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A Report on Workshops:

General Circulation Model Study of Climate-Chemistry Interaction

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Abstract

This report summarizes the discussion on *General Circulation Model Study of Climate-Chemistry Interaction* from two workshops, the first held 19-21 August 1992 at Oslo, Norway and the second 26-27 May 1993 at Albany, New York, USA. The workshops are the IAMAP activities under the Trace Constituent Working Group, co-chaired by Wei-Chyung Wang and Ivar S. A.. Isaksen (Wang et al., 1992).

The main objective of the two workshops was to recommend specific general circulation model (GCM) studies of the ozone distribution and the climatic effect of its changes. The workshops also discussed the climatic implications of increasing sulfate aerosols because of its importance to regional climate. The workshops were organized into four working groups: observation of atmospheric O_3; modeling of atmospheric chemical composition; modeling of sulfate aerosols; and aspect of climate modeling. Specific recommendations are indicated.

1. Introduction

One of the major findings from the reports of the Intergovernmental Panel on Climate Change (Houghton et al., 1992) and the Ozone Assessment (WMO, 1991) was the identification of possible climatic effects over the last few decades resulting from anthropogenically-induced changes in atmospheric O_3 and sulfate aerosols. The current quantitative estimates of their direct climatic effects indicate significant impacts, although large uncertainties exist. These two atmospheric constituents differ from greenhouse gases in that they are formed by chemical processes in the atmosphere due to the emission of different source gases (e.g. H_2O, CO_2, N_2O, CH_4, SO_2 and the CFCs) of which most are greenhouse gases. Therefore, it is important to include the climate-chemistry interaction in GCMs.

Ozone acts as a greenhouse gas by absorbing outgoing longwave radiation. It also absorbs solar radiation, in particular, the UV-B radiation. Changes in O_3 vertical distribution can perturb the solar and longwave radiative forcing of the troposphere-surface climate system. The effects on the solar and longwave radiative forcing due to changes in atmospheric O_3 are sensitive to the altitudes where O_3 changes. For example, a decrease in stratospheric O_3 will provide a warming effect due to increased available solar radiation for absorption, and a cooling effect due to decreased downward longwave radiation. The net effect will depend on the location and time of the year. On the other hand, an increase in tropospheric O_3 can warm the troposphere-surface system through increases in absorption of both the solar and longwave radiation. The key to the role of O_3 as a climate gas is due to changes which occur in the lower stratosphere and upper troposphere. Changes in absolute O_3 densities in these regions have been demonstrated to lead to the most pronounced impact on surface temperatures (Wang et al., 1980; Lacis et al., 1990). The discussion are therefore focused on the observed ozone changes in these regions.

Sulfate aerosols interact mainly with solar radiation. Increased levels of aerosol concentration have the direct effect of enhancing the Earth's albedo, thereby cooling the surface temperature. The indirect effect is associated with changes in climate forcing through affecting cloud condensation nuclei (CCNs) and composition of the cloud droplets. However, very little is known about this indirect effect on the climate.

Two considerations make O_3 and sulfate aerosols distinctly different from other greenhouse gases, which is of major concern in quantifying their climatic impacts.

- *Ozone and sulfate particles are secondary constituents formed by chemical reactions in the atmosphere*

The relation between these two constituents and their precursors (NO_x, hydrocarbons and CO for ozone, and SO_2 for sulfate particles) depend on the oxidation process in the atmosphere. This process varies strongly with time and space and depends on several other chemical constituents. Key constituents for the O_3 production process are, in addition to the aforementioned precursors, hydrogen radicals like OH and HO_2, and for sulfate formation O_3, OH, and H_2O_2. In order to calculate the formation with sufficient accuracy, extensive chemical modeling is necessary. The O_3 forming process is found to be highly nonlinear with regard to emissions of the precursors. It is also possible that nonlinear effects are present in the sulfate forming process through cloud-chemistry interactions. The modeling of the oxidation process leading to O_3 and sulfate formation in the atmosphere is, therefore, far from straightforward. It is obvious that calculation of changes in O_3 and sulfate particle, and the climatic effects have larger uncertainties than the calculation of changes and the direct climatic effect of greenhouse gases that are emitted into the atmosphere.

- *Anthropogenically-induced changes in the ozone and the sulfate distributions show large spatial and temporal variations in the atmosphere*

The impact from O_3 and sulfate particles is in striking contrast to the impact from other greenhouse gases of which the concentration changes in most cases are uniform throughout the troposphere. The large spatial and temporal variations in O_3 and sulfate particles are a result of their short chemical life times in the atmosphere. In the troposphere, life times are days to weeks, where as in the lower stratosphere, O_3 life times are of the order of months. The climate forcing from O_3 and sulfate particles will therefore be highly non-uniform and the impact on temperatures more difficult to assess than for the well-mixed greenhouse gases. Due to the complex nature of the anthropogenically-induced changes of O_3 and sulfate particles, and our limited knowledge of their impact on climate change, reliable calculations of the climatic impact of O_3 and sulfate particles require the GCMs to include coupling between their atmospheric distribution and climatic impact.

2. Observation of Atmospheric Ozone

The observed decrease in total O_3 amounting to 2-3% over the past two decades is well documented and occurs globally except for the equatorial region. Most of the decrease is thought to be due to the lower stratospheric O_3 decreases. Recent analysis of satellite data (SAGE II between 1984 and 1991) indicate that lower stratospheric O_3 (16-25 km region) at mid-latitudes Northern Hemisphere shows a downward trend of approximately 10% per decade. This is in agreement with ozonesonde observations over Europe for the same time period. At altitudes close to the tropopause, the satellite observations show less O_3 depletion, which is in agreement with the ozonesonde observations. At the 8-12 km altitude range, O_3 actually increased during the past two decades, at least in the middle latitudes of the Northern Hemisphere. In general, ozonesonde observations over Europe show significant O_3 increases in the upper troposphere over the last two decades or so (in some cases up to 2% per year). There are, however, some discrepancies in the analysis of upper tropospheric O_3 trends for other stations. This raises the questions of the geographic extent of the tropospheric O_3 increase, as well as the magnitude of the trend. In order to resolve the discrepancies, a better understanding of the methodology used in analyzing ozonesonde data is warranted. Additionally, there are no indications of tropospheric O_3 increases in the Southern Hemisphere.

One limitation of the ozonesonde data in describing free tropospheric O_3 changes is their limited geographical coverage. Note, however, that on a campaign basis over a limited time, a large number of ozonesonde launches have taken place. The number of ozonesonde stations which conduct regular long term observations is not sufficient to give a good spatial coverage of large scale tropospheric O_3 changes. The number of sonde stations operating on a regular basis are too small and should be increased to get better geographic coverage, especially at low latitudes and in the equatorial region. All the existing ozonesonde network should perform continuous long term observations on a more regular basis than has been done up to present.

Evidence is mounting that the observed decreases in lower stratospheric O_3 are related to chlorine released from CFCs. These relations can be estimated as CFCs are well-mixed constituents in the lower stratosphere. Calculations of the changes in radiative forcing will, however, depend on accurate knowledge of the height profile of O_3 changes. Only limited information on the change in the O_3 height profile is available at present.

Three basic questions have to be addressed when estimates of middle and upper tropospheric O_3 changes from anthropogenic activities are performed:

- What is the in situ O_3 formation in this altitude region of the atmosphere?
- How efficient is the transport of O_3 and its precursors from the planetary boundary layer where these pollutants are emitted ?
- Does the exchange of O_3 between the lower stratosphere and the upper troposphere change with time, and how efficient is this exchange?

Free tropospheric O_3 formation depends critically on the NO_x levels. It has been demonstrated that NO_x levels in large parts of the free troposphere are too low for O_3 formation. Important factors include the release of NO_x from airplanes, formation from lightening, and transport from the boundary layer and from the stratosphere. The short chemical life times of NO_x lead to large variations in its tropospheric distribution. Therefore, reliable calculations of the O_3 production can only be made if we are able to obtain a realistic representation of the global NO_x distribution in the GCMs. Ozone transport from the stratosphere could probably vary substantially from year to year due to changes in weather patterns, masking anthropogenically-induced changes on a short time scale.

It is suggested that continued comprehensive analysis of the vertical distribution of O_3 and the long term trend in the distribution throughout the atmosphere, with special emphasis on the changes in the upper troposphere and lower stratosphere. The analysis should take into account ozonesonde data, satellite data, Umkher data, and lidar observations.

3. Modeling of Atmospheric Chemical Composition

The development of models to calculate O_3 and sulfate formation in the atmosphere has to include the non-linear chemical processes which vary in time and space. For the O_3 chemistry, it is particularly important to consider the strong interaction between the O_3 forming chemistry and the chemical oxidation of the O_3 precursors (NO_x, CO, CH_4 and NMHC, the non-methane hydrocarbons). This necessitates the use of an interactive chemical scheme with a large number of chemical species. In addition to modeling O_3 changes, the model studies should include calculations of the distribution and changes in the O_3 precursors. Calculations of CH_4 and NO_x are probably the most important constituents to be included in the models.

Although CH_4 has a long atmospheric life time (approximately 10 years) and is well-mixed in the troposphere, it interacts strongly with the O_3 chemistry and the atmospheric oxidation process, thereby affecting its own life time (through the abundance of OH). It has been estimated that this interaction with the oxidation process represent a positive feedback for CH_4 in the atmosphere which enhances its atmospheric concentrations by as much as 30%. The global increase in CH_4 (about 0.8-1%/year) that has been observed for more than a decade is probably significant for free tropospheric O_3 formation.

A major problem in modeling the tropospheric O_3 is the limited knowledge of the spatial and temporal distribution of NO_x in the free troposphere. More observations are needed and priority should be given to model its distribution and chemistry. The chemical models should have sufficient spatial resolution to account for variation in NO_x as well as in other short-lived O_3 precursors (e.g., NMHC). There are also indications that the CFC-induced O_3 reductions in the lower stratosphere lead to enhanced UV fluxes in the troposphere, thereby perturbing the chemistry and affecting chemically active greenhouse gases (e.g., CH_4 and O_3). This raises the importance of considering climate-chemistry interactions.

In the sulfate oxidation scheme, studies up to now have shown that there is less interaction with the oxidation process (O_3-OH chemistry). The interaction may turn out to be stronger than previously assumed when liquid phase O_3 chemistry in the troposphere has been fully explored (Jonson and Isaksen, 1993).

There has been a significant development in chemical modeling of the troposphere over recent years with a hierarchy of models. Several global or hemispheric three-dimensional chemical/transport models, which operate either interactive with the transport or in an off-line mode, have been developed. The chemical schemes used vary in complexity. The major refinements include: development of more complex hydrocarbon-NO_x chemistry; the inclusion of heterogeneous chemistry; better representation of the source distribution of the O_3 precursors; and a better representation of the stratosphere/troposphere exchange process.

It is recommended that model studies of the free tropospheric O_3 generation are conducted and particular attention be placed on O_3 and its changes in the upper troposphere and lower stratosphere. Necessary for improvements in model formulation will be a better understanding of the spatial distribution of NO_x in the free troposphere and of the role of cloud processes for the O_3 chemistry.

4. Modeling of Sulfate Aerosols

It has been demonstrated that the direct effect of increasing sulfate burden in the troposphere is likely to have a significant effect on the solar radiative forcing. As these particles are short-lived constituents in the troposphere, the effect is restricted to Northern Hemisphere where most of the anthropogenically-released sulfur emissions occur. The impact on radiative forcing is particularly large over industrial regions. The increased sulfur concentrations are estimated to yield a surface cooling similar in magnitude to the warming from the greenhouse gases over large regions. However, these studies indicate large uncertainty (up to a factor of three) in the magnitude of the radiative effect of enhanced sulfate particles.

There are several areas in which measurements are needed: the chemical composition; number and particle size; and scattering and absorption of aerosol particles. It has been suggested to increase the monitoring activities and campaigns, and to establish new stations in order to study anthropogenic perturbations in different parts of the world.

There are some quantitative estimates of the indirect effects on the radiative forcing from sulfate particles through changes in the CCNs or in the radiative properties of clouds. Estimates of the changes in radiative forcing from these processes are considered to be highly uncertain. It has been shown that industrial emissions influence the CCN and cloud droplets over large areas. There are indications that a change of 30% in CCN could give a significant change in global radiative forcing of 1 Wm^{-2}.

Although the uncertainties are large, new information over the next few years could help reduce uncertainties. This information includes: satellite observations of aerosol and cloud albedo; differences in cloud sizes between the two hemispheres; and albedo sensitivity to droplet numbers. Improved understanding of the processes involved in the formation CCN and more data on the spatial and size distribution of particles is needed in order to improve the estimates of the climatic impact of sulfate particles.

5. Aspect of Climate Modeling

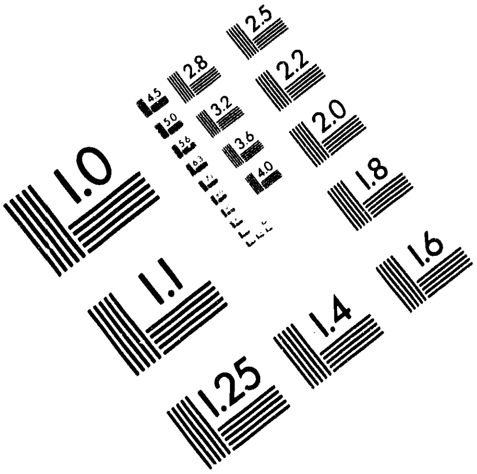
It is well-recognized that the thermal structure of the atmosphere is influenced by the presence of water vapor (H_2O), CO_2, O_3, CH_4, CFCs and N_2O, which, as revealed from satellite measurements, can trap the outgoing longwave radiation at the top of the atmosphere by up to 100-200 Wm^{-2}. Because of the large on-going climate modeling effort as recently updated by Houghton et al. (1992), the discussion focused on the needs for studying the chemical-climate interaction in GCMs.

One of the key issues is the GCM simulated distribution of H_2O, which is part of the hydrologic cycle and exhibits strong seasonal and geographical distribution ranging from a 5 cm column precipitable amount in the tropics to a few tenths of a centimeter in high latitude winter time. Because of the existence of absorption bands across the whole infrared spectrum, a small change of H_2O concentration will have a large effect on the radiative forcing and thus climate. In addition, CH_4 is a source for stratospheric H_2O while tropospheric H_2O is a source for OH, both are important aspect involving climate-chemistry interaction. However, the available data for atmospheric H_2O is not adequate for validating the GCM simulation.

For CH_4, N_2O, $CFCl_3$ and CF_2Cl_2, it has been demonstrated that these gases provide an important radiative energy source for the present climate and that their vertical distribution of infrared opacity are different from CO_2 which may induce different climatic effects especially on a regional basis. These results suggest that these gases need to be explicitly included for studying the greenhouse effect. The concentrations of CO_2, CH_4, N_2O, $CFCl_3$ and CF_2Cl_2 are specified in the general circulation climate models, which is an adequate approximation due to their long life time in the troposphere. However, improvement of the O_3 climatology is needed mainly because the prescribed data for most of GCMs is outdated and does not include a longitudinal variation. Calculations of the climatic impact of the observed lower stratospheric O_3 decreases clearly demonstrated that the decrease could reduce significantly the radiative forcing, particularly at high latitudes. Calculations of the effect of observed changes in the upper troposphere during the 1980s (versus the 1970s) could lead to changes in the radiative forcing with magnitude comparable to the changes from the other greenhouse gases.

Because of the large effect of O_3 changes on the absorption of solar radiation (and thus local temperature), so far two approaches have been used to calculate the radiative forcing due to lower stratospheric O_3 depletion. The first uses fixed-temperature treatment and the second uses adjusted-temperature according to fixed-dynamical heating assumption. The first provides the instantaneous perturbation to the radiative forcing while the second yields the forcing, allowing the rapid adjustment of stratospheric temperature assuming constant dynamical heating. Because of the decreases of lower stratospheric temperature in response to local O_3 depletion, the second approach calculates a negative radiative forcing while the first approach calculates a positive radiative forcing. Radiative forcing calculations also suggest that changes of the vertical O_3 distribution may have large climatic effect, but much work is needed, in particular the GCM studies.

In climate model studies of the impact of O_3 changes, the spatial and temporal variations should be realistically represented. Recognizing the early development of the coupled chemistry-climate models, there is a need to conduct un-coupled GCM simulations so that a better understanding of the importance of climate-chemistry interaction can be achieved. It is recommended that a two-steps approach be



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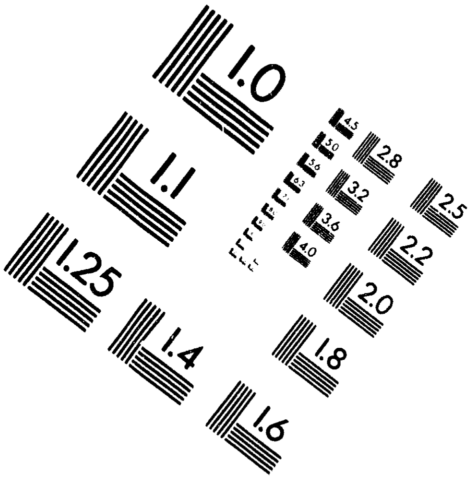

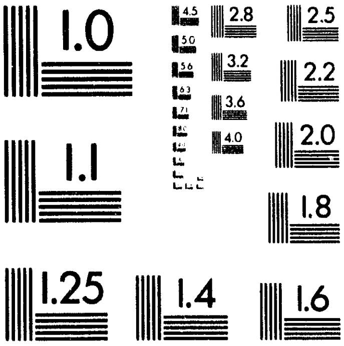

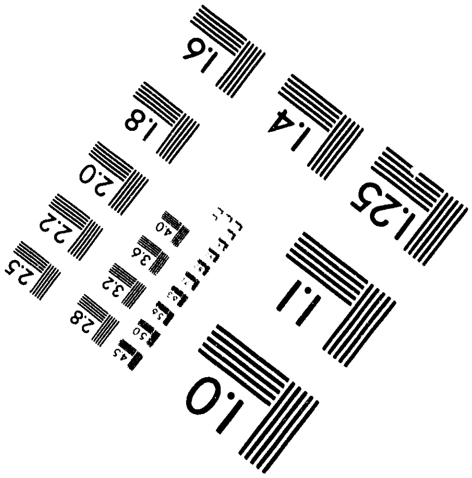

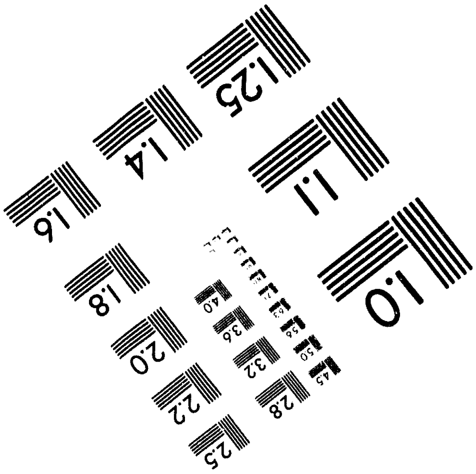

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