Working Paper 1997:1

Impacts of reduced NOx emissions on radiative forcing through changes in tropospheric O_3 and CH_4 - a global 3-D model study

by

Jan S. Fuglestvedt[1], Terje K. Berntsen[1,2],
Ivar S.A. Isaksen[1,2], Huiting Mao[3]
Xin-Zhong Liang[3] and Wei-Chyung Wang[3]

January 1997

This paper is accepted for publication in
"Proceedings of the Ozone Symposium-96"
(where it will appear in abridged form).

[1]CICERO, University of Oslo, Norway
[2]Department of Geophysics, University of Oslo, Norway
[3]Atmospheric Sciences Research Center, State University of New York, Albany, USA

ABSTRACT

A three-dimensional global chemical tracer model and a radiation transfer model have been used to study the role of reduced NOx emissions on radiative forcing. Through production of tropospheric O_3, NOx emissions lead to positive radiative forcing and warming. But NOx also reduces the levels of CH_4, thereby giving negative forcing and cooling. The lifetime of NOx varies from hours to days, giving large spatial variations in the levels of NOx, and, due to non-linearities in the chemistry, there are also large geographical differences in the effects of NOx on O_3 as well as on OH and CH_4. We have selected six geographical regions representing different chemical and physical conditions, and the emissions of NOx in these regions are *reduced* by 20%. The sensitivity in the chemical responses and the radiative forcing due to changes in O_3 and CH_4 show large variations between the regions. The ozone and methane forcing are of opposite sign and generally of similar magnitude.

TABLE OF CONTENTS

1. INTRODUCTION

Several studies have shown the importance of tropospheric O_3 as a climate gas (e.g. Wang et al., 1980; Lacis et al., 1990; Wang et al., 1993; Hauglustaine et al., 1994a). There are some important differences between ozone and greenhouse gases such as CO_2 and N_2O since O_3 itself is not emitted in significant amounts, but is produced through chemical reactions in the atmosphere. Furthermore, due to a lifetime in the order of a few weeks to a few months depending on season and region, there are, in contrast to the long-lived gases, large spatial variations in the concentrations of O_3. Furthermore, the vertical distribution of changes in O_3 is important since the climate sensitivity to O_3 changes reaches a maximum in the upper troposphere and lower stratosphere (Wang et al., 1980; Lacis et al., 1990).

The concentrations of tropospheric ozone have increased (e.g. Bojkov, 1988), and observations and model studies indicate that the amount of O_3 in the Northern Hemisphere may have doubled since pre-industrial times (IPCC, 1994). The enhanced levels of ozone can be attributed to increased emissions of NOx, CO and hydrocarbons that produce tropospheric ozone under the influence of solar radiation. As shown i.a. by Isaksen et al. (1978), Liu et al. (1987) and Lin et al. (1988) there is a strongly non-linear relation between the levels of NOx and the ozone production. NOx emitted or being transported to the remote troposphere is more efficient in producing ozone than if it were emitted in a polluted region and oxidised there.

But NOx does not only affect climate by controlling the levels of ozone. NOx has a dual role with respect to radiative forcing and climate change since emissions of this gas also increase the concentrations of OH radicals. This reduces the lifetimes of gases removed by OH. Several climate gases belong to this group, e.g. CH_4. Emissions of NOx therefore lead to a *positive indirect effect* on climate through O_3 enhancements, and a *negative indirect effect* through reduced lifetimes of CH_4 and other climate gases removed by OH (IPCC, 1992; Fuglestvedt et al., 1996). Lelieveld and Dorland (1995) studied the changes in atmospheric composition since pre-industrial times using a 3-D model and found that the emissions of NOx had given a positive radiative forcing through ozone changes that was of similar magnitude to the negative forcing through reduced lifetime of CH_4.

2. MODELS

To be able to take the large zonal variations in the concentration of NOx into account we use a 3-D chemistry/transport model of the global troposphere. The model is based on the 3-D Chemical Tracer Model (CTM) developed at NASA/GISS (Prather et al., 1987), with the incorporation of an extensive chemical scheme to calculate transport and chemical transformation of species affecting the photochemistry in the troposphere. A detailed description of the model is given in Berntsen and Isaksen (1996). The chemistry scheme contains 50 chemical species and a full diurnal cycle is calculated with a time step of 30 minutes. The chemical species are removed through dry deposition at the surface and through scavenging of soluble species by rain. The NOx emissions and the regional distribution are taken from Dignon et al. (1992). NOx emissions from lightning are distributed according to deep convective activity with a total of 5.2 TgN/yr. Aircraft emissions (0.6 TgN/yr) are obtained from British Aerospace (1994). The total emission of CO is taken from Hough (1991) with the same distribution as for NOx emissions. NMHC emissions are based on Watson et al. (1990) and Isaksen and Hov (1987). The model horizontal resolution is 8° latitude x 10° longitude. There are 9 vertical layers from the surface to 10 hPa. The upper two layers (above ~14 km altitude) considered to be in the stratosphere, are treated as boundary conditions, with pre-set, climatological concentrations of O_3 and NOy-species.

The radiative forcing from the modelled ozone changes is calculated using radiation code from SUNYA/NCAR GENESIS model (Thompson et al., 1995; Wang et al., 1995). The radiative forcing due to ozone changes is calculated by comparing changes in the net radiation flux (the solar and longwave radiation fluxes) at the tropopause between the reference ozone distribution and the one with changes in ozone modelled with the CTM. The other input data which include temperature, moisture,

cloudiness, incoming solar radiation and surface albedo are based on the climate model simulations of present climate documented in Wang et al. (1995), and all these parameters are functions of latitude, longitude, altitude and season.

3. RESULTS

In order to study the geographical variation in the O_3 and CH_4 responses to reductions in NOx emissions six regions representing different chemical and physical conditions are selected (Figure 1). The surface emissions of NOx in these regions are *reduced by 20%* in separate model tests. Table 1 shows the emissions and the changes in the emissions for the different groups.

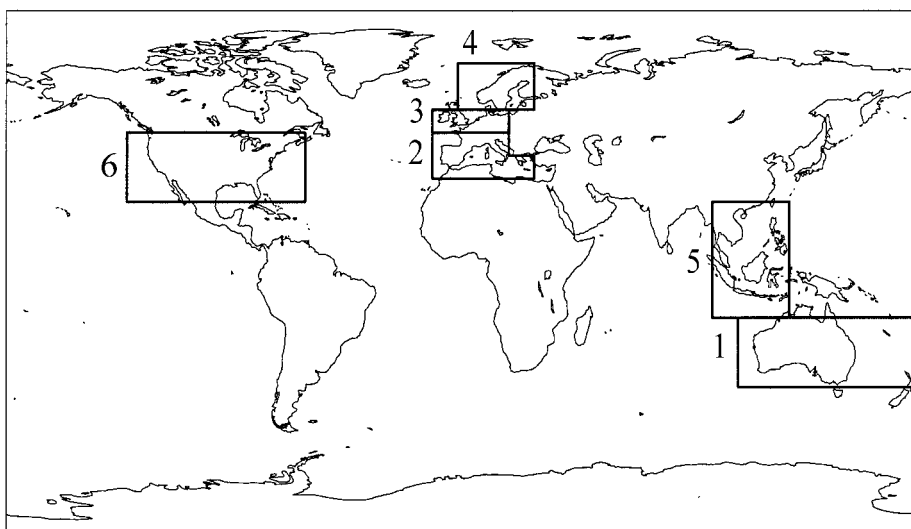A model run with the unperturbed emissions was performed in order to have a reference distribution.

Figure 1. The location of the emission reductions.

Figure 2 shows the modelled monthly mean distribution of NOx (pptv) in the reference run for the lowest layer and for approximately 12 km for June. Due to the short lifetime and the large geographical variations in the emission rates of NOx there are large spatial and temporal variations in the concentrations. In remote regions the NOx levels are as low as a few pptv in the lowest model layer. These regions are generally over the oceans. Over the continents the NOx levels are much higher, especially over the polluted regions where they reach several ppbv. In the upper troposphere the regional variations are somewhat less pronounced, but still the effects of surface emissions can be seen due to effective vertical transport from the surface. The large variations are important for the chemical responses to changes in emissions since NOx is a key species in several chemical processes.

Table 1. Changes in the surface emissions of NOx in the various regions.

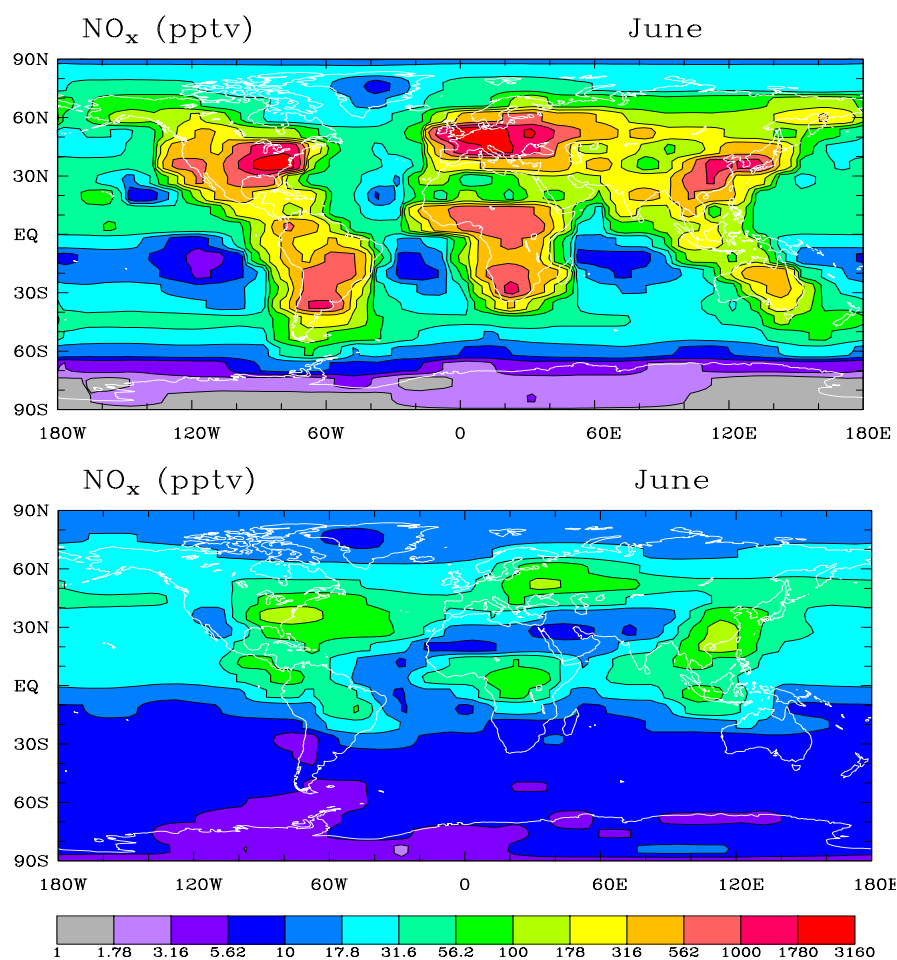| Region	1 Australia	2 Southern-Europe	3 Central Europe	4 Scandinavia	5 Southeast Asia	6 USA
NOx emission (TgN/yr)	0.84	1.97	2.22	0.21	1.18	7.53
Emission reduction (-20%) TgN/yr	0.169	0.393	0.444	0.043	0.236	1.506

Figure 2. Reference distribution of NOx (pptv) at 0.25 km (upper figure) and 12 km for June (lower figure).

Figure 3 shows the modelled distribution of O_3 (ppbv) in the reference run for the lowest layer and for approximately 12 km in June. The figures show that for this month the ozone distribution broadly follows the NOx distribution, although the pattern is somewhat smoothed out due to the longer lifetime of ozone.

Figure 4 shows the modelled distribution of OH (molecules/cm^3) in the reference run for the lowest layer in June. The OH levels are highest at latitudes with strong solar insolation and high humidity. The OH levels are also higher over the continents due to higher O_3 and NOx levels here. The marked increase in OH at low latitudes are important for the spatial distribution of the methane oxidation. High OH levels correlate with high temperatures which make low latitudes important for oxidation of CH_4 and other hydrocarbons. Due to the reduction in temperature and air density with height, most of the oxidation occurs in the lower troposphere. This means that OH changes at low altitudes and low latitudes are most important for changes in methane.

Figure 5 and 6 show the calculated changes in ozone at 12 km altitude for 20% reductions in NOx emissions in USA and Southeast Asia, respectively. For Southeast Asia the ozone decreases are mainly confined to the region where the emission reductions occur and reach 6% at 12 km. For emission
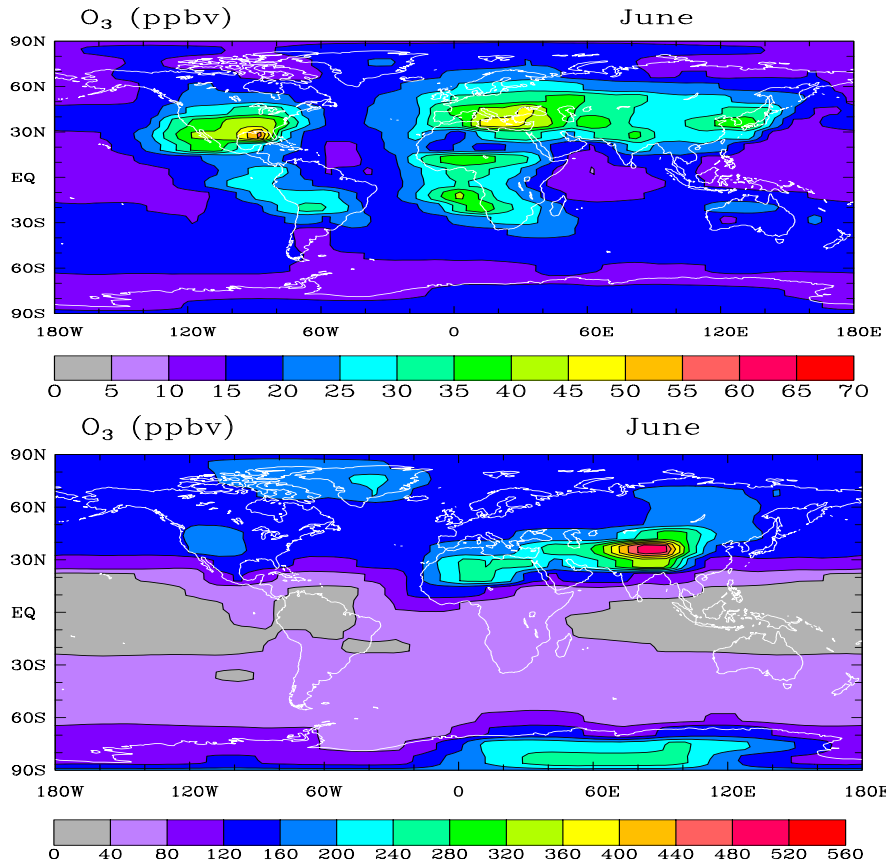
9

Figure 3. Reference distribution of O$_3$ at 0.25 km (upper figure) and 12 km for June (lower figure).

reductions in USA the changes are somewhat more smoothed out longitudinally, and the reductions are typically around 1-2%.

As shown in table 1 there are large differences in the magnitudes of the emission changes between the regions. In order to compare the responses and the sensitivities of the responses between the regions a sensitivity factor is defined as

$$\alpha \quad = \quad \frac{\Delta \overline{O}_3}{\Delta E_{NOx}} \left(\frac{ppbv}{TgN / yr} \right)$$

where $\Delta \overline{O}_3$ is given as change in global mean (ppbv) and ΔE_{NOx} is the change in NOx emissions in the region under consideration. Figure 7 shows α as function of month for emission reductions in USA and Southeast Asia. The highest sensitivities to NOx reductions are calculated for Southeast Asia and Australia, while USA and southern Europe have the lowest sensitivities. In contrast to the low latitude regions, the O$_3$ changes for regions at high latitudes show significant seasonal variation, particularly in the upper troposphere due to the seasonal cycle in convective activity.

Changes in NOx emissions will not only affect ozone concentrations but also the concentrations of OH and thereby also the level of methane. When emissions of NOx to the atmosphere are changed, the following sequence of events will take place:
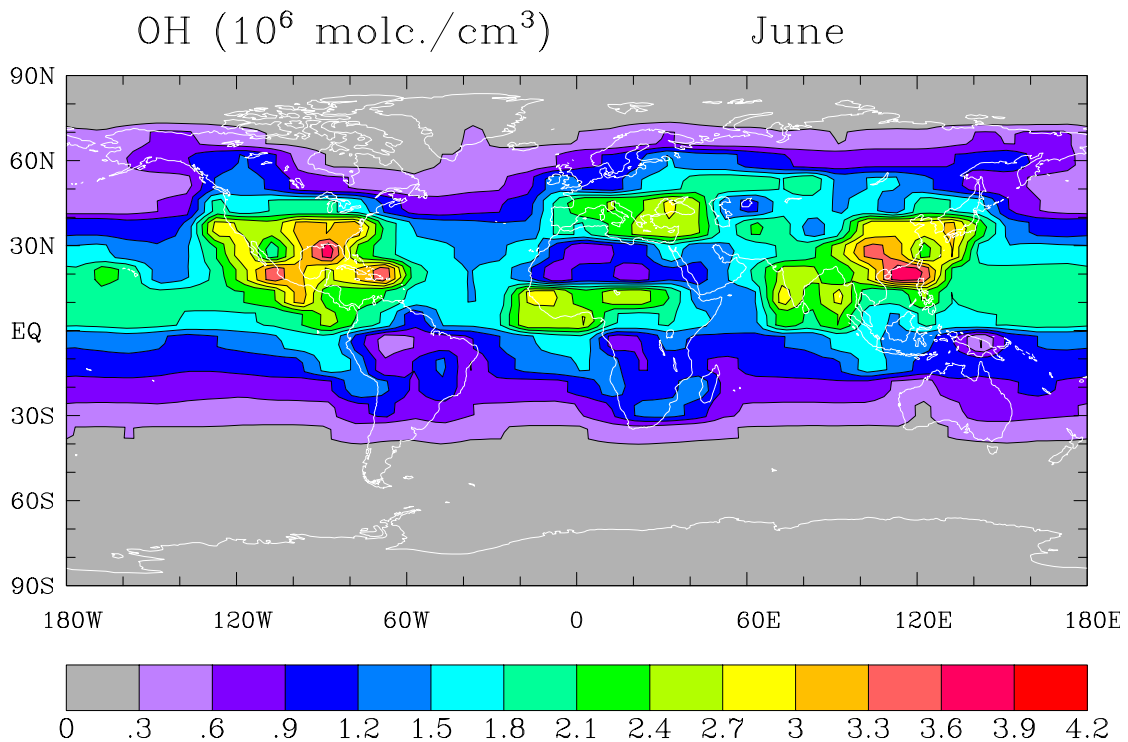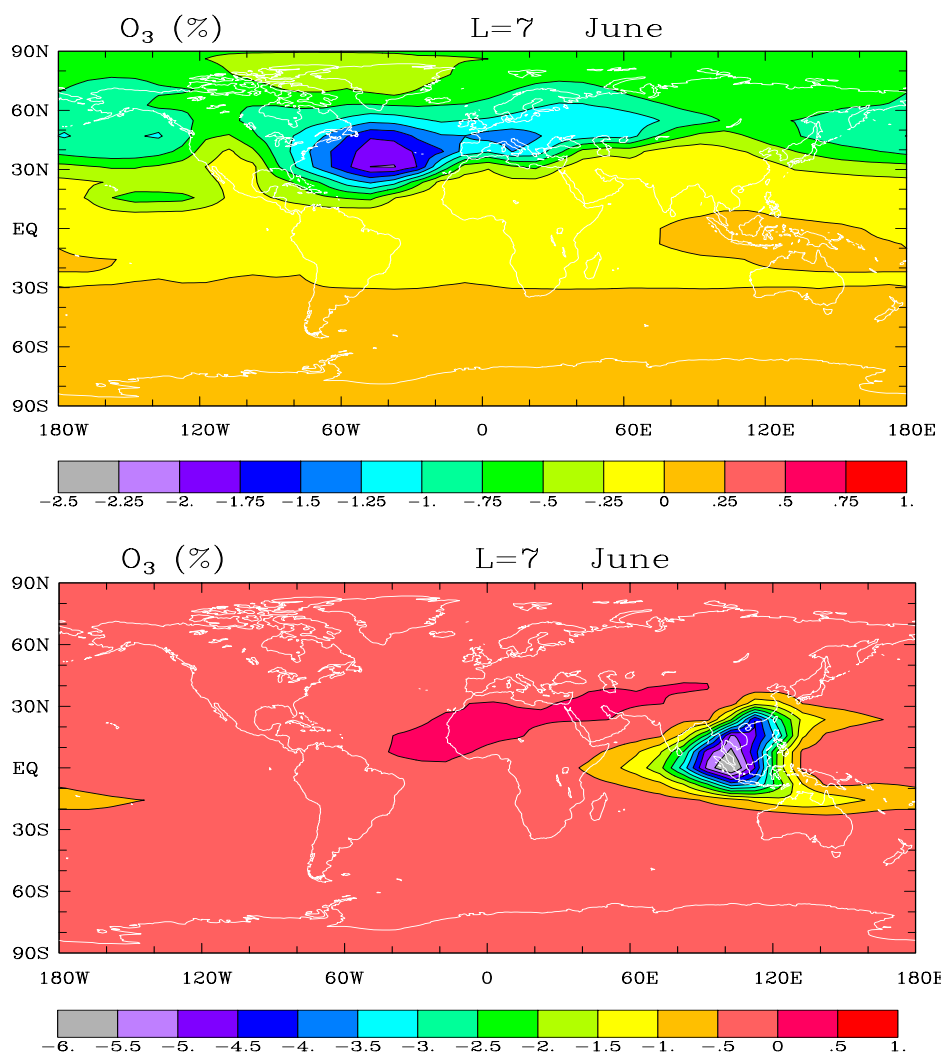
Figure 4. Modelled distribution of OH in the reference case for the lowest layer in June.

$$\Delta E_{NOx} \Rightarrow \Delta[OH]_0 \Rightarrow \Delta[CH_4]_0 \Rightarrow \Delta[OH] \Rightarrow \Delta[CH_4] \Rightarrow \Delta[OH] \Rightarrow \text{etc.}$$

The additional change in CH_4 concentration following the initial change in the concentration of this gas ($\Delta[CH_4]_0$), is a consequence of the feedback effect between CH_4 and OH. Since CH_4 has an adjustment time in the order of 9-15 years, the model has to be run for several decades in order to reach steady state for this gas. Instead only the initial change in CH_4 and OH were calculated by the 15 months model simulation, and, based on previous studies with the same model, the steady state change in lifetime and concentration of methane were calculated assuming that the feedback does not vary significantly with the magnitude of change in concentration. Karlsdóttir and Isaksen (1997) calculated a 7% increase in the lifetime of methane for a 20% increase in concentration. The calculated changes in CH_4 for the various model tests are given in table 2.

Table 2. Normalized forcing for 20% reduction in NOx emissions.

Perturbation	ΔCH_4 (ppbv)	$\Delta F(CH_4)$ (10^{-2} W/m^2)	$\Delta F(CH_4)/\Delta E_{NOx}$ (10^{-2} W/m^2/TgN/yr)	$\Delta F(O_3)/\Delta E_{NOx}$ (10^{-2} W/m^2/TgN/yr)
Group 1, Australia	7.27	0.317	- 1.88	
Group 2, S-Europe	3.85	0.168	- 0.43	
Group 3, Central-Europe	3.22	0.141	- 0.32	
Group 4, Scandinavia	0.34	0.015	- 0.35	0.29
Group 5, Southeast Asia	11.5	0.501	- 2.12	2.37
Group 6, USA	15.7	0.684	- 0.45	0.35

Figure 5 and 6. Changes (%) in O_3 at 12 km for June in response to 20% reduction in NOx emissions in USA and Southeast Asia, respectively.

As for the changes in O_3, the CH_4 responses show large variation between the regions. Differences in the chemical environment of the regions, especially in the background level of NOx, are of great importance. Two regions, Australia and Southeast Asia stand out as being much more sensitive to NOx changes than the other regions.

For the modelled changes in O_3, radiative forcing is calculated with the radiation transfer model described above. The calculated O_3 forcing due to 20% reductions in NOx emissions is noticeable on a regional basis, but in terms of global annual averages the effects are negligible. However, if the radiative forcing is regionally heterogeneous the impact on the dynamics of the atmosphere can be significant even for a relatively small forcing. For USA, the forcing reaches 0.1 W/m^2 at 30°N and 40°W. The O_3 forcing shows a strong seasonal variation for emission reductions in USA, with minimum in February and maximum in July. For Southeast Asia the variation is small. Table 2 shows the calculated global annual forcing due to changes in CH_4 and O_3.
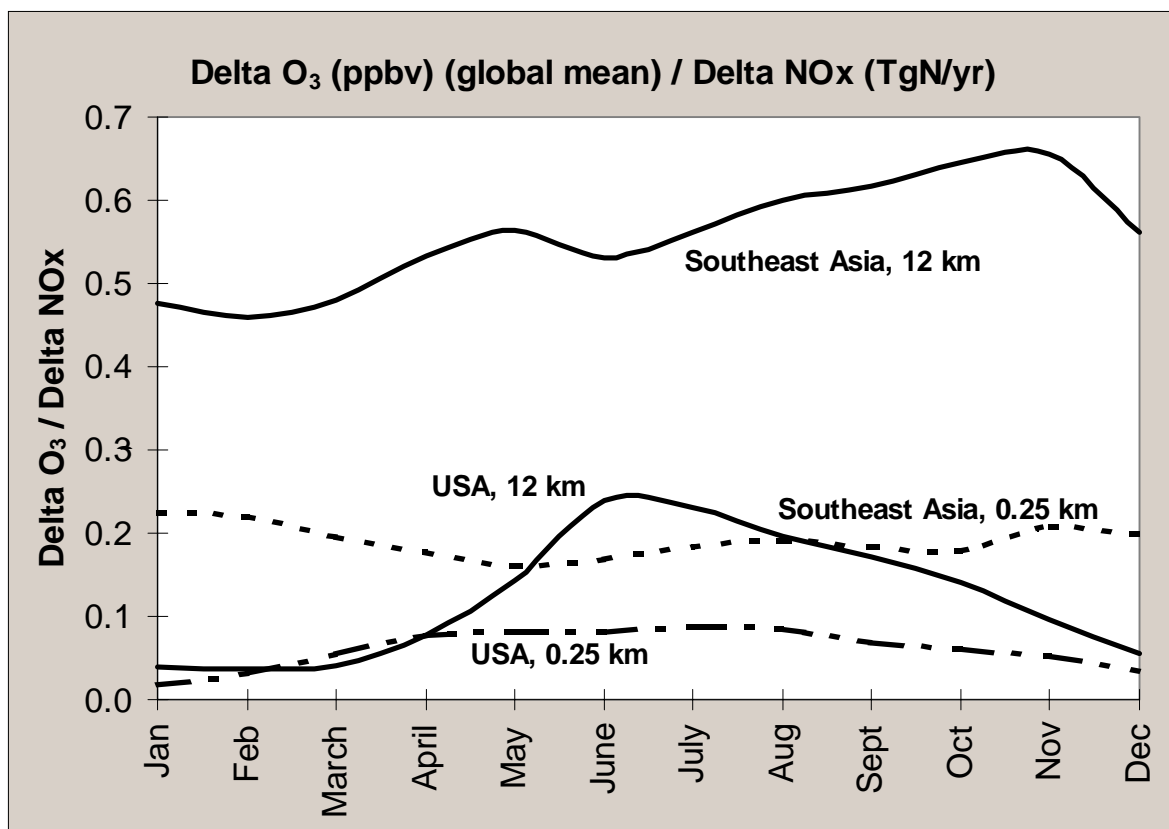
Figure 7. Calculated O$_3$ change sensitivities (α).

4. DISCUSSION AND CONCLUSIONS

The calculations show large variations between the regions in the chemical responses to changes in emissions and the responses vary significantly both with respect to annual cycle of the changes, as well as magnitude of the changes. The climatic impacts in terms of radiative forcing also show significant regional variation. Due to spatial variations in climate parameters such as surface and tropospheric temperatures, cloud cover etc., identical changes in ozone concentrations in different regions do not give equal radiative forcing.

Since the ozone and methane responses to NOx reductions counteract each other it is of interest to compare the radiative forcing from these changes. Such comparisons are however hampered by the very different natures of these two responses. The radiative forcing from methane changes shows a quite homogeneous global pattern, while the ozone effect is much more regional, in spite of the longer lifetime for O$_3$ at higher altitudes. In addition, due to the relatively long lifetime of methane, the response is delayed accordingly, while the ozone response is occurring during a few weeks.

Figure 8 shows the global annual radiative forcing from changes in O$_3$ and CH$_4$ in response to 20% NOx reductions in the three regions USA, Southeast Asia and Scandinavia. The forcing in Southeast Asia and USA are of similar magnitudes (both for ozone and methane), while the effects for Scandinavia are very small compared to the two other groups. For all groups, the ozone forcing and the methane forcing are of similar magnitude, but of opposite sign.

If the numbers for methane and ozone forcing are normalised to the NOx changes in the regions, the picture is somewhat different, see table 2. Significantly higher sensitivities in terms of radiative forcing to reductions in NOx emissions are found for Southeast Asia compared to the other regions. On a per mass basis, the radiative forcing sensitivity to NOx changes are of similar magnitudes for USA and Scandinavia. The ozone forcing sensitivity for Southeast Asia is larger than for Scandinavia by a factor of ~8, while the sensitivity for USA is ~20% larger than for Scandinavia. For the methane forcing, the highest sensitivity is calculated for Southeast Asia, and the lowest for Central Europe (a ratio of ca. 7).

Global annual radiative forcing from changes in O$_3$ and CH$_4$ in response to 20% reduction in NOx emissions
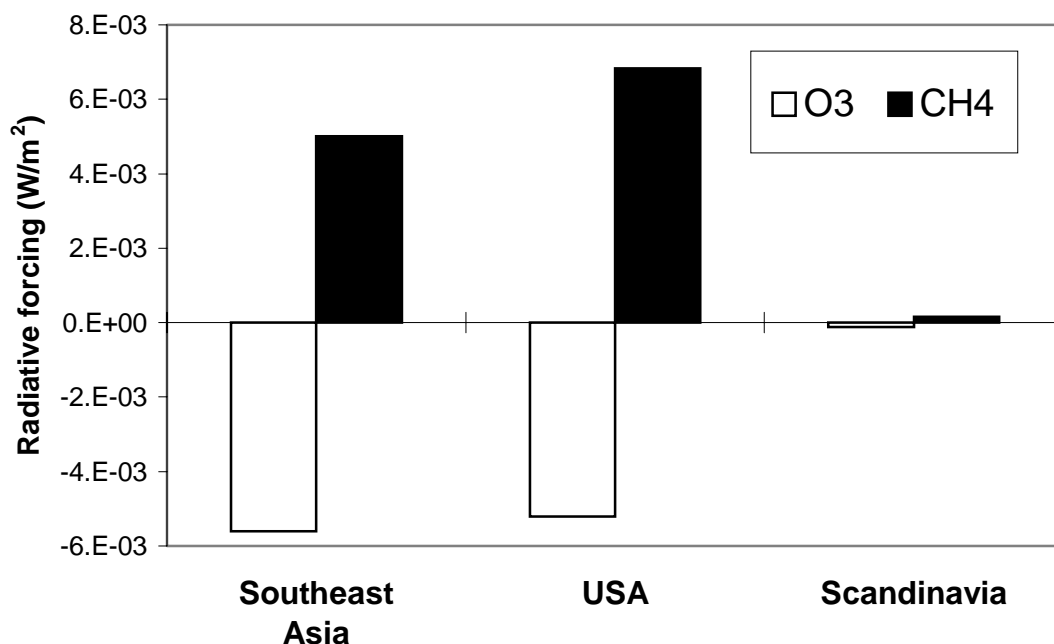

Figure 8. Global annual forcing from changes in O$_3$ and CH$_4$ in response to 20% reduction in NOx emissions from surface sources.

The opposing effects of O$_3$ and CH$_4$ responses are also found in several other studies, e.g. Lelieveld and Dorland (1995); Johnson and Derwent (1996), Fuglestvedt et al. (1996); Hauglustaine et al. (1994b). For a 100% increase in global surface emissions of NOx, Hauglustaine et al. (1994b) calculate with a 2-D model a sensitivity of $0.34 \cdot 10^{-2}$ Wm^{-2}/TgNyr^{-1}. In this 3-D study the estimated sensitivities for USA and Scandinavia are of similar magnitude, $0.35 \cdot 10^{-2}$ Wm^{-2}/TgNyr^{-1} and $0.29 \cdot 10^{-2}$ Wm^{-2}/TgNyr^{-1}, respectively, while Southeast Asia shows a much higher sensitivity, $2.37 \cdot 10^{-2}$ Wm^{-2}/TgNyr^{-1}.

This study shows that emissions of NOx have complex and potentially important impacts on climate through atmospheric chemistry interactions. These impacts are, however, very different in nature; one global methane effect with a delay of approximately a decade, and an ozone effect that is of regional character with an almost instantaneous adjustment. The sensitivity of the responses in methane and ozone shows large variations between the regions considered.

5. ACKNOWLEDGEMENTS

6. REFERENCES

Berntsen, T.K., and Isaksen, I.S.A., 1996. A global 3-D chemical transport model of the troposphere; 1 Model description and CO and O_3 results. Submitted to *J. Geophys. Res.*

Bojkov, R.D., 1988: Ozone changes at the surface and in the free troposphere. In: Tropospheric Ozone, I.S.A. Isaksen (ed.), D. Reidel Publishing Company, 83-96.

British Aerospace (1994). Personal communication, C. J. Hume.

Dignon, J. 1992. NO_X and SO_X emissions from fossil fuels: a global distribution. *Atmos. Environ.,* 26A, 1157-1163.

Fuglestvedt, J.S., Isaksen, I.S.A. and Wang, W.-C. 1996. Estimates of Indirect Global Warming Potentials for CH_4, CO and NO_X. *Climatic Change*, 34, 405-437.

Hauglustaine, D. A., Granier, C., Brasseur, G. P. and Mégie, G. 1994a. The importance of atmospheric chemistry in the calculation of radiative forcing of the climate system. *J. Geophys. Res.,* 99, 1173-1186.

Hauglustaine, D. A., Granier, C., Brasseur, G. P. and Mégie, G. 1994b. Impact of present aircraft emissions of nitrogen oxides on tropospheric ozone and climate forcing. *Geophys. Res. Lett,* 21, 2031-2034.

Hough, A.M. 1991. Development of a two-dimensional tropospheric model. Model chemistry. *J. Geophys. Res.,* 96, 7325-7362.

Intergovernmental Panel on Climate Change (IPCC). 1992. *Climate change 1992. The Supplementary Report to The IPCC Scientific Assessment,* Houghton, J.T., Callander, B. A. and Varney, S.K. (Eds.). Cambridge University Press, Cambridge, U.K.

Intergovernmental Panel on Climate Change (IPCC). 1994. *Radiative forcing of Climate change.* The 1994 Report of the Scientific Assessment Working group of IPCC.

Isaksen, I.S.A., Hov, Ø. and Hesstvedt, E. 1978. Ozone generation over rural areas. *Envir. Sci. Tech.*, 12, 1279-1284.

Isaksen, I.S.A. and Hov, Ø. 1987. Calculations of trends in the tropospheric concentrations of O_3, OH, CO, CH_4 and NO_X. *Tellus*, 39B, 271-283.

Johnson, C. E. and Derwent, R.G. 1996. Relative Radiative Forcing Consequences of Global Emissions of Hydrocarbons, Carbon Monoxide and NO_X from Human Activities Estimated a Zonally-averaged Two-dimensional Model. Accepted for Publication in *Climatic Change*.

Karlsdottir, S. and Isaksen, I.S.A. 1997. Feedback calculations of methane; a 3-D model study. In prep.

Lacis, A. A., Wuebbles, D. J. and Logan, J. A. 1990. Radiative forcing of climate by changes in the vertical distribution of ozone. *J. Geophys. Res.*, 95, 9971-9981.

Lelieveld, J., and van Dorland, R.: 1995, Ozone chemistry changes in the troposphere and consequent radiative forcing of climate, in Wang, W.-C. and Isaksen, I.S.A. (eds.), *Atmospheric ozone as a Climate gas*, NATO ASI series, Springer-Verlag Co. Heidelberg.

Lin, X., Trainer M. and Liu S. C. 1988. On the nonlinearity of the tropospheric ozone production. *J. Geophys. Res.*, 93, 15879-15888.

Liu, S. C., Trainer M., Fehsenfeld F. C., Parrish D. D., Williams E.J., Fahey G. Hübler D.W. and Murphy, P.C. 1987. Ozone production in the rural troposphere and the implications for regional and global ozone distributions. *J. Geophys. Res.,* 92, 4191-4207.

Prather, M., McElroy, M., Wofsy, S., Russel, G. and Rind, D. 1987. Chemistry of the global troposphere: fluorocarbons as tracers of air motion. *J. Geophys. Res.,* 92, 6579-6613.

Thompson, S. L. and Polland. 1995. A global climate model (GENESIS) with a land-surface-transfer scheme (LSX). Part 1: Present-day climate. *J. Climate*, 8, 732-761.

Wang, W.-C., Pinto, J.P. and Yung, Y.L. 1980. Climatic effects due to halogenated compounds in the earth's atmosphere. *J. Atmos. Sci.*, 37, 333-338.

Wang, W.-C., Zhuang, Y.-C. and Bojkov, R.D. 1993. Climate implications of observed changes in ozone vertical distributions at middle and high latitudes of the Northern Hemisphere. *Geophys. Res. Lett.*, 20, 1567-1570.

Wang, W.-C., Liang, X.-Z., Dudek, M.P., Polland, D. D. and Thompson, S.L., 1995: Atmospheric ozone as a climate gas. *Atmos. Res.*, 37, 247-256.

Watson J.J., J.A. Probert, S.D. Piccot, J.W. Jones, 1991. Global inventory of volatile organic compound emissions from anthropogenic sources. EPA-report EPA-600/8-91-002.