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Indirect long-term global radiative cooling from NO_x emissions

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Abstract. Anthropogenic emissions of short-lived, chemically reactive gases, such as NO_x and CO, are known to influence climate by altering the chemistry of the global troposphere and thereby the abundance of the greenhouse gases O_3, CH_4 and the HFCs. This study uses the characteristics of the natural modes of the tropospheric chemical system to decompose the greenhouse effect of NO_x and CO emissions into (i) short-lived modes involving predominantly tropospheric O_3 and (ii) the long-lived mode involving a global coupled CH_4-CO-O_3 perturbation. Combining these two classes of greenhouse perturbations—large, short-lived, regional O_3 increases and smaller, long-lived, global decreases in CH_4 and O_3—we find that most types of anthropogenic NO_x emissions lead to a negative radiative forcing and an overall cooling of the earth.

1. Introduction

The direct radiative forcing (RF) of the greenhouse gases CH_4, O_3 and the HFCs has been the subject of numerous studies, and has been reviewed in recent assessment reports [*Schimel et al.*, 1996; *Granier and Shine*, 1999]. However, the chemical lifetime of many of these species in the troposphere is strongly influenced by short-lived gases with little or no radiative effects in the wavelength range important for climate. The 'indirect' climate effects of these reactive species have been shown to make a significant contribution to the total RF [*Hauglustaine et al.*, 1994; *Johnson and Derwent*, 1996; *Fuglestvedt et al.*, 1996; *Daniel and Solomon*, 1998], but detailed quantification of the effects has proved difficult due in part to the wide range of temporal and spatial scales involved.

This paper presents a new method of quantifying these effects using the natural modes of the tropospheric chemical system. The technique allows the full, time-integrated impacts of emissions changes to be assessed, including the effects of temporal and spatial variations, but without the need for decades-long model runs. We apply the technique to NO_x emissions from a variety of sources, and demonstrate that currently incremental increases in anthropogenic NO_x

emissions have a negative integrated RF. In this case long-term cooling due to decreases in CH_4 and O_3 dominates short-term warming from regional O_3 increases.

2. Excitation of the Primary Mode

The coupling of chemical processes with atmospheric transport leads to the existence of natural modes in the tropospheric system which describe how chemical species co-vary in space and time through the different chemical environments present in the atmosphere. The modes correspond to the eigenstates of the chemical system, and each has a characteristic time-scale corresponding to the respective eigenvalue of the system [*Prather*, 1994, 1996]. In previous work [*Wild and Prather*, 2000], we demonstrated the structure and behaviour of these global chemical modes using a tropospheric Chemical Transport Model (CTM) and showed how the primary natural mode, describing the CH_4-CO-OH coupling, could be excited by regional emissions of short-lived species. This mode, with a decay time about 50% longer than the chemical lifetime of CH_4, dominates the long-term radiative impacts of most tropospheric perturbations.

These studies are performed using the University of California, Irvine (UCI) CTM described in earlier work [*Wild and Prather*, 2000]. The model is run using a single year of meteorological fields from the GISS II' GCM [*Rind and Lerner*, 1996] at $4° \times 5°$ resolution. The CTM resolution is degraded to $8° \times 10°$ to reduce computational costs, and the fields are recycled for the multiple-year runs needed here. The model includes a detailed tropospheric chemistry scheme with hydrocarbon oxidation and the effects of stratospheric chemistry via first-order rates of change above a tracer-diagnosed tropopause. Emissions data for 1985 are used, and are described in the previous study.

The CTM is initialized and run for 10 years with fixed emissions to provide a control. The perturbation runs are initialized likewise and run for 10 years but with emissions from particular sources increased by a factor for the first year and returning to the standard emissions for years 2 to 10. The differences between perturbation and control runs are used to integrate the short-lived perturbations to tropospheric O_3 and CH_4 and to determine the asymptotic behaviour of the perturbation (i.e., the amplitude of the primary mode). Since the primary mode has an exact e-fold
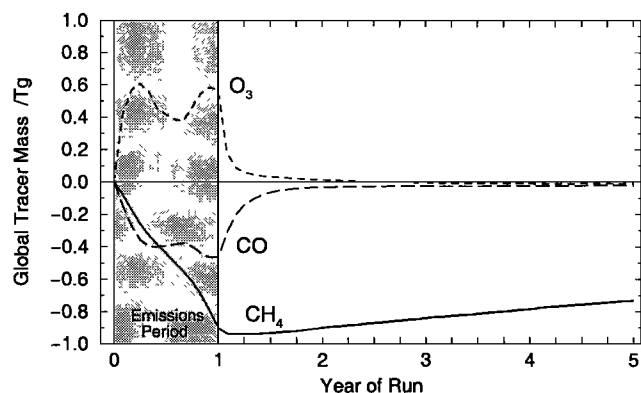
Figure 1. Evolution of the global mass perturbations of O$_3$, CO and CH$_4$ due to the additional emission of 0.5 TgN of global fossil fuel NO for one year.

decay (with annually recycling meteorology), the long-term integration can be analytically evaluated from the product of the amplitude and the e-fold time, saving much computational effort over a steady-state CTM calculation that would take several decades to achieve even 10% accuracy. The incremental NO$_x$ emissions excite the primary mode in a negative sense; CH$_4$ decreases are paralleled by an increase in OH and a decrease in O$_3$, CO, NO$_y$ and peroxy radicals [*Wild and Prather*, 2000]. After the transient, regional effects have decayed, perturbations of all species decay with the timescale of the primary mode, 14.2 years in this model. Although we follow the decay of a pulse, the integral of a single year's emissions is equivalent to the steady state that would accumulate from continuous emissions.

3. Estimates of Radiative Forcing

The direct RF for CH$_4$ and O$_3$ perturbations in the current atmosphere is based on previous studies: +0.37 mW m^{-2} ppb^{-1} for CH$_4$ [*Schimel et al.*, 1996] for a constant mixing ratio perturbation through the troposphere, and +42 mW m^{-2} per Dobson Unit change in column tropospheric O$_3$ [D. Hauglustaine, personal communication, 2000]. The RF per unit O$_3$ depends on region and season [*Fuglestvedt et al.*, 1996] and particularly on altitude [*Lacis et al.*, 1990]. While the short-lived perturbations here are regionally driven, the changes in O$_3$ tend to be dispersed through the mid-troposphere in mid-latitudes and hence may be represented by the mean factor above. The long-lived, primary-mode perturbations are more globally uniform, and hence this simple approach to scaling the RF to the change in total tropospheric O$_3$ provides a suitable estimate. We adopt an uncertainty of 20%, based on the range of forcings from a recent model intercomparison [*Ramaswamy et al.*, 2001], to account for this effect.

We calculate the time-integrated forcing for the primary mode from the amplitude of excitation and the decay time. For a 1.0 Tg perturbation in CH$_4$, 99.3% of the oxidation is accounted for by the primary mode [*Wild and Prather*, 2000], and hence the integrated perturbation is 14 Tg-yr of CH$_4$ with an RF of 1.9 mW m^{-2} yr. The O$_3$ perturbation tied to this excitation of the primary mode is 0.016 Tg-yr of O$_3$ with an integrated RF of about 0.87 mW m^{-2} yr, 45% of the magnitude of the CH$_4$ RF. This long-lived O$_3$ perturbation occurs whenever the primary mode is excited, although

it is often masked initially by large O$_3$ perturbations present in short-lived modes.

4. NO$_x$ perturbation studies

In Figure 1 we show the evolution of the perturbation in global mass of O$_3$, CH$_4$ and CO for a one-year, 0.5 TgN increment in NO$_x$ emissions from global fossil fuel sources. The extra NO$_x$ leads to additional O$_3$ formation in the region of emissions and to increased OH, exciting the primary mode by increasing oxidation of CH$_4$ and CO. Immediately following the period of additional NO$_x$ emissions, the O$_3$ and CO perturbations associated with short-lived modes decay rapidly with e-fold times of 2–3 months, but the CH$_4$ perturbation decays with an e-fold time of 14.2 years. Two years later, the O$_3$ perturbation changes sign, as the global decrease associated with the primary mode dominates the short-term regional increases.

The evolution of the integrated RF due to these changes in O$_3$ and CH$_4$ is shown in Figure 2. While the initial RF driven by O$_3$ formation is positive, long-term cooling due to the primary mode gradually reduces this until, after about 24 years, the integrated RF becomes negative and tends to a total net of −0.54 mW m^{-2}. The net integrated perturbations and RFs due to NO$_x$ emissions from a variety of sources are shown in Table 1. Industrial and fossil fuel sources have been subdivided by region to highlight differences in chemical and meteorological environment; while all perturbations follow a pattern similar to that shown in Figure 1, their magnitudes differ greatly, dependent on the O$_3$ production efficiency of the added NO$_x$ and on the additional OH formation. For aircraft emissions, predominantly in the upper troposphere at northern mid-latitudes, the additional O$_3$ dominates the CH$_4$ removal and leads to net radiative warming. The effects are proportional to, but slightly lower than, those derived in the recent IPCC Aircraft Assessment [*Penner et al.*, 1999] (−15 mW m^{-2} for CH$_4$, +24 mW m^{-2} for O$_3$). For lightning emissions, principally in tropical regions, the perturbation to OH and hence CH$_4$ is larger per unit NO$_x$ emission and dominates the warming due to O$_3$. This reduces the likelihood of positive climate feedbacks between O$_3$ and lightning [*Toumi et al.*, 1996].

For all surface emissions studied here, the primary mode dominates the RF, with a small net cooling in each case
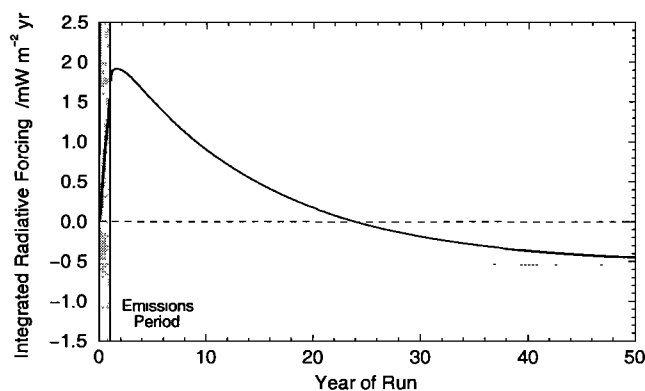

Figure 2. Evolution of the integrated radiative forcing due to changes in CH$_4$ and O$_3$ caused by the additional emission of 0.5 TgN of global fossil fuel NO for one year.

Table 1. Integrated perturbations to CH$_4$ and tropospheric O$_3$ (Tg-yr) and their estimated radiative forcing (RF: W m^{-2} yr) due to a 0.5 TgN increase in emissions for one year.

Perturbation	ΔCH$_4$ Total (Tg-yr)	ΔO$_3$ Total (Tg-yr)	ΔO$_3$ (Tg-yr)[a] Short-lived Modes	Primary Mode	R.F. (mW m^{-2} yr) ΔCH$_4$	ΔO$_3$	Net
Aircraft NO	−67.1	3.79	4.91	−1.11	−9.1	14.6	5.4
Lightning NO	−87.8	2.37	3.83	−1.46	−11.9	9.1	−2.8
Biomass Burning NO	−24.1	0.53	0.93	−0.40	−3.3	2.0	−1.2
Industrial NO	−14.2	0.36	0.59	−0.23	−1.9	1.4	−0.5
— Europe	−5.2	0.08	0.17	−0.09	−0.7	0.3	−0.4
— N.America	−8.9	0.20	0.34	−0.15	−1.2	0.8	−0.4
— E.Asia	−15.4	0.50	0.76	−0.26	−2.1	1.9	−0.2
— Tropics	−39.6	1.26	1.92	−0.66	−5.4	4.9	−0.5
Industrial NO and CO[b]	−6.8	0.68	0.79	−0.11	−0.9	2.6	1.7

[a]The O$_3$ perturbation is split into contributions from the short-lived modes and the long-lived primary mode.
[b]Includes also CO emissions (10 Tg CO for one year).

For CH$_4$, the long-term integrated RF per increment in annual NO$_x$ emissions ranges from −1.4 to −10.8 mW m^{-2} per TgN/yr, similar to the −5 mW m^{-2} found by *Fuglestvedt et al.*, [1996] from step increases in continuous emissions. For O$_3$, the long-term integrated RF ranges from +0.6 to +9.7 mW m^{-2} per TgN/yr. Previous 2-D model studies have suggested +3.1 [*Fuglestvedt et al.*, 1996] and +1.4 to +6.4 mW m^{-2} [*Johnson and Derwent*, 1996], with strong regional variations. [*Fuglestvedt et al.*, 1999] find CH$_4$ and O$_3$ RF of −5.0 and +3.5 mW m^{-2} per TgN/yr, respectively, from US emissions. We find lower but proportional forcings of −2.4 and +1.6 mW m^{-2} for fossil fuel emissions over a similar region. The relationship between the transient response in O$_3$ associated with short-lived modes and the long-term perturbation in CH$_4$ associated with the primary mode is shown in Figure 3. With the exception of aircraft emissions (open circle), all scenarios for NO$_x$ increases fall below the zero-forcing line, indicating a net negative integrated forcing.
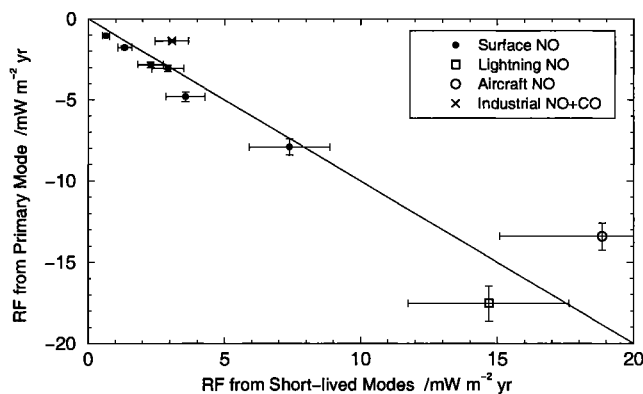

Figure 3. A scatter plot of the integrated radiative forcing (RF) from CH$_4$ and O$_3$ in the primary mode against that of O$_3$ in short-lived modes for model runs with additional one-year emissions of 0.5 TgN from a variety of sources (see Table 1). Above the zero-forcing line, the warming effects of short-term O$_3$ increases dominate; below, radiative cooling due to the primary mode dominates. Error bars of 20% are indicated for the RF from O$_3$.

In contrast to NO$_x$, emissions of CO reduce OH concentrations and therefore excite the primary mode in a positive sense. As NO$_x$ and CO have similar combustion sources, we consider the impact of coupled emissions by pairing the 0.5 TgN/yr increase in global industrial NO$_x$ sources with a proportional increase in CO emissions (10 TgCO/yr). The primary mode is still excited in a negative sense but with about half the amplitude of that due to increased NO$_x$ emissions alone. This highlights the greater sensitivity of OH to NO$_x$ rather than to CO emissions near the surface. Additional O$_3$ formation attributable to CO emissions leads to about a third more O$_3$ in the short-lived modes, and drives the net RF positive for this coupled emission (shown with a cross in Figure 3).

5. Conclusions

The identification of RF caused by emission of short-lived, chemically reactive gases with both short-lived and long-lived chemical modes is extremely useful. This separation of the problem can greatly reduce the computational effort for tropospheric chemical modeling, enabling a wide range of sensitivity studies such as the examples here. Further, it provides both insight and a clear quantitative separation of RF that is regional (short-lived modes) from that which is global (the long-lived CH$_4$-CO-O$_3$ mode). Since these two different patterns of radiative forcing may not cancel, even with opposite signs, it may become important to diagnose them separately.

Anthropogenic surface emissions of NO$_x$ produce a net RF, due to changes in O$_3$ and CH$_4$, that is consistently negative but can vary by an order of magnitude depending on region. Combined industrial emissions of CO and NO$_x$, however, yield a positive RF. Thus decisions to control global tropospheric O$_3$ and hence greenhouse warming by cutting NO$_x$ emissions alone would produce the opposite effect when the long-term, global changes to both CH$_4$ and O$_3$ are considered.

This study has some clear inadequacies. For one, the treatment of urban emissions in the CTM does not include the effects of non-linearities in small-scale plumes where NO$_x$ concentrations may be sufficiently high to suppress O$_3$ production or to remove NO$_x$ rapidly. It is not clear in these

cases that the OH effect is also proportionally suppressed. Uncertainties such as these can be evaluated by comparing the global CTM with carefully designed, high-resolution regional models. Second, the inhomogeneity of the short-term O$_3$ increases adds uncertainty in estimating its RF that is estimated here to be about $\pm 20\%$. Also, we have assumed that RF is additive in terms of climate impact and hence that tropospheric O$_3$ heating cancels CH$_4$ cooling. These latter assumptions can only be tested in a climate model.

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