Final Technical Report

on

Atmospheric Ozone as a Climate Gas

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Abstract

This report summarizes the major research accomplishments of the project "Atmospheric Ozone as a Climate Gas" for the period July 1, 1994–March 31, 1998. The report is divided into three sections: research summary, publications and participation of graduate students.

The objectives of the research program were: (1) to improve understanding of the physical, chemical and dynamical processes that control mid-latitude O_3 in the lower stratosphere and free troposphere; and (2) to develop improved predictions of future O_3 changes in these regions and their influence on (and response to) future climate changes. The research team includes a subcontractor, Professor Ivar Isaksen of the University of Oslo.

1. Research Summary

1.1 Climatic Effects of Observed Ozone Changes

The observed O_3 climatology, together with radiation and climate models, were used to evaluate the importance of atmospheric O_3 to radiation balance, and to study how the observed O_3 changes during the decades 1980s and 1990s affect radiative forcing and climate as well as their comparison with increasing other greenhouse gases. [Shine et al., 1995; Liang and Wang, 1995; Mohnen et al., 1995a, b; Wang et al., 1995a, b]

1.2 Climate-Chemistry Interaction

Climate-chemistry interactions were studied based on the NCAR GCMs (CCM1, GENESIS, CCM3) and the University of Oslo's chemical-transport models (CTMs). First, the CTMs were used to calculate tropospheric O_3 distribution using a variety of emission scenarios for NO_x , HC, and CO. Next, the O_3 and other gases consistently calculated from the CTMs were used as input to radiation models to study the effect on radiative forcing and UV flux reaching the surface. These experiments were conducted for both the current climate state and the global warming climate state induced by increasing concentrations of greenhouse gases (such as CO_2 , CH_4 , N_2O and CFCs). These studies were summarized in several publications [Berntsen et al., 1996, 1997; Fuglestad et al., 1995, 1996, 1997; Wang et al., 1997]

A 2-D CTM of the global troposphere was used to study the chemical response to: (1) increased UV radiation from stratospheric O_3 depletion and (2) increased temperature and H_2O densities caused by global warming as simulated from GCM. Increased UV radiation increases the photolysis rates for several tropospheric gases, in particular O_3 . This leads to enhanced levels of odd hydrogen and reduced concentrations of tropospheric O_3 . Increases in temperature and water vapor densities reduce the levels of tropospheric O_3 through temperature dependent reaction rates and increased production of odd hydrogen. In both cases, the CH_4 levels are also reduced. Thus the results indicate that the considered mechanisms constitute damping effects on global warming. For a global warming of $3.9^\circ C$ and column moisture increase 27% calculated in Wang et al. (1992, Inadequacy of effective CO_2 as a proxy to assess the greenhouse effect of

other radiatively active gases. (*Geophys. Res. Lett.*, 19, 1375-1378), the levels of tropospheric O_3 and CH_4 are reduced by about 10% and 18%, respectively.

Through chemical interactions in the atmosphere, emissions affect climate indirectly via effects on gases with direct radiative effects on climate. An overview of mechanisms is given together with a preliminary assessment of the climatic impact of changes in tropospheric O_3 and CH_4 in response to emission increase. For a 10% increase in the CH_4 emissions the relative increase in concentration has been estimated to be about 35% larger. Radiative forcing from enhanced levels of tropospheric O_3 is estimated to be about 50% of the direct forcing from CH_4 . Inclusion of these indirect effects (including the effect on stratospheric H_2O) approximately doubles the climatic impact of CH_4 emissions. The effect of NO_x emissions through changes in tropospheric O_3 and CH_4 are of opposite signs. The tropospheric levels of O_3 are increased, while the levels of CH_4 are reduced. For emissions of NO_x from aircraft, the positive effect via O_3 changes are significantly larger than the negative through changes in CH_4 . For NO_x emitted from surface sources, the effects through changes in O_3 and CH_4 are estimated to be of similar magnitude and large uncertainty is connected to the sign of the net effect. Emissions of CO have a positive indirect effect on climate through enhanced levels of tropospheric O_3 and increased lifetime of CH_4 .

We examined the effects of cirrus clouds on O_3 photodissociation rates. The study evaluated the sensitivities of $O(^1D)$ and $O(^3P)$ photodissociation rates to optical thickness, cloud altitudes, solar zenith angle and surface reflectivity. The results showed that: above the cloud $O(^1D)$ rate increases versus clear sky condition and below the cloud the sign of the change depends on the thickness of the cloud layer. Cloud altitude affects the magnitude of $O(^1D)$, but not on $O(^3P)$. The rate decreases as solar zenith angle increases under both clear and cloudy sky conditions. At large solar zenith angles, below-cloud rate is reduced even though the cloud layer is very thin; above-cloud rate is slightly increased. As surface reflectivity increases, the rate below a fairly thick cloud layer can be increased versus the clear sky condition.

A 2-D CTM of the global troposphere was used to study the chemical response to increased temperature and H_2O caused by global warming. Increases in these climate variables reduce the levels of tropospheric O_3 through temperature dependent reaction rates and increased production of odd hydrogen. In this case, the CH_4 levels are also reduced. The results thus indicate that the considered mechanisms constitute a negative feedback effect on global warming.

A 3-D CTM was used to calculate the tropospheric O_3 distribution using a variety of emission scenarios for NO_x , HC, and CO. Changes in O_3 and other gases consistently calculated from the CTMs were used to study the effect on radiative forcing and UV flux reaching the surface. These experiments were conducted for both the current climate state and the global warming climate state induced by increasing concentrations of greenhouse gases (such as CO_2 , CH_4 , N_2O and CFCs). The results indicate that there is a strong differences in the seasonal and geographical distributions of O_3 and radiative forcing perturbation.

1.3 Development of a Coupled Climate-Chemistry Model

We continue the development of a coupled climate-chemistry global model. Progress is being made to incorporate a chemical code used in the University of Oslo's 3-D CTM into CCM3. The chemical code has been tested against a more accurate Gear code. The comparison shows that the distribution of the key gases in the O_3 chemistry is accurately simulated. During the initial phase, we focus on the interface of input and output as well as the transport of trace species. In addition, in parallel to the coupled model development, we examine the consistency in the physical and chemical processes important to both climate and chemistry.

2. Publications

There were six (6) refereed journal articles, four (4) book articles, and two (2) proceedings manuscripts.

Berntsen, T., I. S. A. Isaksen, W.-C. Wang and X.-Z. Liang, 1996: Impacts of increased anthropogenic emissions in Asia on tropospheric ozone and climate: A global 3-D model study. *Tellus*, 48B, 13-32.

Berntsen, T., I. S. A. Isaksen, J. S. Fuglestad, G. Myhre, F. Stordal, R. S. Freckleton, and K. P. Shine, 1997: Effects of anthropogenic emissions on tropospheric ozone and its radiative forcing. Submitted to *J. Geophys. Res.*

Fuglestad, J. S., I. S. A. Isaksen, and W.-C. Wang, 1996: Estimates of indirect global warming potentials for CH_4 , CO , and NO_x . *Climatic Change*, 34, 405-437.

Fuglestad, J. S., J. E. Jonson, W.-C. Wang, and I. S. A. Isaksen, 1995: Responses in tropospheric chemistry to changes in UV fluxes, temperatures and water vapor densities. in *Atmospheric Ozone as a Climate Gas*, (Eds) W.-C. Wang and I. S. A. Isaksen, 145-162, NATO ASI Series, Springer-Verlag, Berlin.

Fuglestad, J. S., T. K. Berntsen, I. S. A. Isaksen, H. Mao, X.-Z. Liang and W.-C. Wang, 1997: Impacts of reduced NO_x emissions on radiative forcing through changes in tropospheric O_3 and CH_4 : A global 3-D model study. *Proceedings of XVIII Quadrennial Ozone Symposium-96*, September 12-21, 1996, L'Aquila, Italy.

Liang, X.-Z. and W.-C. Wang, 1995: A GCM study of the climatic effect of 1979-1992 ozone trend. *Atmospheric Ozone as a Climate Gas*, (Eds) W.-C. Wang and I. S. A. Isaksen, 259-288, NATO ASI Series, Springer-Verlag, Berlin.

Mohnen, V. A., W. Goldstein, and W.-C. Wang, 1995a: A new challenge: Assessing tropospheric ozone as a climate gas. *Atmospheric Environment*, 29, 641-642.

Mohnen, V. A., W. Goldstein, and W.-C. Wang, 1995b: The potential role of tropospheric ozone as a climate gas. *Bull. World Meteorol. Org.*, 44, 38-42.

- Shine, K. P., B. P. Briegleb, A. S. Grossman, D. Hauglustaine, H. Mao, V. Ramaswamy, M. D. Schwarzkopf, R. Van Dorland, and W.-C. Wang, 1995: Radiative forcing due to changes in ozone: A comparison of different codes. *Atmospheric Ozone as a Climate Gas*, (Eds) W.-C. Wang and I. S. A. Isaksen, 373-396, NATO ASI Series, Springer-Verlag, Berlin.
- Wang, W.-C., H. Mao, I. S. A. Isaksen, J. S. Fuglestad, and S. Karlsdottir, 1997: Indirect Effects of Increasing Atmospheric Methane on the Radiative Forcing Through Climate-Chemistry Interactions. Proceedings of XVIII Quadrennial Ozone Symposium-96, September 12-21, 1996, L'Aquila, Italy.
- Wang, W.-C., M. P. Dudek, and X.-Z. Liang, 1995a: Chapter 9: The greenhouse effect of trace gases. 317-346, *Future climates of the World*, (Ed.) A. Henderson-Sellers, Elsevier Science Publ. Number 16 of the World Survey of Climatology.
- Wang, W.-C., X.-Z. Liang, M. P. Dudek, D. Pollard and S. L. Thompson, 1995b: Atmospheric ozone as a climate gas. Atmospheric Research, 37, 247-256.

3. Participation of Graduate Students

Huiting Mao, a Ph.D. candidate at the Department of Earth and Atmospheric Sciences, SUNYA, has been studying radiative forcing and climate issues related to tropospheric O_3 . She is currently working on the study of the consistency of physical and chemical processes between the CTM and global climate model.

Jostein Sundet, a Ph.D. student at the Department of Geophysics, University of Oslo, has developed a new version of the CTM (OSLO CTM2) which will be used in the project. This model is flexible with regard to resolution (T21-T63) and uses the winds from ECWMF. Another Ph.D. student, **Sigrun Karlsdottir**, has studied CH_4 feedback processes with 3-D CTM (OSLO CTM1) and the impact of climatic change (through changes in temperature and H_2O).