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Perturbation to global tropospheric oxidizing capacity due to latitudinal redistribution of surface sources of NO_x, CH₄ and CO

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Abstract. Economic and social projections indicate that during next several decades there will be major geographical redistribution of surface emissions of O₃ precursors, such as NO_x, CH₄ and CO. A net decrease in their emissions from northern hemispheric mid-latitudes will be accompanied by substantial increases from the tropics. We have investigated a hypothetical scenario of currently underway transition of such emission patterns using a global two-dimensional photochemical model. With overall O₃ precursor releases held constant, a simultaneous transfer of their emissions by 25% from the latitude belt 75°N-35°N to 5°S-35°N increases tropospheric oxidizing capacity such that the methane global lifetime and concentrations fall by more than 3%. Seasonally dependent changes in surface O₃ concentrations are also calculated. In influencing OH concentration, redistribution of surface NO_x emissions is 2-3 orders of magnitude more efficient per unit mass than CO emissions. Shifts in methane sources have insignificant effects on global photochemistry, but lead to a decrease in its interhemispheric gradient.

Introduction

Since the beginning of the industrial era, human activities have been influencing the global atmosphere, causing shifts from its natural state. Current projections of socio-economic indicators [Population Reference Bureau, 1997; World Resources 1996-97; Hargreaves et al., 1994] suggest that the developed countries (which we refer to as the North) are entering into an era of social and economic stability while the developing countries (here the South) are in the process of sustaining or even enhancing their industrial and urban growths. These disparities in demographic and economic change are imposing geographically variable human impacts on the global environment through the emissions of air pollutants.

In the future, major redistribution of human-related emissions of NO_x (defined as NO₂+NO), hydrocarbons and CO are all likely. A net decrease is predicted from the North accompanied by an increase in emissions from the South [World Energy Council, 1993; EPA, 1995; OECD, 1995; EMEP, 1997]. This redistribution may be accompanied by a

net increase in the global source strength for each of these pollutants [Graedel and Crutzen, 1991; Alcamo et al., 1994; Lee et al., 1997]. Some developed countries are already reducing their CO and NO_x emissions by as much as 15% [World Resources 1996-97]. Qualitatively similar to trends in SO₂ emissions and concentrations in the Eastern United States [Husain et al., 1998], OECD [1995] has reported decreases in NO_x concentration over several European countries and the United States. Note also that in Eastern Europe, changes in political system accompanied by shifts in industrial management from governments to the private sector are already resulting in a control on the emissions of air pollutants from the NH mid- and high latitudes [Benkovitz et al., 1996; Schooner et al., 1997]. On the other hand, for 1980-1987, Kato and Akimoto [1992] estimated a 35-55% increase in NO_x emissions from developing Asian countries. Dignon and Hameed [1989] calculated that between 1970-1980, more than 50% of the increase in global NO_x emissions originated in Asia with only 9% and 18% coming from North America and Europe respectively. A case study referenced to 1990 by World Energy Council [1993] projects that by the year 2020 there will be a 13% increase in global surface NO_x emission accompanied by a net decline of 36% from the North and an increase of 80% from the South. For South Asian and Pacific regions, the estimated increases in NO_x emissions are 118% and 60% respectively. With reference to 1990, EPA [1995] and EMEP [1997] expect significant decreases in surface emissions of NO_x and CO from the North.

The global tropospheric photochemical tendency is quite inhomogeneous in space and time. Redistribution of surface sources of NO_x, CH₄ and CO emissions will lead to variable trends in surface O₃ mixing ratios. For example, slowdown in its growth rate over NH high latitudes and Europe [Oltmans et al., 1998], and increase in its growth from Asian regions [Lee et al., 1998]. Similar variable trends are also expected in OH radical concentration. Much of the effort of photochemical modelers has been devoted to study the effects of uniformly increased or decreased global/regional emissions of air pollutants on the chemical balance of the troposphere. In this paper, we study the changes in tropospheric chemistry implied by the geographical redistribution in surface emissions of NO_x, CH₄ and CO with their global source strengths fixed.

Simulations and Model Description

For the present study, we consider 25% decreases in surface emissions of NO_x, CH₄ and CO from the developed countries (considered to be located between 35°N-75°N) and

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simultaneous increases in their emissions from the NH developing countries (considered to be located between 35°N-5°S) by the same absolute amounts for which they were reduced from the developed countries. This scenario allows geographical redistribution in surface emissions of NO_x, CH₄ and CO but leaves their global source strengths unchanged. To study the non-linearity in the chemical system induced by this scenario, multiple variations were introduced in the surface emissions of NO_x, CH₄ and CO. To examine the relative partial effects of adjustment in individual sources of NO_x, CH₄ and CO on the tropospheric oxidizing capacity, individual model simulations altered emissions for each of these species separately. In the future, global surface emissions of NO_x, CH₄ and CO may in fact increase. In the present study, this possibility is set aside. Moreover, we realize that the future changes in geographical emission patterns may or may not be the same for all three air pollutants.

All calculations were made using a modified version of the Oslo two-dimensional (altitude/latitude) global tropospheric photochemical model. This model has a horizontal resolution of 10° and extends from surface to 24.5 km with a vertical spacing of 0.5 km. Both advective and diffusive coefficients are derived from the GFDL GCM [Plumb and Mahlman, 1987]. The treatment for convective transport is based on the scheme of Langner *et al.* [1990]. The transport features have been tested with observed distributions and trends of CFC-11, CFC-12 and ⁸⁵Kr. The photochemical scheme accounts for complete oxidation of most of the hydrocarbons containing up to 3 carbon atoms. In the model, the annual surface source strengths for NO_x, CH₄ and CO are 37 Tg of NO_x as N, 500 Tg of CH₄ and 1280 Tg of CO respectively. North to south ratios of their corresponding surface emissions are 4.2, 2.1 and 2.6 respectively. More details of their distinct latitudinal emission distributions are given in Gupta [1996] and Gupta *et al.* [1998]. For the reference case, the model latitude belts (75°N-35°N) and (35°N-5°S) account for 149 Tg of CH₄, 484 Tg of CO, 20 Tg of NO_x as N and 207 Tg of CH₄, 491 Tg of CO, 12 Tg of NO_x as N respectively. For 25% adjustment, about 5, 37 and 121 Tg of surface emissions of NO_x, CH₄ and CO respectively were assumed to have shifted from (75°N-35°N) to (35°N-5°S). The model was typically allowed to simulate for at least four decades so as to attain steady state concentrations of CH₄ throughout the domain. In this paper, our analysis will concentrate on relative chemical changes brought by the redistribution of surface emissions of NO_x, CH₄ and CO. For steady state distributions of reactive species corresponding to the reference simulation, see Gupta [1996]. Simulations corresponding to the 25% shift of emissions of all three species combined and NO_x, CH₄ and CO individually are termed as *ALL*, *NOX*, *CH4* and *CO* respectively.

Results and Discussion

Emissions of the air pollutants such as NO_x, hydrocarbons and CO from the developing countries, which are mostly confined to tropical latitudes, very effectively perturb the global atmospheric chemical balance for two primary reasons. First, tropical regions experience year-round intensive photochemical activity. Also, at low latitudes, sub-scale convective events draw polluted air out of the planetary boundary layer and redistribute it in the photochemically efficient free troposphere. Along with non-linearities in the production of tropospheric ozone [Liu *et al.*, 1987] and increasing tropical emis-

sions, these two factors significantly affect the distributions of chemically reactive trace gases and oxidizing capacity of the global troposphere. Gupta [1996] has described some photochemical effects of increased tropical emissions of NO_x, hydrocarbons and CO on the tropospheric chemical balance.

Our calculations show that redistribution of emissions of NO_x, CH₄ and CO has significant effects on the tropospheric oxidizing capacity and distributions of chemically active trace species. For simulations *ALL* and *NOX*, Figure 1 shows the calculated percent changes in OH concentration for July. For case *ALL*, a combined decrease in sinks and sources of OH radicals from mid-latitude regions to the tropics led to an overall increase (ranging from 1.4-4.2% at various latitudes and altitudes) in OH concentrations throughout the mid-latitude NH troposphere. This net increase in OH concentration at mid-latitudes is caused mainly by the decrease in emissions of CH₄ and CO and is counteracted somewhat by the decrease in NO_x surface emissions. For case *NOX*, reduction in NO_x emissions alone led to a decrease in OH concentration by 2.6% and 1.6% in the lower and middle NH troposphere respectively. This is due primarily to decrease in O₃ concentration (see Figure 2). For tropical regions of the NH, an increase in OH concentration in the range of 3-8% is calculated. For *ALL*, despite an increase in emissions of CO and CH₄, an increase in OH is obtained because of enhanced NO_x emissions which are relatively more efficient in producing O₃, and hence OH radicals. A comparison of the increase in OH concentration at NH tropics shows that increased emissions of NO_x contributed the most to the calculated increase in OH concentration, with a small opposing effect from increased emissions of CO and CH₄.

On the annual average, changes in global OH concentration weighted by CH₄ mixing ratio and rate of reaction for CH₄+OH [Prather and Spivakovsky, 1990], calculated for *ALL*, *NOX* and *CO* cases are +3.6%, +3.9% and -0.2% respectively. For a scenario with only an increase in surface NO_x emissions by 5 Tg as N (which is 25% of surface NO_x emissions between 35°N-75°N) between 35°N and 5°S, the calculated increase in global annually averaged OH concentration is 5.2%, indicating that the largest factor for increase in global OH concentration for case *NOX* is increase in tropical NO_x surface emissions. For *NOX* and *CO*, the calculated percent changes in global annual averaged OH concentration

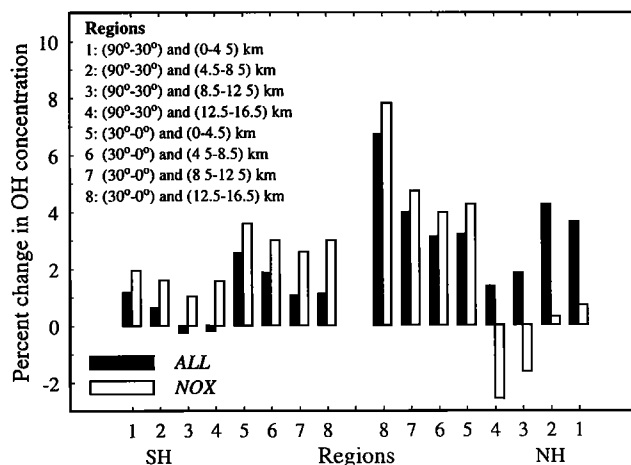


Figure 1. Calculated percent changes in regional OH concentration for July simulated for scenarios *ALL* and *NOX*.

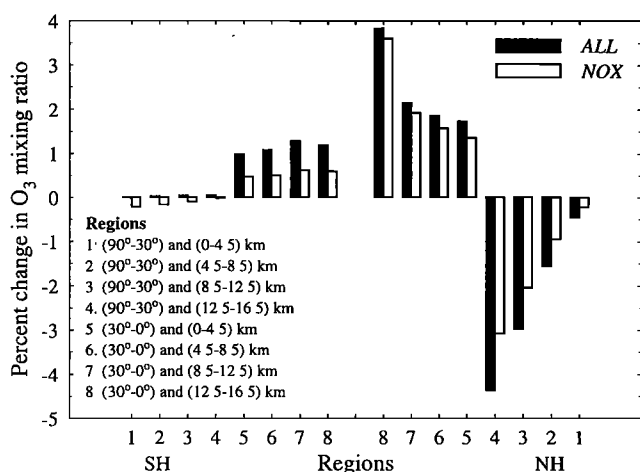


Figure 2. Calculated percent changes in regional O₃ mixing ratios for July simulated for scenarios ALL and NOX.

per Tg adjustment in surface source are +0.78% and $-1.6 \times 10^{-3}\%$ respectively. Due to nonlinear photochemical interactions, the calculated changes in annually averaged global OH concentration per Tg shift in NO_x emission are +0.86% and +0.68% respectively for 10% and 50% shifts in surface emissions of NO_x. For scenario CH₄, no net change in global annually averaged OH concentration is calculated.

Figure 2 shows the calculated percent changes in O₃ mixing ratio for cases ALL and NOX in July. Because of decreased emissions of O₃ precursors, a decrease in O₃ mixing ratios ranging from 0.2% to 4.4% is calculated for all mid- and high latitude regions of the NH troposphere. The largest contribution is from the decreased NO_x surface emissions. On the other hand, an increase in O₃ is calculated in the tropics of both the hemispheres. This is caused by enhanced oxidation of CH₄ and CO in the presence of increased NO_x concentrations. For the NH tropics, the increase ranges from 1.3% to 3.8% and is localized in the lower troposphere. Over the entire tropical domain, most of the calculated increase in O₃ can be attributed to increased tropical NO_x surface emissions. For cases ALL and NOX, the annual average increase in surface O₃ mixing ratios for the NH tropics lies between 5–6% (Figure

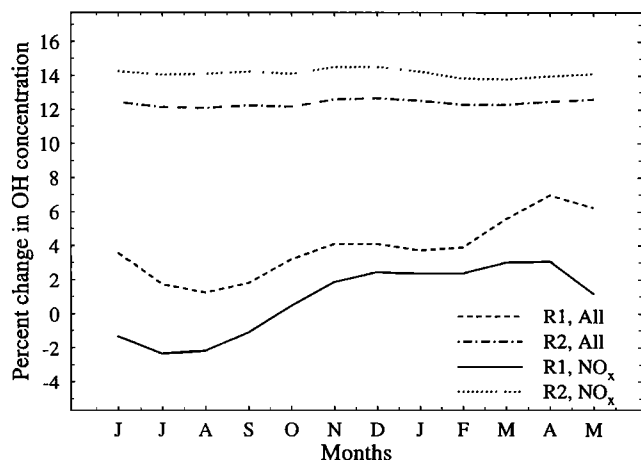


Figure 3. Monthly variations in calculated percent changes in regional surface O₃ mixing ratios simulated for scenarios ALL and NOX. R1 and R2 stand for latitude belts (90°N–30°N) and (30°N–equator).

Table I: Calculated Percent Changes in Annually Averaged CH₄ Surface Mixing Ratios Brought by Latitudinal Redistribution of Surface Sources of NO_x, CH₄ and CO

Case	Model Latitude Region					
	MR1	MR2	R1	R2	R3	R4
ALL	-4.66	-3.04	-4.39	-3.03	-3.00	-2.98
NOX	-3.43	-3.70	-3.45	-3.71	-3.76	-3.75
CO	+0.05	+0.13	+0.05	+0.13	+0.17	+0.17
CH4	-1.13	+0.59	-0.94	+0.62	+0.69	+0.64

Note: MR1, MR2, R1, R2, R3 and R4 correspond to latitudes (75°N–35°N), (35°N–5°S), (90°N–30°N), (30°N–0°), (0°–30°S) and (30°S–90°S) respectively.

3). A variable change in surface O₃ mixing ratio is obtained for mid- and high- latitude NH regions. For case NOX, the surface O₃ mixing ratio decreases by 0.4–3.4% for the months of June–November. At other times of the year, an increase of 0.5% to 2.1% is calculated for surface O₃ mixing ratios despite decreased surface NO_x emissions. These seasonally dependent changes in surface O₃ mixing ratio are partially governed by the extent of O₃ losses by NO forming NO₂.

The changes in OH and O₃ distributions are expected to alter the steady state lifetime, mixing ratio and interhemispheric gradient of CH₄. For cases ALL and NOX, the decreases in CH₄ time constants are 3.3% and 3.7% respectively. Annually averaged surface mixing ratios of CH₄ for the region (75°N–35°N) changed by -4.7%, -3.4%, +0.05% and -1.1% for ALL, NOX, CO and CH4 respectively (Table I). For the region (35°N–5°S), the values are -3.0%, -3.7%, +0.1% and +0.6%. Table I also displays these changes on semihemispheric basis. The stated changes show that the scenario CO will lead overall to a minor increase in CH₄ surface mixing ratios in the NH. For scenario NOX the effect is more pronounced and have opposite sign. For scenario CH4, the calculated changes in methane surface mixing ratios are variable. About 1% decrease is computed for model region (30°N–90°N) and is partially caused by the decreased emissions there. In other parts of the model, suppression of

Table II: Seasonal Variations of Calculated Changes in Interhemispheric Gradient of CH₄ Surface Mixing Ratios Brought by Latitudinal Redistribution of Surface Sources of NO_x, CH₄ and CO

Season	Percent Change in CH ₄ Interhemispheric Gradient					
	ALL		NOX		CH4	
	R1-R4	R2-R3	R1-R4	R2-R3	R1-R4	R2-R3
JJA	-26.3	-6.1	+0.6	-2.9	-25.4	-2.1
SON	-27.5	-7.7	+1.4	-2.5	-26.7	-3.1
DJF	-21.8	-5.4	-0.1	-1.4	-20.7	-2.7
MAM	-20.8	-4.6	-0.1	-2.1	-19.7	-1.5

Note: Regions R1, R2, R3 and R4 are defined in Table I.

OH radical led to about 0.6% increase in CH₄ surface mixing ratios. Hydroxyl is lowered by the increase in CH₄ emissions in the NH tropics and by enhanced transport of CH₄ to the SH. For simulations *ALL*, *NOX* and *CH4*, Table II displays percent changes in the CH₄ interhemispheric gradient. For simulations *ALL* and *CH4* at all seasons, decreases of more than 19% are calculated in north-south gradient (for 90°N-30°N and 30°S-90°S) of CH₄ surface ratios. The reduced gradient is found to be seasonally dependent with a maximum value for season SON and minimum for MAM. For case *NOX*, the change in this gradient is positive for seasons JJA and SON with magnitudes 0.6% and 1.4%. It is slightly negative for other seasons. The change in north-south gradient for tropical surface mixing ratios is negative for all three scenarios. For *ALL*, decrease in this gradient lies between 4.6-7.7%. These results show that the reduced CH₄ interhemispheric gradient observed by *Dlugokencky et al.*, [1998] could be partly due to geographical redistribution of surface sources of CH₄ and NO_x.

Most intense economic and industrial transitions are currently being experienced in the northern hemispheric Asian and Pacific developing countries. In future, this transition is expected to spread to the so far neglected African continent, resulting in larger tropical emissions to the atmosphere and a more pronounced increase in tropospheric oxidizing capacity. Our present calculations do not consider increases in tropical aircraft NO_x emissions and also the effects of biogenic hydrocarbons which are mostly confined in tropical regions. These two factors could enhance the magnitude of chemical effects described here. Within given latitude band, the longitudinal surface emissions of O₃ precursors, especially NO_x, are inhomogeneous, therefore, three-dimensional chemical models should also simulate the stated emissions scenarios to better quantify the results of this study.

Conclusion

Surface emissions of NO_x, CH₄ and CO are decreasing from developed countries which lie mainly in the latitude belt (75°N-35°N). The opposite is true for the developing countries of the tropics. We have conducted two-dimensional photochemical modeling investigations of the shift in emissions. Assuming constant global source strengths, a shift of 25% of the surface emissions of NO_x, CH₄ and CO from the developed countries led to a decrease in steady state methane lifetime by more than 3%, and also caused a net increase in tropical surface O₃ and variable changes in its concentrations at NH mid-latitudes. These changes are mostly attributable to variations in emissions of NO_x, primarily from the tropics. Further, the interhemispheric gradient of CH₄ concentrations, a variable often used to deduce CH₄ sources, was decreased.

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