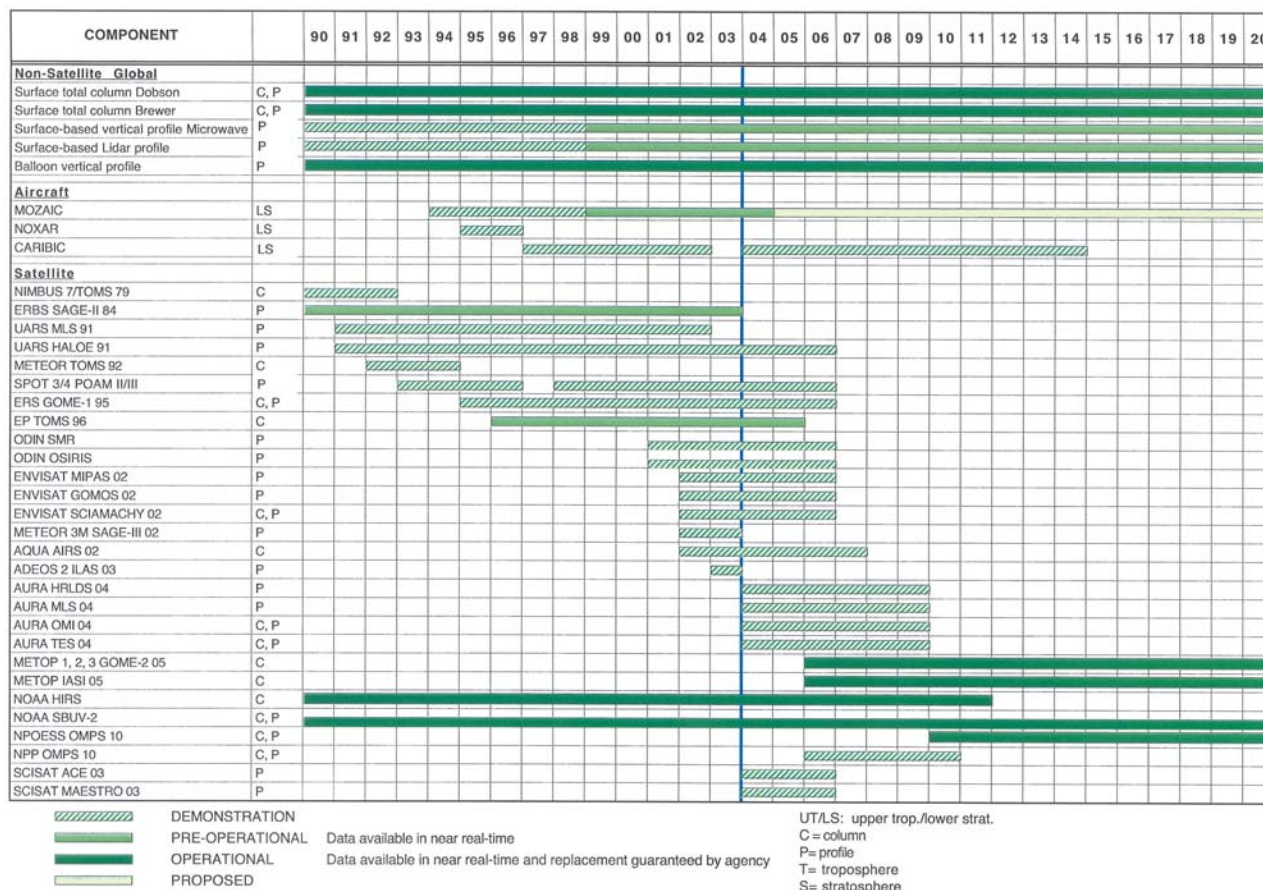


Figure 4.2: An Overview of satellite, ground-based and aircraft measurements for stratospheric O₃.

ESTIMATED GLOBAL COLUMN OZONE NETWORK: 2003
Stations with data submitted since at least 1 Jan. 1999

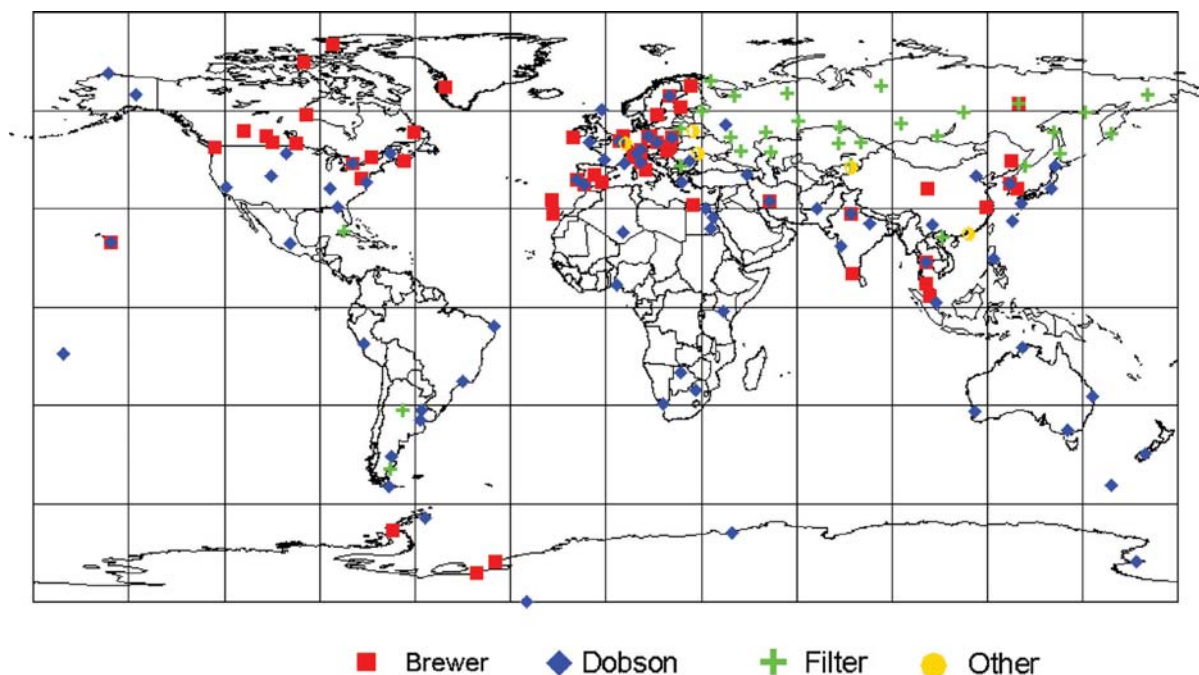


Figure 4.3. Global Column Ozone Network, 2003 [Courtesy of WMO/GAW World Ozone and UV Data Centre, Meteorological Service of Canada, Toronto.]

total column network as indicated by those submitting data is shown in Fig. 4.3. Today global coverage is attained through measurements by three instrument types: the Dobson and Brewer spectrophotometers and the Russian filter instrument.

GAW is building a systematic programme of routine calibration of instruments linking them to global standard reference instruments maintained by WMO members and collecting observations at the World Ozone and UV Data Centre in Toronto. There is a need to replace the Russian filter instruments with the more accurate Dobson or Brewer and to encourage those stations that are not submitting data to do so. This would greatly enhance the network in many regions showing gaps in Fig. 4.3.

Systematic ground-based observations of *in situ* ozone are made at 37 GAW global and regional stations. They are linked to a global standard at NIST in Washington and quality assured by the GAW World Calibration Centre for *in situ* ozone in Zurich, Switzerland. The data are archived at the World Data Centre for Greenhouse Gases in Japan. There is much work to be done with partners to collect a global data set. Many environmental agencies support ozone measurements at regionally representative sites. This is one gap that needs to be filled by IGACO.

Systematic measurements of *in situ* O₃ from commercial aircraft are now available for selected routes from Europe through the MOZAIC and CARIBIC programmes (see

Section 3.2). NOAA/CMDL is beginning a programme of routine vertical profiling of the lower troposphere at a large number of locations using light aircraft. Aircraft observations are in the demonstration stage because coverage is not comprehensive, vertical profiles are limited and the data are not yet freely available. Expansion of these observations is needed.

Atmospheric forecast models that include stratospheric ozone are currently assimilating non-satellite and satellite observations to produce UV forecasts. They are in demonstration mode and moving into pre-operational mode as more data become available in real time. These same models can be used in a re-analysis of all observations (real time and non-real time) as the integrated observational system improves and delivers more data to a central archive.

Carbon Monoxide: CO

CO is a reactive gas that displays a strong temporal and spatial variation. Its origin is predominantly anthropogenic, being mainly produced by the combustion of fossil fuel in the northern hemisphere and bio-mass burning in the southern hemisphere. It has a direct influence on the greenhouse gas concentrations of CO₂ and O₃; indirectly, it affects the concentration of CH₄, because CO is the major sink for OH, which breaks down CH₄. The global average lifetime of CO is about two months, yielding significant hemispheric spread after emission.

Ground-based CO concentrations are monitored globally at 28 GAW stations shown in Fig. 4.5. These measurements are routinely quality assured by the World Calibration Centre for CO in Zurich and linked to the GAW global reference standard for CO maintained by NOAA/CMDL. Stations are located at regionally representative sites and data are available from the World Data Centre for Greenhouse Gases in Japan. There are large gaps in the global GAW surface *in situ* network that could be filled through collaborative agreements with regional networks. Although total column CO observations are possible at the NDSC monitoring sites that operate FTIR instruments, there is no systematic programme to do this. The data is critical for calibration and validation of satellite observations and evaluation of models. Total column observations would provide corroborating data for checking models.

Systematic measurements of *in situ* CO from commercial aircraft are now available for selected routes from Europe through the MOZAIC and CARIBIC programmes (Section 3.2), including vertical profiles in the troposphere. NOAA/CMDL is beginning a light-aircraft programme of routine vertical profiling of the lower troposphere at a large number of locations using automated flask sampling. Airborne observations are in the demonstration stage because coverage is not comprehensive, vertical profiles are limited, the data are not yet freely available, and the long-term continuation is still uncertain.

The best satellite CO data available is that from the Terra MOPITT instrument, which was specifically designed to measure CO column and vertical profiles. This instrument is operating on 4 out of the 8 channels originally intended yet still provides useful global

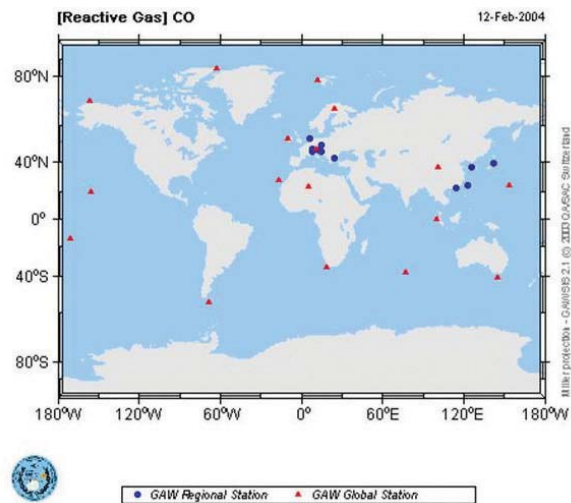


Figure 4.5. Global and Regional stations in GAW that measure CO and are quality-assured by the GAW World Calibration Centre in Zurich Switzerland.

coverage. Satellite data are also available from ENVISAT SCIAMACHY and Aura. In the future, METOP IASI, to be launched in 2005, will provide operational column CO but not profiles. Satellite CO column measurements are expected to be produced at 5 and 10% precision, depending on location, with a 3 to 6 day repeat cycle, which will be well within the observation and modelling requirements.

Ultraviolet Radiation: UV-A, j(NO₂) and UV-B, j(O¹D)
 Ultraviolet radiation is an important parameter throughout atmospheric chemistry and thus plays a role in all four issues. The photo-dissociation of ozone leads to the formation of the hydroxyl radical, which initiates the removal of pollutants through photo-oxidation and so directly determines the oxidising



Figure 4.4. An overview of satellite, ground-based and aircraft measurements for CO.

efficiency of the troposphere. It also directly affects the air quality. Similarly, the photolysis of NO_2 is the only source of atomic oxygen in the troposphere needed for the formation of ozone.

UV radiation also has an indirect effect on climate through the oxidising efficiency, which impacts upon climate-related species.

The appropriate variable for atmospheric chemistry is the *actinic flux*. It is required in the wavelength ranges responsible for the photo-dissociation processes, namely UV-B (290-315nm), which determines the photolysis rate of ozone, $j(\text{O}^1\text{D})$, and UV-A (315-400nm), which determines $j(\text{NO}_2)$. Actinic flux conversion from measurements of UV irradiance at the surface is currently feasible by an empirical method with an accuracy of about 10% if the sky conditions are not known. Accuracy is much improved when the sky conditions are known.

The current state of the global measurement system and integration of observations is as follows:

a. For satellite observations.

UV-A and UV-B irradiances at the Earth's surface can be determined for clear-sky conditions from absolute Earth radiances in the UV, measured from nadir-viewing satellites. The accuracy is still limited to, at best, around 20%, which is equivalent to that achievable with radiation transfer models for clear-sky conditions. Corrections due to cloudiness and tropospheric aerosol are generally taken from dedicated satellite observations unless this information is already available with adequate geographical resolution. Significant work is needed to improve the reliability of surface irradiance determinations.

b. For non-satellite observations.

Earth surface UV irradiance networks are currently operated on a global scale, with the usual sparseness in the southern hemisphere and over the oceans. Studies are in progress to convert irradiance data to actinic flux data at the surface with a reasonable degree of precision, for complex atmospheric and sky conditions.

Vertical distribution of tropospheric actinic flux measurements are only made on a campaign basis by means of balloon or airborne instruments. The comparisons with radiative-transfer-model calculations demonstrated the need to complement the measurements with relevant atmospheric parameters such as ozone concentrations and aerosol vertical distribution in the troposphere, with a resolution of 2 to 3 km. Reliable satellite information for those para-

meters, when available, will be a valuable tool with which to calculate actinic fluxes adequately in the troposphere using radiative transfer models. Such determinations will require validation campaigns with *in situ* measurements.

Many gaps still exist in the measurements and in the integration methodology, mainly due to the lack of adequate and detailed information on the various atmospheric conditions with the appropriate altitude resolution. Albedo measurements are also required to provide a full set of coherent data. The integration of cloudiness is one of basic quantities needed from satellites to calculate the UV irradiance and actinic fluxes.

Water Vapour: H_2O

Water vapour plays a key role in all four issues. It is a major greenhouse gas. It plays a role in air quality and oxidation efficiency through its reaction with O^1D atoms to form OH radicals, which in turn initiate the photo-oxidation reactions that cleanse the atmosphere. H_2O also influences the size and composition of hygroscopic aerosol particles and, through the equilibration between the vapour and liquid phases, it has impacts on the optical properties of aerosols and, hence, on visibility and radiative forcing, as well as the heterogeneous reactions of gases with aerosols.

Finally, water vapour is a source of clouds and precipitation, which directly affect the climate, and which disperse, transform and remove soluble gases and aerosols in the atmosphere. Increasing concentrations of water vapour in the lower stratosphere are of growing concern because of its influence on the radiation balance in this climate-sensitive region, and the potential changes in the homogeneous and heterogeneous chemistry affecting O_3 .

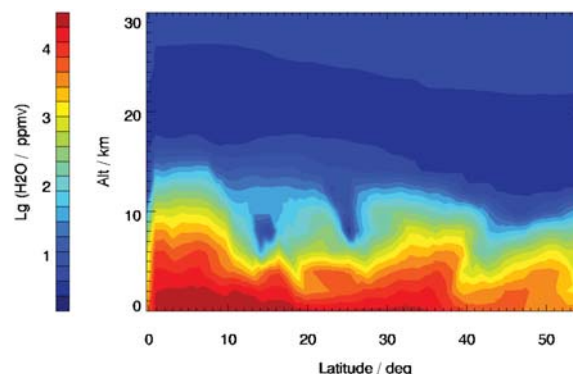
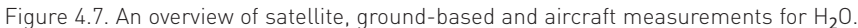


Figure 4.6. Example of a meridional cross-section of water vapour mixing ratio, taken from meteorological analysis, mixed with climatology above 20 km. The key role of water vapour and its high variability in horizontal, vertical and time domains make it a primary target for global high-resolution measurements. [Courtesy of B. Kerridge, RAL]



comprehensive, vertical profiles are limited, the data are not yet freely available, and the long-term continuation is still uncertain.

Assimilation modelling capabilities for water vapour are operationally advanced and will be able to take immediate advantage of any improvements in the global observation system, leading to major enhancements in predictive capability and climate modelling. An important shortcoming is the inability of current models to reproduce the large ice supersaturation found in the upper troposphere by the aircraft measurements.

HCHO is a reactive trace gas in the lower troposphere; some is emitted directly into the atmosphere, but its principal importance is as a product from the atmospheric oxidation of CH_4 and other VOCs. It is removed by subsequent oxidation, and also by deposition and rainout.

Background mixing ratios of HCHO are generally well below 1 ppbv and therefore difficult to measure on a routine basis. However, elevated mixing ratios of up to 10 ppbv or more can be found near forested areas where HCHO is produced from the oxidation of isoprene and terpenes. Elevated HCHO concentrations



Figure 4.8. An overview of satellite, ground-based and aircraft measurements for HCHO.

have also been observed in and downwind of urban areas as a result of anthropogenic VOC emissions and in regions of biomass burning.

Techniques to measure HCHO from ground-based sites have been developed in the last 10 years, but the number of operational measurements is limited and there remain open issues with data quality. Measurements with long-path DOAS and MAXDOAS are being made at a few locations. There are presently no routine aircraft measurements for HCHO.

GOME-1 and SCIAMACHY have demonstrated that the tropospheric column of HCHO can be measured from LEO satellites. OMI aboard EOS-Aura will provide additional LEO measurements. HCHO exhibits an appreciable diurnal variation and is highly variable on small spatial scales, so that observations on short timescales are necessary to use it fully within models.

The total-column HCHO measurements, now obtainable from spaceborne sensors, provide information on

regional source strengths of both natural and anthropogenic VOCs throughout the World. They provide useful validation products for CTMs.

The importance of HCHO to IGACO is in the validation of models of oxidation and photo-oxidant production in the troposphere.

Volatile Organic Compounds (VOCs)

VOCs are emitted by the biosphere and are products of the petroleum industry. They are removed from the atmosphere by reaction with the hydroxyl radical and subsequent photo-oxidation to CO_2 and H_2O . VOCs are responsible, together with NO_x , for the photochemical formation of O_3 and other photo-oxidant pollutants including secondary aerosol. The lifetime of VOCs ranges from several months in the case of C_2H_6 to hours for the most reactive ones such as isoprene or anthropogenic olefins. The main importance of VOC is in the lower troposphere and especially over, and downwind of, populated areas. Whilst the oxidation products of VOCs are distributed globally, the need to monitor reactive

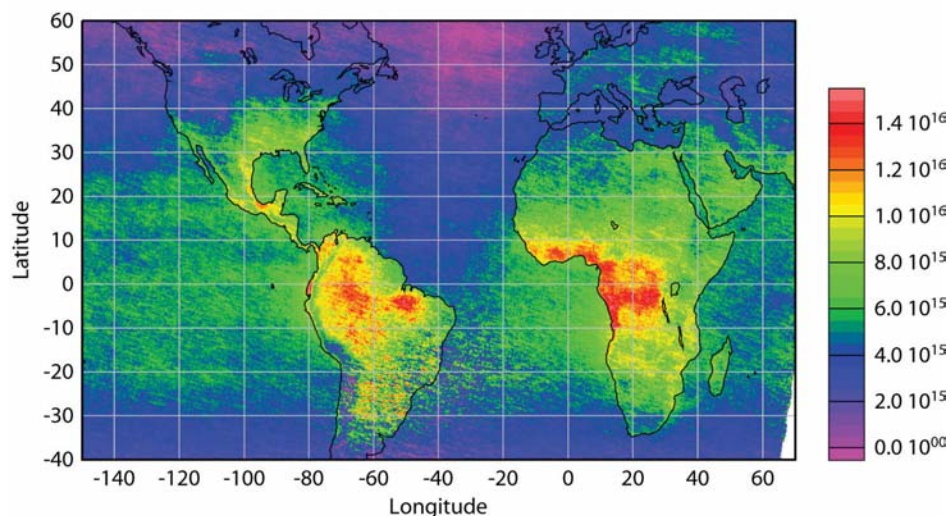


Figure 4.9. Vertical columns of HCHO derived from SCIAMACHY measurements in 2003. The highest values are mainly due to isoprene emissions from tropical rainforests above Africa and South America. [Courtesy of F. Wittrock, University of Bremen]

VOCs themselves is confined to the regions of emissions, because of their short atmospheric lifetimes.

Of the hundreds of VOCs emitted into the atmosphere, only a limited number are currently measured routinely at a few well-equipped ground stations and from a small number of aircraft platforms. Only a few components of the VOC family (e.g. C_2H_6) have the potential to be observed by current or proposed satellite instruments (e.g. MIPAS). However, because of its relatively low reactivity, C_2H_6 is not a suitable indicator for anthropogenic and biogenic VOC emissions related to air quality. Furthermore, the observational challenge focuses on the distribution of VOCs in the boundary layer, which is currently not feasible from space. However, satellite measurements of important VOC reaction products, such as organic aerosol, are a target for future satellite missions (e.g. CALIPSO).

Assimilation of VOC data is currently restricted to special cases in selected emission regions and cannot be done on a global basis due to the lack of data and suitable modelling tools. WMO/GAW is currently harmonising the fragmented networks and has established a World Calibration Centre for VOC in Germany.

Obviously there is a great demand to further develop and integrate the existing ground-based regional VOC monitoring networks, including regular measurements from small aircraft.

Active Nitrogen: $NO_x = NO + NO_2$

The reactive nitrogen species, NO and NO_2 , often referred to as NO_x , play a critical role in tropospheric, stratospheric and mesospheric chemistry.

NO_x is released into the troposphere by the combustion of fossil fuels, biomass burning and

lightning. It is also produced during the oxidation of NH_4^+ , and in the reduction of NO_3^- in the biosphere. NO_x is removed from the troposphere by gas-phase and heterogeneous reactions, which form nitric acid, nitrates and other species; these are subsequently deposited to the Earth's surface and/or biosphere, or they are rained out. NO_x in the stratosphere originates from the reaction of $O(^1D)$ atoms with N_2O , transported from the troposphere, and from the transport of NO from the mesosphere and thermosphere, where it is produced by ion-molecule reactions.

In the troposphere, NO_x is a dominant factor in the *in situ* photochemical catalytic production of O_3 , and in determining the amount of hydroxyl radical, the most important tropospheric oxidising agent. Thus it plays a critical role in determining the oxidising efficiency of the troposphere. NO_x provides a measure of the amount of the reactive nitrogen present, but the ratio of the two species (NO/NO_x) is highly variable depending primarily on the actinic flux, which photolyses NO_2 to NO, and on the amount of O_3 present, as well as that of peroxy radicals (HO_2 and RO_2). In the stratosphere, atomic oxygen and other radicals, which react with NO to form NO_2 , also influence the ratio.

In the stratosphere and lower mesosphere, reactive nitrogen impedes the efficiency of catalytic destruction cycles involving halogens by the formation of temporary reservoirs, which bind active chlorine and bromine, for example chlorine and bromine nitrates ($ClONO_2$ and $BrONO_2$). At high latitudes in winter and spring, the stratosphere is de-nitrified by the formation of polar stratospheric clouds, through the reactions of N_2O_5 , resulting in the release of labile halogen compounds. In spring the returning sunlight photolyses these, releasing halogen atoms which then participate in efficient catalytic cycles removing O_3 and producing



Figure 4.10. An overview of satellite, ground-based and aircraft measurements for tropospheric NO_2 .

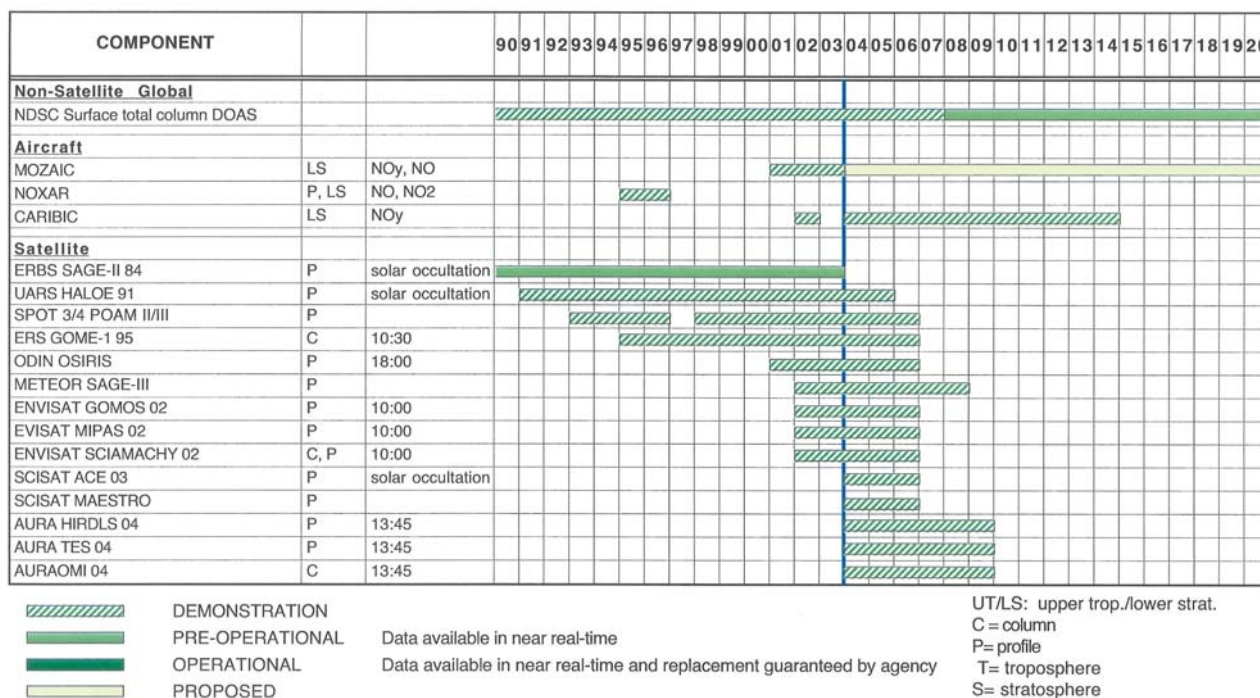


Figure 4.11. An Overview of satellite, ground-based and aircraft measurements for stratospheric NO₂.

the so-called ozone hole. The amount of reactive nitrogen present in polar regions is critical to the efficiency of the formation of intermediate products that inhibit the formation of the seasonal Antarctic ozone hole.

The monitoring of reactive nitrogen is complicated by the fact that NO is easier to measure by *in situ* methods, whereas the NO₂ amount and distributions can be retrieved by remote sensing with instrumentation located on the ground, or balloon or aircraft borne, and satellite platforms. Differential optical absorption spectroscopy (DOAS) is generally used to retrieve NO₂ concentrations. A reasonable estimate of NO_x can be obtained through the simultaneous measurement of either NO or NO₂ with O₃ and *j*(NO₂), which provide the necessary information to calculate the NO/NO₂ ratio.

Although NO₂ has a significant diurnal cycle in the stratosphere, except in winter, NO_x is not expected to vary much, NO and NO₂ being inter-convertible. In contrast NO_x varies greatly in the troposphere during the day due to the variability of the source and sink processes, which depend strongly on atmospheric conditions. Thus frequent sampling of NO_x is required to achieve a realistic representation during the day.

Remote sensing from LEO satellite platforms yields the vertical distribution in the stratosphere, the total atmospheric column and the tropospheric column. An important byproduct of the spaceborne NO₂ measurements has been the capability of mapping the

regions having significant tropospheric sources of tropospheric NO_x.

Ground-based measurements of the near-surface concentrations of NO and NO₂ are made at some GAW/baseline stations and by environmental agencies; however, the data quality achieved by some networks is insufficient for the IGACO needs. The remote sensing of NO₂ columns by DOAS over long light paths is a growing application area. The NDSC provides a limited set of accurate column measurements of NO₂ around the globe in relatively clean tropospheric environments. Additionally, the same instrumentation is deployed within other networks in the polluted troposphere. The development of MAXDOAS (multi axial DOAS) now enables the atmospheric column to be separated into upper and lower parts.

Systematic measurements of *in situ* odd nitrogen species (NO_y) from commercial aircraft are now available for selected routes from Europe (Section 3.2). The observations are in the demonstration stage because coverage is not comprehensive and there is a need to include both NO and NO₂ in the *in situ* measurements.

There are gaps in the measurements of NO_x in that there are no routine aircraft measurements for UT/LS, and there is no comprehensive ground-based network. While the global tropospheric and stratospheric monitoring of NO₂ will be a major contribution to IGACO, much development work is still required to obtain profiles routinely.

Nitrogen Reservoir Species: HNO_3

HNO_3 is the most important reservoir for odd nitrogen in the atmosphere. It is eliminated from the stratosphere by downward transport to the troposphere and rain out. HNO_3 plays an important role in heterogeneous chemistry in polar stratospheric clouds (PSCs) since PSC type I particles contain HNO_3 , either in the form of liquid ternary solutions with water or as solid HNO_3 hydrates. Sulphuric-acid aerosols, which find their way into the stratosphere from large volcanic eruptions, convert NO_x and the temporary odd nitrogen reservoir, N_2O_5 , into HNO_3 .

Ground-based measurements of nitric acid column abundances were initiated in the 1950s, with *quasi*-continuous monitoring since 1985; they are among the priority tasks of the NDSC. Vertical profiles have been measured in the stratosphere initially with balloon-borne *in situ* filter sampling and later with infrared spectrometry.

Routine aircraft measurements of NO_y provide proxy information on HNO_3 , which comprises 90% of NO_y in the lower stratosphere.

Spaceborne measurements were initiated in 1978 by LIMS on board Nimbus 7, followed by ATMOS from 1985 to 1994 and CRISTA in 1994 and 1997. Global time series have been obtained by means of satellite observations (UARS) from 1992 to the present day. Other observations are currently made with MIPAS on board ENVISAT.

Nitrous Oxide: N_2O

Nitrous oxide originates from both natural Earth land-surface sources and anthropogenic sources. It is a greenhouse gas (Global Warming Potential of ~ 300 relative to CO_2) with a tropospheric mean residence time of 120 years. Its reaction with $\text{O}(^1\text{D})$ oxygen atoms in the stratosphere yields NO , which makes it the major source of all the nitrogen oxides involved in stratospheric ozone chemistry. Its concentration is increasing in the atmosphere at a rate of 0.25% per year. There are major unknowns in its global cycle that remain to be resolved.

N_2O is monitored *in situ* at 13 ground-based stations by GAW and NASA/AGAGE. In addition, total column N_2O is measured by NDSC at many locations. With its long lifetime, the present ground-based measurement systems are sufficient for IGACO. N_2O profiles will be measured with upcoming satellite instruments; grab samples are taken on some CARIBIC flights. NOAA/CMDL is beginning a programme of routine vertical profiling of the lower troposphere at a large number of locations using light aircraft and automated flask sampling.

Halogen Sources: CH_3Br , CFC-12, HCFC-22, halons

Chlorine finds its way into the atmosphere as HCl in volcanic eruptions; salt (NaCl) forms aerosol over the oceans and sea coasts as well as over salt flats. It is thought that some atomic chlorine can be formed from NaCl in the polluted boundary layer and that this may play a small role in the initiation of photo-oxidation of VOC. Removal is by conversion to HCl followed by rainout or deposition.

Chlorofluorocarbons (CFCs) are solely of anthropogenic origin, having been extensively used as refrigerants, propellants, and for foam blowing. Because of their chemical stability, CFCs are only destroyed in the stratosphere, where they are photolysed by short-wavelength UV radiation to form chlorine atoms which catalyse the removal of stratospheric O_3 . Due to the Montreal Protocol and its amendments, the industrial production and usage of CFCs is now limited to a few special sectors and, as a result, the amount of CFCs in the troposphere appears to have peaked. The concentrations in the stratosphere are expected to continue to rise for some time and then only to decrease slowly.

The hydrochlorofluorocarbons (HCFCs), which are now being produced to replace the CFCs, are removed in the troposphere by OH radicals and, in consequence, only a small fraction of the emissions reach the stratosphere, leading to a much reduced ozone destruction potential. However they have appreciable global warming potential and are thus not as ideal replacements for CFCs as originally thought. After a sharp initial increase in the nineties, the quantity of HCFCs in the troposphere appears to have levelled off.

The primary sources of organic bromine in the troposphere are surface emissions of methyl bromide, halons and short-lived organo-bromine compounds. Halons are primarily used as fire-extinguishing agents and account for about 40% of total organic bromine. The mixing ratios of halons continued to increase in 2002 at a mean rate of 0.1 ppt Br per year. This rate is slower than that observed in the mid-nineties, when Br from the most abundant halons (1211 and 1301) was increasing at a rate of 0.2 to 0.3 ppt per year.

Methyl bromide has both natural and anthropogenic sources and accounts for about 50% of the global organic bromine emissions. Measurements and trends in methyl bromide determined since 1992 are poorly documented so there are large uncertainties in the global trend of total organic bromine in the troposphere.

In the stratosphere, few measurements of bromine compounds were performed during the 1990s and thus the inorganic bromine burden in this region can

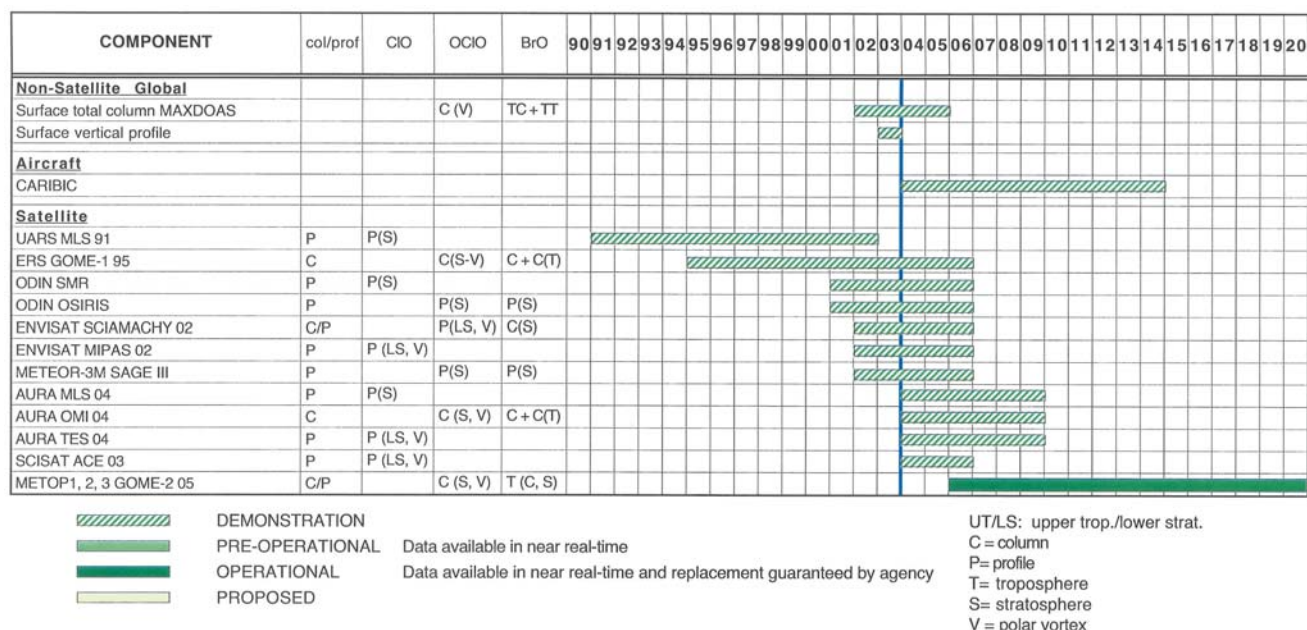


Figure 4.12. An overview of satellite, ground-based and aircraft measurements for ClO OCIO and BrO.

only be estimated indirectly. However, the few measurements suggest that, as-yet unmeasured, organic bromine compounds from natural tropospheric sources are being transported into the stratosphere.

Halogen Reservoir Species: HCl, ClONO₂

HCl is a major reservoir compound for inorganic stratospheric chlorine. It is formed by the reaction of hydrocarbons with chlorine atoms, Cl, resulting from the photodissociation of organic or inorganic chlorine compounds by solar UV radiation. It is removed from the stratosphere by downward transport to the troposphere, where wet deposition removes it.

HCl was first measured in the stratosphere with a balloon-borne infrared spectrometer in 1974, followed by ground-based measurements of column abundances starting at the end of the 1970s. These ground-based observations are currently included in the key species monitored by the NDSC. Vertical profile retrieval has been recently developed from the same observations and could become operational by 2010.

Spaceborne measurements were initiated with the Space Shuttle flights from 1984 to 1994. Global time series are obtained by means of satellite observations (UARS/HALOE).

Stratospheric long-term trends in HCl column abundances have been determined from ground-based observations, and since 1992, from satellite measurements. The updated ground-based observations show that the total column abundances of HCl reached a maximum value around 1998 and are

currently starting to decrease. These results provide robust evidence of the impact of the regulation of the Montreal Protocol and its subsequent amendments on the inorganic chlorine loading of the stratosphere.

Active Halogen Compounds

Active halogen compounds comprise the elements (Cl and Br atoms), and some oxides (ClO, OCIO, BrO). Their principal importance is in the stratosphere where they participate in chain reactions which remove O₃. They are formed by the photo-dissociation of the natural and anthropogenic source and reservoir compounds (see above) with low-wavelength UV light. They are removed from the atmosphere by gradual conversion to HCl and other halogen compounds, diffusion into the troposphere, followed by rain out or deposition.

ClO, OCIO and BrO can be observed from space. Satellite measurements include NASA-UARS-MLS measurements of ClO, ESA-ERS-2-GOME-1 and ESA-ENVISAT-SCIAMACHY measurements of BrO and OCIO, and ESA-ENVISAT-MIPAS measurements of ClO.

Ground-based DOAS and MAXDOAS measurements to obtain profiles of ClO, BrO and OCIO are being made at a limited number of sites.

Cl and Br atoms cannot presently be observed directly in the atmosphere.

The concentrations and trends of these constituents depend upon the concentrations and trends in the precursors and reservoir compounds. However, the details of their variation with atmospheric conditions are essential in the verification of the models used to



Figure 4.13. An overview of satellite, ground-based and aircraft measurements for SO₂.

describe the atmosphere, and to obtaining an accurate picture of the state of the atmosphere.

Sulphur Dioxide: SO₂

SO₂ is a key species in sulphate particle formation. It is emitted directly by volcanoes, and produced from the oxidation of biogenic compounds. Anthropogenic emissions of SO₂ stem mainly from fossil-fuel combustion. They continue to be globally very large, despite the effective desulphurisation technology developed and applied in most developed countries. The oxidation of SO₂, heterogeneously on aerosol and cloud droplets or by homogeneous gas-phase reactions, produces sulphuric acid (H₂SO₄) which, in the gas phase, generates aerosol including cloud condensation nuclei. In the stratosphere, SO₂ is an

important precursor for stratospheric aerosol. SO₂ has a very short lifetime in the troposphere and highly variable sources.

Measurements of SO₂ and sulphate (SO₄²⁻) are made with a network of ground-based stations around the World. The QA/QC is variable.

The data from the NASA TOMS instruments were used first to detect the SO₂ emitted by volcanoes from space. GOME and SCIAMACHY have demonstrated that SO₂ from pollution in the troposphere and from volcanic eruptions can also be measured from space. The measurements of OMI will also provide SO₂ data.



Figure 4.14. An overview of satellite, ground-based and aircraft measurements for CO₂.

Carbon Dioxide: CO₂

CO₂ is the most important anthropogenic greenhouse gas. It is a long-lived atmospheric species and therefore it is well mixed in the atmosphere. Its global mean concentration has increased by more than 30% since pre-industrial times. The available ground-based measurement network operated by WMO/GAW and partners (Fig 3.1) is adequate to detect trends in global mean and meridional concentrations on a seasonal basis. However, it is not adequate to deduce carbon sinks on a regional or continental scale using inversion modelling.

Significant uncertainty exists in the CO₂ uptake by the oceans and the terrestrial biosphere. In the planetary boundary layer over vegetation, measurements of CO₂ display a strong diurnal variation, especially during the growing season. The daily cycle induces significant horizontal variability in the CO₂ column over land. Forests, oceans and other sinks may prove highly variable in CO₂ uptake from year to year depending on meteorological conditions.

Thanks to the GAW scientific advisory group on greenhouse gases and the activities of the carbon-dioxide measurements group, gaps in the measurement network are being filled. Routine ship measurements are being expanded. Aircraft measurements made by Japan Airlines and the Meteorological Research Institute of Japan between Tokyo and Sydney are continuing into a second phase, with continuous measurements replacing flask sampling. NOAA/CMDL is beginning a programme of routine vertical profiling of the lower troposphere at a large number of locations using light aircraft and automated flask sampling. In addition, steps are being taken to encourage researchers engaged in carbon exchange flux studies to link their measurements to the GAW global CO₂ reference standard maintained at the Central Calibration Laboratory operated by NOAA/

CMDL and to submit their data to the GAW World Data Centre for Greenhouse Gases in Tokyo. The carbon flux community is the key to the implementation of the Carbon Cycle Theme of IGOS.

Measurements of CO₂ total column amount over cloud free regions by satellites has the potential to fill some gaps left by the non-satellite ground-based and aircraft. The Orbital Carbon Observatory satellite will be launched by NASA in 2007. It is the first satellite dedicated to measuring CO₂ in the atmosphere with sufficient precision to be of use in carbon sink studies. In the meantime, limited information on CO₂ column density may be forthcoming from ENVISAT and EOS-AURA. It is a challenge to decide which combination of ground-based, aircraft and satellite observations most effectively meets the needs of carbon-cycle researchers. The inverse modelling of CO₂ needs to be explored for establishing global source-sink magnitudes.

Methane: CH₄

Current anthropogenic CH₄ emissions dominate natural emissions by a factor of about 2. CH₄ is now an important greenhouse gas with its present contribution to global radiative forcing being about 17%; concentrations have more than doubled since pre-industrial times. CH₄ is oxidised in the atmosphere by OH constituting its main sink. In turn, CH₄ is one of the main sinks for OH, the prime oxidant of the troposphere, and these interactions provide a firm link between chemistry and climate. CH₄ is also an important source for stratospheric water vapour.

CH₄ emissions are subject to the Kyoto Protocol. The CH₄ growth rate (approx. 0.4% per year) has varied and has slowed during the past decade. As a result, the share of CH₄ in the annual increase in greenhouse forcing decreased substantially in the nineties. Large uncertainties (typically a factor of 2) prevail on

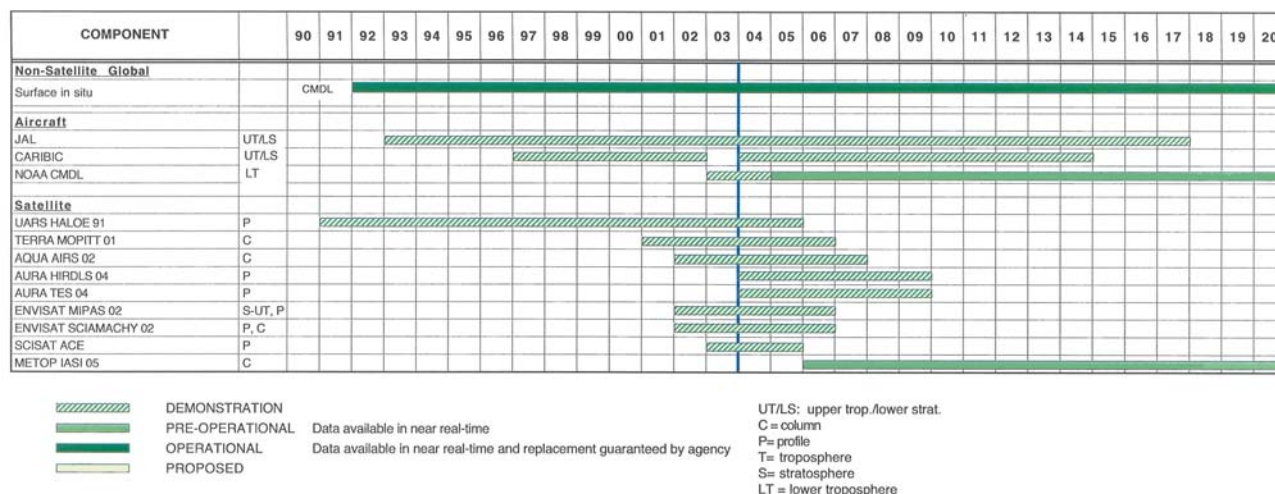


Figure 4.15. An Overview of satellite, ground-based and aircraft measurements for CH₄.

magnitudes of both natural and anthropogenic CH₄ sources, and consequently on the causes of past variations as well as future trends.

The measurement-accuracy requirements for methane are determined by the role that methane plays in climate, stratospheric ozone depletion and oxidising efficiency issues. From the climate perspective, the emission reduction commitments of the Kyoto Protocol, which is approximately a 10% reduction by the 2008-2012 period from 1990 levels, require a measurement accuracy of 1% or better. Owing to its relatively short atmospheric lifetime (ca. 8 years) compared with other greenhouse gases, methane concentrations display relatively large variations in space and time. The spatial resolution needed to capture these variations should be of the order of the source/sink scale of 100 km or less. The temporal resolution should capture at least the seasonal variation and avoid potential biases caused by poor diurnal sampling or cloud cover.

The present ground-based network for methane coordinated by GAW is almost identical to that for carbon dioxide shown in Fig. 3.1. It is sufficient to quantify the global spatial and temporal distribution of methane, but not for reducing uncertainties in CH₄ emissions using inverse modelling techniques. However, improvements in the global network described above for CO₂, namely the addition of ship, flux tower and aircraft measurements, will address this gap in the next decade.

Aircraft measurements made by Japan Airlines and the Meteorological Research Institute of Japan between Tokyo and Sydney are continuing into a second phase with continuous measurements replacing flask sampling. NOAA/CMDL is beginning a programme of

routine vertical profiling of the lower troposphere at a large number of locations using light aircraft and automated flask sampling.

Satellite observations of tropospheric CH₄ are presently limited in accuracy and scope. Recently launched and soon to be implemented instruments (SCIAMACHY, TES, IASI) will improve the situation, but the limitations on accuracy, spatial sampling and sensitivity mean that emissions cannot be quantified well enough with satellites alone. The ground-based and aircraft measurement network improvements are likely to be the most effective. Stratospheric measurements of sufficient quality to address some of the questions related to the role of CH₄ will be available from research satellites (MIPAS, TES, HIRDLS).

Aerosol Optical Properties

Aerosols play a role in all four atmospheric chemistry issues. For air quality, the effects of anthropogenic particulate matter smaller than 2.5 micrometre (PM_{2.5}) on human health is well documented and beginning to be regulated in many countries. For climate, the IPCC assessment of 2002 has recognised that aerosol forcing of the heat and energy balance of the atmosphere is as important as greenhouse gases and highly uncertain. In particular, the scattering of solar radiation by particles and the absorption of radiation by the black carbon component of particles is critical. For tropospheric air quality and oxidising efficiency, the detailed role of aerosols in gas-particle reactions, in cloud chemistry and in precipitation processes is still uncertain. For the stratosphere, it is now well recognised that gas-particle reactions can affect the chemistry controlling the ozone layer.

Unlike atmospheric gases, atmospheric aerosols cannot be characterised by a single concentration. Instead they have physical, chemical and optical properties, as well as their state of mixture. Therefore, a variety of measurement techniques are required. Accuracy requirements for aerosol measurements are generally expressed in different terms for different applications. Target and threshold requirements have been compiled for an ESA Atmospheric Chemistry mission (ACECHEM) for the purpose of studying atmospheric chemistry and climate. The WMO/CEOS report provides further target and threshold requirements for heterogeneous stratospheric chemistry and climate. These requirements are compiled and updated in Table 4.3.

Current State of the Global Measurement System and Integration of Observations

The global measurement system for tropospheric aerosols currently includes a network of ground-based *in situ* aerosol monitoring stations operated by WMO members under GAW and national or regional monitoring programmes such as EMEP. It also consists

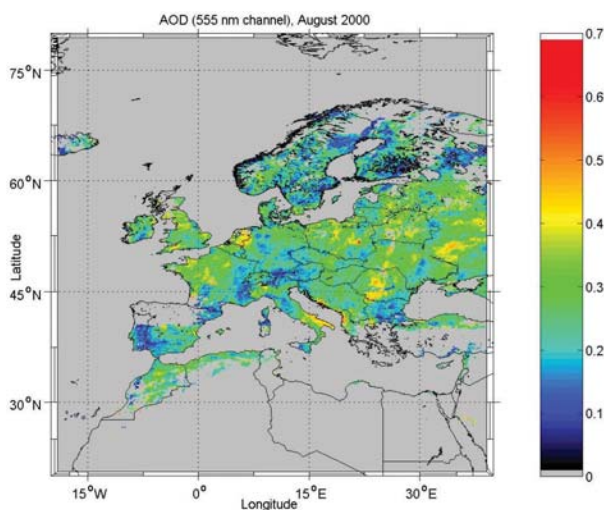


Figure 4.16. Aerosol optical depth over Europe from ATSR-2, average over August 2000. [Courtesy: G. de Leeuw, TNO-FEL, The Netherlands]

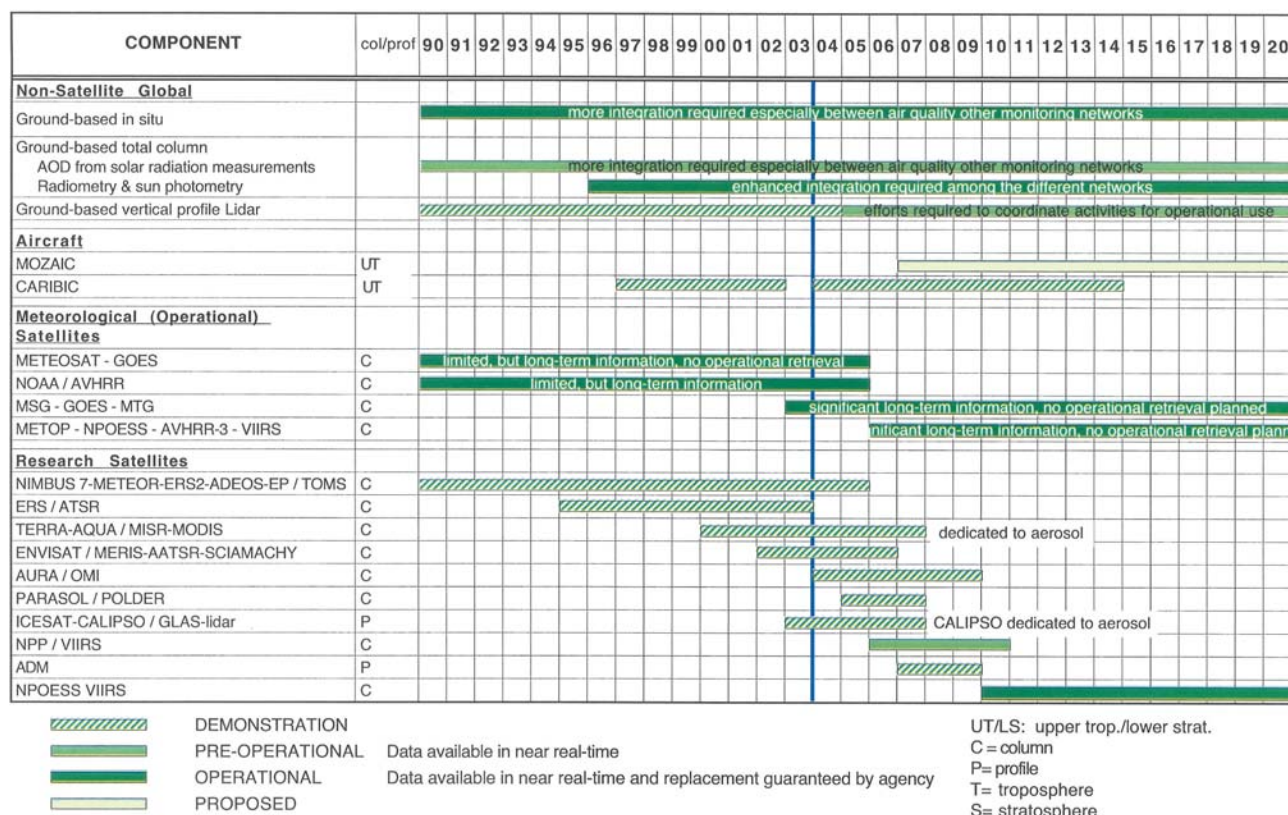


Figure 4.17. An overview of satellite, ground-based and aircraft measurements for tropospheric aerosol optical properties.

of a ground-based network of aerosol optical-depth measurement stations operated by the GAW community, the Baseline Surface Radiation Network (BSRN) of the World Climate Research Programme, NASA/AERONET, PHOTONS, AEROCAN, DOE/ARM, GMA and SKYNET. This network is currently not well coordinated, but efforts are being made by GAW to remedy this.

In addition, there are ground-based aerosol LIDAR networks operated by several regional programmes including the European EARLINET, the NASA MPLS, NDSC and members of GAW. They are extremely important in providing missing information on the vertical distribution of aerosols, as well as on the aerosol mixing depth in the atmospheric boundary layer. Globally, these types of observations are largely uncoordinated and lack a solid foundation for long-term operation.

Routine aircraft measurements of aerosol optical properties are still in a demonstration mode with no substantial geographical coverage. Systematic observations of aerosol radiative properties in the lower 3 km of the troposphere have begun under a pilot study conducted by DOE/ARM in Oklahoma in cooperation with NOAA/CMDL. Twice weekly, an instrument package is flown by a light aircraft. Such observation are

important in order to parameterise the remote-sensing observations of aerosols. They need to be extended to a number of other locations around the World.

Satellite observations of aerosol optical properties have progressed to a point where they range from pre-operational to operational, although there are demonstration-mode instruments on a number of research satellites (see Fig. 4.17). These carry an unprecedented array of spaceborne sensors with unique characteristics enabling quantitative observations on a global scale. Other instruments such as TOMS provide valuable long-term measurements of aerosols, although with a lower accuracy and resolution. These spaceborne measurements are complemented by the ground-based networks already mentioned. Although occasionally covered on a research-campaign basis, there is an obvious lack of continuous, high-quality *in situ* measurements of aerosols in Africa, South America and Asia. The largest changes in aerosol loading are expected to occur in the coming decades over these continents, and downstream of them; satellites are an obvious way to close the gap.

It is important to note that, as their characteristics improve, the new generation of operational meteorological satellites (geostationary: SEVIRI on MSG;



Figure 4.18. An Overview of satellite, ground-based and aircraft measurements for stratospheric aerosol optical properties.

Sun-synchronous: AVHRR-3 on METOP and VIIRS on NPOESS) will provide some basic column properties of aerosols (aerosol optical depth, Ångström exponent, ratio of coarse to fine mode). However, aerosol is of such importance that the routine measurements of its optical properties need to be extended to include other properties such as physical size distributions, the UV radiative properties and vertical profiles. The succession of several spaceborne lidars (GLAS, CALIPSO), following the LITE Space Shuttle experiment, should, in the future, complement the monitoring of aerosol vertical profiles in both the troposphere and stratosphere by ground based LIDARS and aircraft. The ground-based networks need to continue to provide ground truth, retrieving parameters not measurable from space, and filling the gaps in spaceborne monitoring. The networks also need to be enhanced in some regions of the World.

The measurement system for stratospheric aerosols currently includes the ground-based measurement networks mentioned above with NDSC, AERONET and GAW playing an important role. A number of satellite instruments, e.g. SAGE-III, POAM, HALOE, MIPAS, and TOMS, are in place. These measurements include valuable long-term measurements, such as those of the limb-viewing SAM/SAGE instrument series from 1978 onwards, as well as highly specialised satellite instruments like POAM and HALOE, which are capable of detecting polar stratospheric clouds (PSCs) and identifying the type of these clouds (e.g. ice clouds or nitric acid containing clouds). Concerning the Arctic and Antarctic PSC issue, uninterrupted monitoring will be required to fulfil the requirements of the Montreal Protocol for the protection of the ozone layer. Given that the sulphate budget of the upper troposphere and lower

stratosphere is not well understood, a coherent long-term record of the stratospheric aerosol will be essential. With SAGE-III the immediate future is well covered. The enhanced capabilities of satellite instruments for investigating the processes in the tropical tropopause layer will be required in future, including the understanding of deep convective clouds and thin laminar cirrus clouds, and their role in dehydrating air on its way from the tropical upper tropopause into the lower stratosphere. Here lidars, both ground-based and spaceborne, will become particularly useful. The active sensing of the NASA GLAS altimetry instrument has managed to shed some light on aerosol vertical distributions. The LIDAR instrument on NASA CALIPSO is designed specifically to provide vertical profiles.

Gaps and Overlaps in Aerosol Measurements

Although major national and international efforts are underway to characterise the chemical and physical properties of aerosols, there are major gaps that urgently need to be filled if an IGACO system is to be put into place. The biggest gaps are: (i) in coordination of a large number of existing monitoring efforts, (ii) the lack of a data synthesis and analysis centre and (iii) measurements of aerosol chemical composition and the state of internal or external mixing of aerosols. The last can only be filled by ground-based, aircraft and balloon *in situ* measurements. They are needed to reduce the uncertainty in indirect aerosol climate forcing through their interactions with clouds. No global network currently exists but intensive efforts toward establishing one are under way by WMO/GAW.

While it is interesting and economical to obtain aerosol properties as byproducts of meteorological satellite

measurements (e.g. NPOESS, METOP, ADM), there is a risk that planning of satellite missions will not meet the actual need for continuous aerosol measurements. Although they represent a source of reliable and accurate worldwide aerosol products (some of which are not measurable from space), current networks of ground-based Sun photometry rely on uncertain sources of funding, especially in the long-term. Finally it is clear that the integration of a large spectrum of aerosol properties (measured and needed) requires techniques such as sophisticated modelling and data assimilation.

Critical Ancillary Parameters

Modelling will require a number of ancillary parameters to characterise the physical state of the atmosphere where the detailed measurements of concentration are being made. Most important among these are temperature, pressure, wind speed (u , v , w), cloud-top height, cloud coverage, albedo and solar radiation, all measured from the satellite or ground from which the chemical parameters are being estimated.

In addition, satellite measurements of lightning flash frequency, the number of fires and surface properties, such as vegetation index, are necessary for modelling and studies of effects. It is expected that these ancillary measurements will be provided by other satellite systems. There are a number of national ground networks and satellite-based instruments for lightning detection and efforts are being made to establish a database. The coverage, however, is patchy at present.

4.3 Accuracy Requirements

The need for long-term continuity in quality-assured atmospheric chemistry observations has already long been emphasised by the global-change research community. Within the context of IGOS, WMO-CEOS Report No. 140 identified the current deficiencies in the observing system and possible courses of remedial action; it suggested priorities to try to ensure the most effective use of available resources, and for upgrading existing or establishing new systems, in order to provide user-friendly access to the data products called for by the global-change community.

A major concern for IGACO is the continuation, consolidation and harmonisation of calibration and validation support for the observations, as these are critical to ensure their scientific value. In considering quality-assurance requirements for global atmospheric chemistry measurements, it is important to be specific as the requirements vary appreciably with the issues of concern (air quality, oxidation efficiency, chemistry-climate interaction and stratospheric ozone depletion) and also the altitude regime.

Tables 4.2a and 4.2b list the data-quality objectives as "target" and "threshold" values for trueness and precision. They are based on the WMO-CEOS Report No. 140 and the recent SPARC assessments. The set of requirements satisfy the user communities concerned with the focus of IGACO (air quality, oxidation efficiency, chemistry-climate and stratospheric ozone depletion, as well as the essential "end-to-end" ground-truthing for satellite sensors). They are based on "quantitative science", i.e. on measured concentrations, on published trend assessments, and on known concentration differences in the vertical and horizontal distribution of the various parameters. The development of data-quality objectives is an iterative and continuously evolving process, which takes into account user needs and advances in measurement technologies/capabilities. They ultimately lead to the dissemination of data products to the scientific community and policy makers that are of known quality and adequate for their intended use.

The tables summarise the data-quality objectives for the chemical species and parameters chosen. They list values for column amounts (troposphere and total atmosphere) and vertical profiles using the altitude regions (1) lower troposphere 0-5 km, (2) upper troposphere 5 km to tropopause, (3) tropopause to 30 km, and (4) upper stratosphere and mesosphere > 30 km. These values form the basis for deriving globally harmonised data-quality objectives for an integrated observing system. Table 4.3 provides similar data quality objectives for some aerosol parameters.

ATMOSPHERIC SPECIES IN GROUP 1 TO BE MEASURED BY AN INTEGRATED GLOBAL OBSERVING SYSTEM												
Atmospheric Region	Requirement	Unit	H ₂ O	O ₃	CH ₄	CO ₂	CO	NO ₂	BrO	ClO	HCl	CFC-12
1. Lower troposphere	O _x	km	5/25	<5/50	10/50	10/500	10/250	0/250	50			
	O _z	km	0.1/1	0.5/2	2/3	0.5/2	0.5/2	0.5/3	2			
	O _t		1hr	1hr	2hr	2hr	2hr	1hr	1hr			10d
	precision	%	1/10	3/20	1/5	0.2/1	1/20	10/30	10			2*
	trueness	%	2/15	5/20	2/10	1/2	2/25	15/40	15			4*
	delay		(1)/(2)	(1)/(2)	(1)/(2)	(1)/(2)	(1)/(2)	(1)	(2)			
2. Upper troposphere	O _x	km	20/100	10/100	50/250	50/500	10/250	30/250				
	O _z	km	0.5/2	0.5/2	2/4	1/2	1/4	0.5/3				
	O _t		1hr	1hr	2hr	2hr	2hr	1hr				
	precision	%	2/20	3/20	1/10	0.5/2	1/20	10/30				
	trueness	%	2/20	5/30	2/20	1/2	2/25	15/40				
	delay		(1)/(2)	(1)/(2)	(1)/(2)	(1)/(2)	(1)/(2)	(1)				
3. Lower stratosphere	O _x	km	50/200	50/100	50/250	250/500	50/250	30/250	100	100	50/250	1000
	O _z	km	1/3	0.5/3	2/4	1/4	2/5	1/4	1	1	1/4	
	O _t		1d	1d	6-12hr	1d	1d	6-12hr	6hr	6hr	6-12hr	10d
	precision	%	5/20	3/15	2/20	1/2	5/15	10/30	10	10	5/10	6
	trueness	%	5/20	5/20	5/30	1/2	10/25	15/40	15	15	15	15
	delay		(1)/(2)	(1)/(2)	(1)/(2)	(2)/(3)	(2)/(3)	(1)	(2)	(2)		
4. Upper stratosphere, mesosphere	O _x	km	50/200	50/200	50/250	250/500	100/500	30/250	100	100	50/250	
	O _z	km	2/5	0.5/3	2/4	2/4	3/10	1/4	1	1	1/4	
	O _t		1d	1d	1d	1d	1d	1d	1d	1d	1d	
	precision	%	5/20	3/15	2/4	1/2	10/20	10/30	10	10	5/10	
	trueness	%	5/20	5/20	5/30	1/2	10/25	15/40	20	20	15	
	delay		(1)/(2)	(1)/(2)	(1)/(2)	(2)/(3)	(2)/(3)	((1)/(2)	(2)	(2)		
5. Total column	O _x	km	50/200	10/50	10/250	50/500	10/250	30/250	100	100	30/250	1000
	O _t		1d	1d	12hr	1d	1d	12hr	12hr	12hr	6-12	10d
	precision	%	0.5/2	1/5	1/5	0.5/1	1/10	1/10	10	10	4	4
	trueness	%	1/3	2/5	2/10	1/2	2/20	2/20	15	15	6	10
	delay		(1)/(2)	(1)/(2)	(1)/(2)	(2)/(3)	(1)/(2)	(1)	(2)			
6. Tropospheric column	O _x	km	10/200	10/50	10/50	10/500	10/250	10/250	25			1000
	O _t		1hr	1hr	2hr	2hr	2hr	1hr	1hr			10d
	precision	%	0.5/2	5/15	1/5	0.5/1	2/20	1/10				4
	trueness	%	1/3	5/15	2/10	1/2	5/25	2/10				10
	delay		(1)/(2)	(1)/(2)	(1)/(2)	(1)/(2)	(1)/(2)	(1)				

Table 4.2a.

Tables 4.2a and 4.2b. Observational requirements for gaseous atmospheric species and radiation levels. For each altitude range, the target (best case) and threshold (minimum to be useful) values are given as target/threshold. Otherwise the value is given as an overall objective. The requirements are given in terms of horizontal resolution (Δx , km), vertical resolution (Δz , km), temporal resolution (Δt), precision (random error, %) and trueness (total error, %). Delay time between observation and availability of the product: (1) HOURS for operational use in chemical weather forecast, air quality and oxidation efficiency; (2) DAYS to WEEKS for global distributions, ozone depletion, trend analysis and verification of international agreements; (3) MONTHS for climate research and modelling. * refers to surface measurements. ** refers to in situ, or from irradiation measurements with radiative transport calculations under cloud-free conditions. Group 1 and Group 2 attributes are defined in Section 5.3.

ATMOSPHERIC SPECIES IN GROUP 2 TO BE MEASURED BY AN INTEGRATED GLOBAL OBSERVING SYSTEM												
Atmospheric Region	Requirement	Unit	NO	HNO ₃	C ₂ H ₆	CH ₃ Br	Halons	HCFC-22	ClONO ₂	HCHO	SO ₂	UVA <i>j</i> (NO ₂) UVB <i>j</i> (O ¹ D)
1. Lower troposphere	O _x	km	10/250	10/250	50	500*				1	1	
	O _z	km	0.5/3	1/3	?					2-5	2-5	2-5
	O _t		1hr	1d	1hr	10d	10d	10d		1hr	1hr	1hr
	precision	%	10/30	10/30	10	4*	15*	2*		10	5	7/10*
	trueness	%	15/40	15/40	15	8*	20*	4*		15	10	15*
	delay		(1)	(1)/(2)						(1)	(1)	
2. Upper troposphere	O _x	km	30/250	10/250	50					10	10	50/500
	O _z	km	0.5/3	1/3	2					0.5	0.5	3**
	O _t		1hr	1d	1hr						1hr	1hr
	precision	%	10/30	10/30	10					10	5	10
	trueness	%	15/40	15/40	15					15	10	15
	delay		(1)	(1)/(2)						(1)	(1)	
3. Lower stratosphere	O _x	km	30/250	50/250		500	500	1000	50/250			
	O _z	km	1/4	1/4		5	5	5	1/4			
	O _t		12hr	12hr		3d	3d	3d	6-12hr			
	precision	%	10/30	10/30		4	4	8	20			
	trueness	%	15/40	15/40		8	8	15	30			
	delay		(1)	(1)/(2)								
4. Upper stratosphere, mesosphere	O _x	km	30/250	50/250					50/250			
	O _z	km	1/4	1/4					2/6			
	O _t		1d	1d					1d			
	precision	%	10/30	10/30					20			
	trueness	%	15/40	15/40					30			
	delay		(1)/(2)	(2)/(3)								
5. Total column	O _x	km	30/250	30/250	50			1000	30/250		50	
	O _t		1d	1d	1hr			10d	6-12hr		1hr	
	precision	%	1/10	1/10	1			5	20		1	
	trueness	%	2/20	2/20	2			15	30		2	
	delay		(1)	(2)/(3)							(2)	
6. Tropospheric column	O _x	km	10/250	10/250		1000	1000	1000				
	O _t		1hr	1d		10d	10d	10d				
	precision	%	1/10	1/10		4	4	6				
	trueness	%	2/20	2/20		8	8	15				
	delay		(1)	(1)/(2)								

Table 4.2b

Theme		Unit	Aerosol Optical Depth (VIS+IR)	Aerosol Extinction Coefficient (VIS)	Aerosol Absorption Optical Depth (VIS)	PM1, PM2.5, PM10
a, d Climate Studies and oxidising capacity	O _x	km	1/10	10/100	1/10	
	O _z	km		0.5/1		
	O _t		global daily	global weekly	global daily	
	precision		0.005/0.01	0.005/0.01 km ⁻¹	0.002/0.01	
	trueness		0.01/0.02	0.01/0.02 km ⁻¹	0.004/0.02	
	delay		weeks	weeks	weeks	
b Air quality (PBL and free trop)	O _x	km	0.25/1	0.5/2		0.25/1
	O _z	km		0.1 in PBL		0.1 in PBL
	O _t		regional hourly	regional daily		regional sub-daily
	precision		0.005/0.01	0.005/0.01 km ⁻¹		1/10 µg m ⁻³
	trueness		0.01/0.02	0.01/0.02 km ⁻²		1/10 µg m ⁻³
	delay		near real-time	near real-time		near real-time
c Ozone depletion (UT/LS)	O _x	km	10/100	10/100		
	O _z	km		1/2		
	O _t		10d	10d		
	precision		10 ⁻⁵ /10 ⁻⁴	10 ⁻⁶ /10 ⁻⁵ km ⁻¹		
	trueness		10 ⁻⁵ /10 ⁻⁴	10 ⁻⁶ /10 ⁻⁵ km ⁻¹		
	delay		days	days		

Table 4.3. Target and threshold requirements for aerosol (mostly optical) properties. Note that these quantities are not independent (for instance aerosol extinction coefficient and sometimes aerosol optical depth can serve as a proxy for the concentration of particulate matter [PM] at the surface). Here precision and trueness are given in absolute values. Note that "Aerosol Optical Depth" and "Extinction Coefficient" are not independent quantities, but are specified here separately to allow for column and profile information, respectively.

In this chapter, IGACO is described in detail and recommendations are made for its implementation. It draws not only upon the experience gained for meteorological variables, but also takes into account differences in communities and technology required for atmospheric composition measurements. The recommendations are divided into two types: general recommendations (GR) related to the general features of IGACO, and more specific recommendations (SR) related to the implementation.

5.1 A Global System is Required

GR1 Establishment: An Integrated Global Atmospheric Chemistry Observation System (IGACO) should be established for a target list of atmospheric chemistry variables and ancillary meteorological data.

The Integrated Global Atmosphere Chemistry Observation System (IGACO) proposed in this report consists of three measurement components (ground-based, aircraft and satellite), and a modelling component to integrate the observations into a global picture of atmospheric composition and its trends. In addition, there are the crucial additional elements of quality-assurance, data distribution and data archiving/analysis. The proposed system is summarised in Section 5.2.

The proposed system is similar to that used by the meteorological community, which has a long history: the meteorological system was developed by the national hydrological and meteorological services and space agencies, coordinated by the WMO, to integrate measurements of meteorological variables important to weather prediction and climate. Today a system of non-satellite networks and satellites delivers observations of physical variables such as temperature, pressure, wind speed and humidity in real time through a Global Telecommunications System (GTS) to forecast modelling centres around the World. It also delivers a more comprehensive data set consisting of real-time and non-real-time observations to a data archive, where it is accessible for re-analysis using models at the NCEP, ECMWF and JMA. Using sophisticated mathematical descriptions of atmospheric processes and the most powerful computers available, these models serve to integrate the observations from a variety of sources into a comprehensive picture of current and projected global weather patterns.

Establishment of an integrated observational system for chemical variables lags, by several decades, the observational system for physical variables. Now, early in the 21st century, all the components are either in place or, with careful planning, will be readily available

at reasonable cost within the next 10 years. What is missing is a well-focused strategic plan and an internationally coordinated implementation effort, which it is the purpose of this report to provide.

The authors and the reviewers of this document agree that the establishment of IGACO to observe the composition of the atmosphere and its changes is necessary, timely, and feasible. It will be necessary to start planning immediately for a necessary minimum of satellites, ground stations and routine aircraft programmes, with the required modelling support, to produce IGACO.

GR2 Continuity: the data products from satellite and non-satellite instruments, which are to be integrated into a global picture by IGACO, must have assured long-term continuity.

In order to quantify the changing atmospheric composition, an overarching and fundamental requirement for IGACO is that continuity of measurement is maintained for all the system components.

GR3 Management of IGACO. The responsibility for the co-ordination and implementation of IGACO should rest with a single international body. International and national agencies responsible for aspects of IGACO should be committed partners and agree on their appropriate responsibilities.

It is not yet clear where the responsibility for the development of IGACO in its entirety should lie. The current international meteorological, environmental and space agencies all have different mandates with respect to the generation of the data needed, and no one body is responsible for observing the changing atmosphere. This is a critical management issue to be dealt with in the implementation phase of IGACO, which is discussed further in Section 5.3.

5.2 The Proposed Architecture for IGACO

The proposed IGACO system is shown as a flow chart in Fig. 5.1. It should be emphasised that, although various components and elements of the IGACO system are presently available or projected, a complete system does not yet exist for any atmospheric constituent in the target list of variables (Tables 4.2 and 4.3).

Essential to the proper functioning of IGACO is a system for data collection from various sources, a system for distribution of the data to users and of archiving these data for establishing long-term records, as well as an end-to-end quality-assurance and quality control system to quantify the uncertainties in the data.

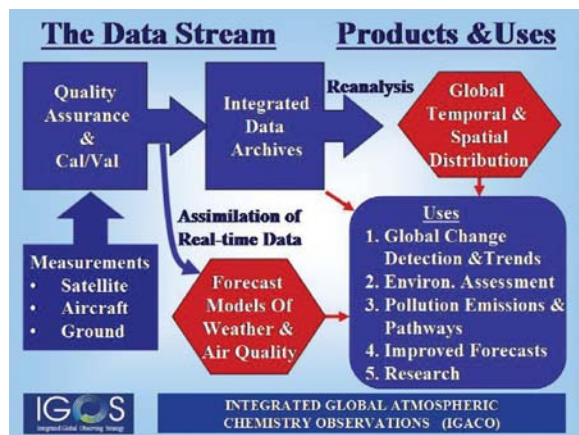


Figure 5.1. The major components and critical elements of the IGACO system. [Figure courtesy of L. Barrie, WMO]

5.2.1 Measurements

GR4 *Gaps in observational coverage: for each target species and variable, the present gaps in the current spatial and temporal coverage should be filled by extending the existing measurement systems.*

The proposed measurement system comprises an optimised network of ground-based stations, routine aircraft flights and satellites in a configuration suitable for addressing the temporal and spatial variability of a specified chemical variable. Since observations are made with a variety of instruments, which vary in their spatial and temporal resolutions, an efficient mathematical approach to assimilating them into a global picture is necessary to define an optimal mix of each instrument type. Currently, there are major gaps in the global coverage of observations for all the target chemicals.

5.2.2 Quality-assurance and Data-handling protocols

GR5 *Long-term validation of satellite observations: in order to ensure accuracy and consistency for satellite measurements, sustained quality-assurance measures, over the entire lifetime of the observations, are essential.*

There are presently large gaps in the quantitative information for each chemical parameter about the comparability of observations by different instruments, which limits our ability to merge observations into a global data set. Quality assurance is the only effective way to ensure the comparability of data. Three elements of quality assurance are essential in the measurement/analysis/assessment hierarchy:

- **Data Quality Objectives (DQO):** defined by trueness, precision, completeness, comparability and representativeness.
- **Quality Control (QC):** achieved through calibration/validation and traceability to globally harmonised standards and reference methods, instrument and method intercomparison, good laboratory practice, etc.
- **Quality Assessment (QA):** achieved through independent verification (e.g. system and performance audits for ground-based stations).

For most *ground-based* and *routine aircraft* observations of the gaseous species, the quality assurance of measurements is already achievable using internationally traceable standard reference materials or reference methods, routine inter-laboratory comparisons, instrument calibration programmes and data analysis aimed at ensuring that the quality-assurance criteria are being met. However, in practice, the effectiveness of delivery of quality assurance in a measurement programme is patchy at best. It is limited by the resources needed for key quality-assurance facilities and functions.

For *satellite-borne sensors* the quality-assurance commonly referred to as calibration/validation is more complicated. It involves the evaluation of the retrieval algorithms used to extract geophysical quantities from calibrated and well-characterised basic data products. Calibration/validation is being addressed by the Working Group on Calibration and Validation of the CEOS subgroup on atmospheric composition. A policy for validating data for satellite ozone and chemistry missions was suggested in WMO/CEOS Report No. 140.

The components in the end-to-end calibration/validation of satellite data are:

1. **Pre-launch calibration.** The purpose of calibration is to assign the correct physical quantities to instrument output. This encompasses a precise characterisation of the radiometric instrument response, including conversion of the measured signal to atmospheric radiances, as well as the accurate determination of the spectral domain and resolution, the viewing direction and, in some cases, the polarisation sensitivity. The uncertainties of these parameters and their sensitivity to environmental conditions needs to be established. For a large class of instruments, the need to carry out calibration under vacuum conditions encountered in space constitutes an important requirement. Calibration requires standards and measurements provided by the recognised national standards institutions to be known and traceable.

2. **In-flight calibration** is needed to quantify changes in the instrument induced by launch vibrations, outgassing upon deployment in space, thermal variations in orbit, ageing and radiation damage to electrical and optical components. It should include radiometric calibration using internal sources, solar radiation or deep-space measurements, spectral calibration with internal sources or solar or terrestrial spectroscopic measurements, pointing calibration, using well-defined terrestrial or celestial references. Usually, special calibration modes are required, which form part of the instrument design and the operational in-orbit scenario.

It must be recognised that both in-flight and on-ground calibration will have inherent limitations. For example, on-ground calibration in vacuum will put limitations on the range and type of measurements that can be carried out, and terrestrial sources available will not always be able to simulate the instrument input prevalent in space. On the other hand, in space independent radiometric and spectral sources are not always available (for example, a source of known polarisation state). Therefore, calibration is often a compromise between what is needed ideally and what is achievable in practice.

3. **Retrieval algorithm consistency.** Multiple satellite systems will be in orbit measuring the same constituent (e.g. O₃ from NPOESS and METOP). Every effort should be made to have a common or well-compared radiative transfer model and the same spectroscopic databases to ensure that these fundamental parameters do not cause biases. The respective algorithms for each system have a different heritage and physical basis. It is highly desirable to have a uniform data set among the systems so as to avoid biases between them. Comparisons of the algorithms using a common radiance data set should be conducted as part of the cal/val programme or even sooner.
4. **Geophysical validation.** Trace gas or aerosol data retrieved from spaceborne sensors are validated by comparison to the most reliable correlative data. Simultaneity and co-location of the measurements must be optimised, and any remaining sampling differences must be bridged using, for example, CTMs. Ideally, validation sources should be based on a different measurement principle or different spectroscopic features; they include ground-based networks and airborne measurements, sometimes complemented by chemical models to get access to additional species. Profile data covering the stratosphere and upper troposphere can often only be validated by scientific instruments operated on a campaign basis, e.g. on

stratospheric balloons and aircraft. Intense geophysical validation campaigns are needed in the early phase of a space mission, followed by periodic short measurement campaigns and continuous use of routine observation networks, to monitor instrument performance throughout its lifetime.

5. **Inter-satellite comparison.** Observations of old and new satellite instruments require inter-comparison in order to establish consistent long-term datasets, despite possible discrepancies between absolute values of data.

Apart from their necessary role within IGACO, ground-based networks are essential components in the validation of satellite observations.

GR6 *Validation of vertical-profile data from satellite observations: a set of high performance scientific instruments using ground, aircraft and balloon platforms, possibly operated on campaign basis, must be maintained to provide the crucial validation data.*

Validation of satellite data also requires a component outside the long-term continuous observation system addressed in this report:

GR7 *Comparability: the ability to merge observations of different types must be ensured by insisting that appropriate routine calibration and comparison activities linking diverse measurements together are part of an individual instrument measurement programme.*

In order for the full power of diverse observations of any parameter to be fully realised, the quality and comparability of a measurement require that the data quality objectives are routinely met, and that they be available with the data. This is an essential requirement of IGACO.

GR8 *Distribution of data: universally recognised distribution protocols for exchange of data on atmospheric chemical constituents should be established.*

The data and associated quality-assurance information need to be in such a format that they are easily accessible in real time (for assimilation in forecast models) or on time scales of months to a year for integration into a comprehensive data archive using model integration techniques. Data distribution protocols are needed that allow for effective transmission of observations together with their associated quality-assurance information to an

established archive and to forecast modellers. The Global Telecommunications System (GTS) of WMO is an example of such a system for real-time meteorological data.

5.2.3 World Integrated Data Archive Centres (WIDAC)

GR9 Multi-stake holder World Integrated Data Archive Centres (WIDAC) should be established so for the targeted chemical variables.

GR10 Storage for raw data should be established so that they can be re-interpreted as models and understanding improves.

In the IGACO system, observations should be made available through a World Integrated Data Archive Centre (WIDAC) for future applications and re-analysis (see Fig. 5.1). The need for re-analysis will recur as more sophisticated tools and improved observational data become available. A WIDAC must be sufficiently robust that it can last for many years.

The optimal configuration of the WIDAC will have to be decided, but it may be centralised or distributed. The meteorological community has such an archive for a short list of meteorological parameters, but is facing major challenges to extend it to other parameters such as water vapour. Furthermore, despite the maturity of meteorological data coordination, there is still no WIDAC for meteorological data that has free access.

There is currently no potential WIDAC for any chemical constituent on the target list. World Data Centres do exist for individual network or satellite observations for particular variables on the target list, but there is no integration at present. For instance, GAW has a World Ozone and UV Data Centre for non-satellite observations and many organisations have surface ozone observations. The satellite agencies have ozone data centres for observations from specific satellite instruments, and there have been releases of quality-controlled data with suitable software tools to manipulate them; these include updates as funding allows. To bring all this information together in a synergistic way is the challenge that IGACO intends to meet.

Stakeholders in a WIDAC are: (i) providers of measurement data, (ii) data analysis and model groups, and (iii) users of the integrated data set. A WIDAC should have periodic updates of observations and reanalysis built into it, so as to cope with the continuous improvement in satellite retrieval algorithms and revisions of non-satellite observations. Access to data should be free of charge and expedited by software tools that are user-friendly.

5.2.4 Models and model inputs needed for the IGACO system

GR11 The development of comprehensive chemical modules in weather and climate models should be an integral part of IGACO.

As the preceding sections illustrate, atmospheric models that encompass the target constituents are needed for use in forecasting air quality and for re-analysis of the global comprehensive database in the WIDAC to produce a 4-D composition distribution.

Data assimilation tools have started to provide consistent distributions of stratospheric ozone and water vapour on a routine basis. They now need to be improved and extended to include other species on the IGACO target list. The assimilation of all of these species into the weather and climate models which include a full chemical scheme will allow for more confident predictions of the distributions of many other compounds not on the target list.

GR12 Strong coordination with meteorological services is essential so that the ancillary meteorological data required by IGACO is accessible.

The requirements for numerical weather prediction have been mandated to and are addressed by the national meteorological services represented by the WMO. The essential ancillary data for IGACO listed in Chapter 4 is, in principle, available from them. However, some coordination is needed to ensure that the data is in fact freely available.

5.3 Implementation

The implementation of the IGACO system will necessarily take time, particularly, as is emphasised by the timelines in Chapter 4, there are gaps looming in the satellite coverage which characteristically take at least 10 years to fill. Taking a hierarchical approach, there are two main phases of implementation of the IGACO system: the short term from 0 to 10 years and long term beyond 10 years. In addition, there are sub-phases within these that depend on the specific activity required. These are addressed below after the specific recommendations (labelled SR).

The target chemical species, listed in Chapter 4, have been divided into two groups depending on the current state of the IGACO components.

Group 1: O₃, H₂O, CO₂, CO, NO₂, BrO, ClO, HCl, N₂O, the CFCs, ClONO₂ and aerosol optical properties.

There is likely to be a reasonably comprehensive set of global observations of these species for both the troposphere and the stratosphere using a sparse number of LEO satellites, ground-based networks and flight measurements. There are also atmospheric modelling capabilities to begin to form them into a global picture. In addition, there is a good network of ground-based and satellite observations that only require maintenance and some gaps to be filled. Data assimilation modelling is in good shape. Routine aircraft observations are made, but are not yet comprehensive enough.

Group 2: CH₄, HCHO, volatile organic compounds (VOC), SO₂, HNO₃, OCIO, NO, CH₃Br, and the halons, together with *j*(NO₂) and *j*(O¹D).

For this group, all the current satellites are in experimental "demonstration" mode and only have a limited lifetime. There are some ground-based *in situ* measurements. With the exception of CH₄, the global network is sparse. The next ten years need to be spent developing instrumentation and putting the monitoring infrastructure in place.

5.3.1 The short term: 0 to 10 years

The aim here is maintain and improve the quality of existing capabilities, demonstrate the workability of IGACO for selected species, develop parts which are not yet ready and plan the deployment of the remainder. Specific recommendations discussed in detail below will result in the:

- establishment of an IGACO system for all *Group 1* variables;
- development of data harmonisation and QA/QC for all aspects of the initial system;
- upgrading of selected ground-based stations and routine aircraft platforms;
- development of automated algorithms necessary for satellite measurements in the troposphere for as many parameters as possible;
- development of modelling and assimilation tools, parameterisations and strategies to produce a reliable overall picture of the atmospheric composition from the various measurement components; and
- planning and establishing initiation of a sustainable network for *Group 2* variables that is available for implementation of an IGACO system in the long term, Phase 2.

The specific actions required in this first phase of IGACO are listed below together with a sub-phase scheduling chart indicating the timing of implementation.

SR 1 Establishment of an IGACO system for selected Group 1 species encompassing data collection, harmonisation, QA/QC, data archiving and model-based integration.

Aerosol optical properties, stratospheric and tropospheric O₃ and water vapour are ripe for demonstration projects in this regard since many components already exist. For the remaining *Group 1* species, observational system gaps are considerably greater and recommendation SR4 should be implemented first.

SR2 Initiate immediately the planning and implementation of a network of satellite platforms to be launched in the long term, with consideration of geostationary as well as enhanced low-Earth-orbit capabilities.

To address climate-chemistry interaction in the UT/LS and stratospheric ozone depletion, high-vertical-resolution profiles are obtained, using the limb-sounding technique which requires a low Earth-orbit. Two satellites are sufficient to achieve 12-hour time sampling.

For air quality, oxidising efficiency and climate, observations of the troposphere down to the surface are made. This is achieved in nadir viewing geometry which provides total-column information or low-resolution profiles. These measurements are required with much better temporal and horizontal resolution than any existing atmospheric chemistry satellite data. In the long term, three to four geostationary satellites or, alternatively, a number of polar-orbiting satellites will be needed. The trade-off between the two options is determined by the performance with respect to temporal resolution, geographical coverage, horizontal resolution, signal-to-noise ratio, pointing stability, and necessarily by technical and cost considerations.

SR3 Develop the necessary data-harmonisation, quality/control and data-exchange protocols using the demonstration projects in SR1.

SR4 Upgrade the missing ground-based (in situ, total column, active and passive profiling, and balloon sonde) measurements to measure the Group 1 variables, and, where feasible, some of those from Group 2.

SR5 Develop a sustainable routine aircraft measurement programme based on the considerable experience obtained in ongoing projects.

Most of the existing aircraft programmes are operated as short-term research projects and have no secure

SPECIFIC RECOMMENDATIONS	YEARS AFTER INITIATION OF IGACO									
	1	2	3	4	5	6	7	8	9	10
SR1a: Establish IGACO System for Ozone, Aerosol & Water Vapour										
SR1b: Establish IGACO Systems for Remaining Group 1 Variables										
SR2: Plan and Initiate New Satellite Missions for the Long Term										
SR3: Develop Data Management/Analysis Systems Including WIDACS										
SR4: Upgrade ground-based observations for Group 1										
SR4: Upgrade ground-based observations for Group 2										
SR5a: Begin Instrument Development Programme for IGACO Aircraft Activities										
SR5b: Add New Networks to MOSAIC and CARIBIC										
SR6: Improve Available Satellite Data Retrieval										
SR7a: Incorporate Models in IGACO for SR1a										
SR7b: Incorporate Models for IGACO for SR1b										

Figure 5.2. Scheduling of the sub-phases of IGACO for phase 1: the short term

future. In order to develop the required capacity for a global routine aircraft programme, it is necessary to expand the fleet of aircraft appreciably, so as to provide global coverage; measurements are particularly needed for the Pacific and the southern hemisphere. This requires the development and the certification of smaller instrumentation packages under aeronautical rules, both for implementation on civil aircraft and for the maintenance in an operational system. The current suite of measurements should be extended as much as possible, in particularly for NO_x (NO and NO_2), aerosol and H_2O in the lower stratosphere.

- SR6 *Develop the necessary algorithms and associated calibration/validation procedures to retrieve operationally, total-column and vertical-profile concentrations from existing and planned satellites for as many of the Group 1 and 2 variables as possible.*
- SR7 *Develop chemical transport modelling and data assimilation so as to accommodate data from the various measurement components.*

5.3.2 The long term: beyond 10 years

As shown by the timelines in Chapter 4, there is a looming gap in the provision of satellite-based instruments for atmospheric-chemistry observations since, by then, the present "demonstration mode" satellites have finished their terms. For IGACO, new operational satellites as well as an enhanced non-satellite measurement network will be required.

Specific long-term requirements are to:

- provide an operational network with sufficient satellites to meet the necessary requirements of the measurement time resolution; immediate action is required from the agencies involved to avoid a time gap in global surveillance;
- provide satellite instruments for measurements of all Group 1 and 2 components;
- upgrade ground-based stations to measure Group 1 and 2 components, and to install new stations at those locations that models show to be particularly sensitive;

- d. secure routine aircraft measurements to implement instrumentation for Group 1 and 2 components, and to add routes to improve global coverage; and
- e. develop further models and assimilation techniques to provide a reliable picture of the atmospheric variability and long-term changes.

The long term should see the implementation of IGACO for both the Group 1 and Group 2 species, completion of the extensions to the ground-based observation system and to the flight measurements, and the launch of the necessary satellites, planned at the beginning of the short-term implementation. If the initial planning is properly and promptly executed, as recommended, then one should see IGACO emerge from initial pilot projects for selected species, using parts of the system in demonstration mode, to a fully grown system with the full complement of satellite, flight- and ground-based measurement systems, backed by quality control, data distribution and modelling with data assimilation for all the chemical components.

5.3.3 Education and capacity building

In order to meet and sustain the needs of IGACO, capacity-building measures will be required throughout all the phases of IGACO. The measures should include the support of graduate and postgraduate research programmes, as well as career development for managers, scientists and engineers working to implement and sustain IGACO. They should aim at facilitating different aspects of the phases of IGACO and should include the support of experimental and instrumental development, together with the further development of data inversion and retrieval theory, numerical retrieval and assimilation techniques. Some of the education and training measures could parallel those sponsored by the WMO for the meteorological community, and the CGMS/WMO Virtual Laboratory for Education and Training. Such activities will support the UNDP Capacity 21 initiative.

5.3.4 The commitment process

IGOS requires that a theme such as IGACO be led by one or more of its partners, and that "unless exceptionally approved, it is expected that one of the four observing programmes GCOS, GOOS, GTOS and GOS/GAW will lead future implementation." Considering that the focus of IGACO is on atmospheric composition, the natural lead is the WMO/GAW programme. Other essential players are the satellite agencies represented by CEOS, the other components of WMO, and the research community in universities and organisations.

Some services that the WMO could offer as the lead partner in IGACO are as follows:

- The WMO would use its proven system for coordinating global atmospheric weather and composition research and measurements to aid enhanced weather, climate and ozone prediction. The WMO already liaises with the various satellite agencies both inside and outside the meteorological services. WMO/GAW can work through the WMO constituent bodies (the Commission of Atmospheric Science (CAS), the Commission of Basic Science (CBS) and the WMO Executive Council) as well as the WMO Consultative Meetings on High-level Policy on Satellite Matters to promote the implementation of IGACO.
- The WMO/GAW programme together with its partners already has strong global ground-based networks for greenhouse gases, UV radiation, ozone, aerosols and reactive gases with global reference standards, quality assurance, technical support and global facilities. These necessary components of IGACO would be maintained and enhanced to fill critical gaps in coverage and in satellite calibration/validation needs.
- The WMO can offer direct coordination with air-quality forecast agencies through the World Weather Research Programme under the Atmospheric and Environment Research programme and the Commission for Atmospheric Science.
- Integration of surface and space-based meteorological observations and the data flow into air-quality forecasting in IGACO could be addressed through the existing mechanisms developed for meteorological variables by the WMO World Weather Watch Programme and the new WMO Space Programme.

The implementation of IGACO might be undertaken in the following way.

- The IGOS partners agree on the lead institution.
- The IGOS partners form an IGACO implementation group consisting of representatives of the major partners together with several scientists prominent in the field. The purpose of the group would be to support the lead institution in implementing IGACO.
- WMO, if appointed as lead, would review and announce its commitment to IGACO, both on the organisational side, and on the measurement side in which its participation is vital.

- The CEOS Strategic Implementation Team (CEOS/SIT) would review the IGACO recommendations and indicate ways in which space agencies could contribute. The CEOS Working Group on Calibration-Validation Team (CEOS/Cal-Val) could review, and include in its work programme, the relevant IGACO implementation recommendations for satellite system observations and associated ground-based observations. The CEOS Working Group on Information Systems and Services (WGISS) could review, and include into its work programme relevant IGACO implementation recommendations as they relate to information exchange. Additionally, WGISS could provide recommendations to the IGACO Implementation Team to ensure consistency with CEOS experience in information systems and services.
- CEOS and WMO should work jointly towards establishing appropriate WIDACs and linking them closely with institutions responsible for quality assessment, harmonisation and re-analysis of chemical observations collected by the individual data centres.
- Collaboration with the IGOS Themes on Carbon Cycle and Water should be initiated to ensure the best use of the data and to avoid duplication of effort.
- The WMO and CEOS should then initiate, together with other national and international agencies, a series of research and development programmes to improve the existing parts of the components of IGACO or to develop the required new components.
- The efforts of CEOS and WMO should ensure that IGACO is seen as part of the long term strategy for integrated observational systems being developed under the Global Earth Observations (GEO) initiative (see the Foreword to this report).

Finally it should be emphasised that with such a framework in place for IGACO, other initiatives, both national and from the research community, will be more effective in contributing towards the development of a global integrated observational system. Once the IGACO Theme Report recommendations are implemented, its overall vision will support proposals for specific research projects funded by national, regional and international research agencies. These projects will help to improve existing components or to develop new components of IGACO that do not currently exist. Collaboration with IGAC (International Global Atmospheric Chemistry) would be beneficial; it would encourage the continued improvement of the understanding of the atmosphere essential to the aims of IGACO.

ACECHEM	Atmospheric Composition Explorer for Chemistry and Climate Interaction; a proposed ESA mission		pollutants in Europe; Monitoring arm of the CLRTAP
ADM-Aeolus	Atmospheric Dynamics Mission	ENVISAT	ESA environmental monitoring satellite
AERONET	Aerosol Robotic Network; managed by NASA, includes AEROCAN and PHOTONS	EOS	Earth Observation System set up by NASA
AOD	Aerosol Optical Depth	EOS-Aura,/Terra	NASA satellites
AEROCAN	Canadian Aerosol Optical Depth Network, Canadian Sun-Photometer Network; a part of AERONET	ERS-2	ESA satellite
AOT40	The sum of the differences between hourly ozone concentration and 40 ppb for each hour when the concentration exceeds 40 ppb during a relevant growing season, e.g. for forest and crops.	ESA	European Space Agency
		EU	European Union
		EUMETSAT	European Organisation for the Exploitation of Meteorological Satellites
ATSR-1, ATSR-2	Along Track Scanning Radiometer: instruments launched on ERS-1 and ERS-2	FAO	Food and Agricultural Organization of the UN
BSRN	Baseline Surface Radiation Network	GACP	Global Aerosol Climatology Project
Capacity 21	UNDP capacity building initiative (http://www.undp.org/capacity21/)	GASP	Global Atmospheric Sampling Program
CAPMoN	Canadian Air and Precipitation and Monitoring Network	GAW	Global Atmospheric Watch
CARIBIC	Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container	GCOS	Global Climate Observing System
CAS/WG	Commission for Atmospheric Sciences (CAS) Working Group on Environmental Pollution and Atmospheric Chemistry; a commission within the WMO	GEO (1)	Geostationary orbit
CCM	Chemistry/Climate Models	GEO (2)	Intergovernmental <i>ad hoc</i> Group on Earth Observations;
CEOS	Committee on Earth Observation Satellites	GEOSS	GEO System of Systems
CEOS/SIT	CEOS Strategic Implementation Team	GHG	Greenhouse Gases
CFC	Chlorofluorocarbon	GOME	Global Ozone Monitoring Experiment: instrument mounted on ERS-2 measuring tropospheric components
CGMS	Coordination Group for Meteorological Satellites	GOME-2	Successor of GOME, to be flown on MetOp
CLRTAP	Convention on the Long Range Transboundary Air Pollution (LRTAP)	GOOS	Global Ocean Observation System
CMDL	Climate Monitoring and Diagnostics Laboratory. NOAA laboratory	GTS	Global Telecommunication System
CTM	Chemical/Transport Models	HALOE	Halogen Occultation Experiment; instrument launched on UARS
DOAS	Differential Optical Absorption Spectroscopy	HIRDLS	High Resolution Dynamics Limb Sounder; instrument aboard the NASA Aura satellite
DQO	Data Quality Objective	ICSU	International Council of Scientific Unions
EANET	Acid Deposition Monitoring Network in East Asia	IGAC	International Global Atmospheric Chemistry
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air	IGACO	Integrated Global Atmospheric Chemistry Observations
		IGBP	International Geosphere Biosphere Programme
		IGOS	Integrated Global Observation Strategy
		IPCC	Intergovernmental Panel on Climate Change
		IPCC-TAR	Intergovernmental Panel on Climate Change third assessment report
		LEO	Low Earth Orbit

ILEAPS	Integrated Land Ecosystem - Atmosphere Processes Study	POPs	Persistent organic compounds; semi-volatile compounds travel slowly through the atmosphere and which are now appearing in hitherto pristine parts of the World. They are generally poisonous.
LRTAP	see CLRTAP	PSC	Polar Stratospheric Clouds
MAXDOAS	Multi Axial DOAS	QA/QC	Quality Assurance/Quality Control
METOP	METOP; a Eumetsat polar-orbiting satellite for operational meteorology to be launched by ESA in 2005	SAGE-III	Stratospheric Aerosol And Gas Experiment: NASA instrument
MIPAS	Michelson Interferometer for Passive Atmospheric Sounding; instrument on ESA satellite, ENVISAT	SBUV	Solar Backscatter Ultraviolet Instrument: NOAA instrument on the NASA satellite, NIMBUS 4
MISR	Multi-angle Imaging Spectroradiometer; instrument on a NASA Terra satellite	SCIAMACHY	Scanning Imaging Absorption Spectrometer for Atmospheric Cartography; instrument launched on ENVISAT
MODIS	Moderate Resolution Imaging Spectroradiometer; instrument aboard NASA Terra and Aqua satellites	SEVIRI	Scanning Enhanced Visible and Infrared Imager
MOPITT	Measurement of pollution in the troposphere; instrument on the NASA Terra satellite	SHADOZ	Southern Hemisphere Additional Ozone Sondes
MOZAIC	Measurement of O ₃ , water vapour, CO and NO _x by Airbus in-service aircraft	SOLAS	Surface Ocean and Lower Atmosphere Study
MSG	Meteosat Second Generation	SPARC	Stratospheric processes and their role in climate
NASA	National Aeronautics and Space Administration	TES	Tropospheric Emission Spectrometer
NDSC	Network for the Detection of Stratospheric Change	TOMS	Total Ozone Mapping Spectrometer; instruments mounted on a number of NASA satellites
NMHC	Non-Methane Hydrocarbons	UARS	Upper Atmosphere Research Satellite; NASA satellite
NOAA	National Oceanic and Atmospheric Administration	UN	United Nations
NOAA/CMDL	NOAA Climate Monitoring and Diagnostic Laboratory	UNDP	United Nations Development Programme
NOXAR	Measurements of nitrogen oxides along air routes; Swiss project using commercial aircraft	UN-ECE	UN Economic Commission for Europe
NPOESS	National Polar-orbiting Operation Environmental Satellite System; a NOAA initiative	UNEP	UN Environment Programme
NPP	NPOESS Preparatory Project	UNFCCC	UN Framework Convention on Climate Change
OMI	Ozone Monitoring Instrument	UT/LS	Upper Troposphere/Lower Stratosphere
PHOTONS	PHOTométrie pour le Traitement Opérationnel de Normalisation Satellitaire; a part of AERONET the European Optical depth network	UV, UV-A, UV-B	Ultra Violet radiation; UV-A: ca 320 to 400 nm; UV-B: ca 280 to 320 nm
PM, PM10, PM2.5, PM1	Particulate Matter; the figures refer to particles with a size in µm less than that stated.	VIIRS	Visible/Infrared Imager/Radiometer Suite
POAM	Polar Ozone and Aerosol Measurement; instrument carried on the French SPOT satellites	VOC	Volatile Organic Compound
POLDER	Polarisation and Directionality of the Earth's Reflectances; French instrument carried on the Japanese ADEOS-1 and ADEOS-2 satellites	WCRP	World Climate Research Programme
		WIDAC	World Integrated Data Archive Centre
		WMO	World Meteorological Organization
		WMO/CAS	Commission on Atmospheric Science
		WMO/CBS	WMO Commission on Basic Science

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Printed by
European Space Agency



ESA SP-1282
GAW No. 159 (WMO TD No. 1235)

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