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
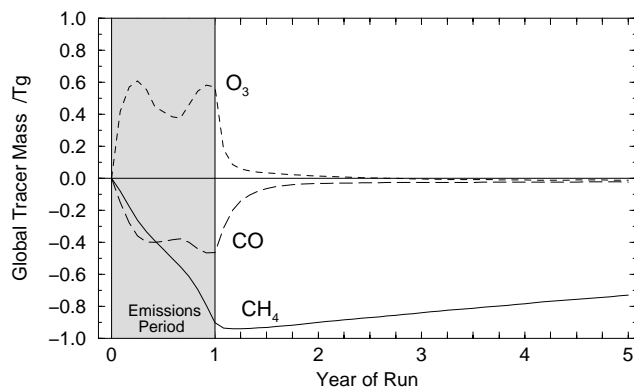
Figure 1. Evolution of the global mass perturbations of O$_3$, CO and CH$_4$ due to the additional emission of 0.5 TgN of global fossil fuel NO for one year.

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].

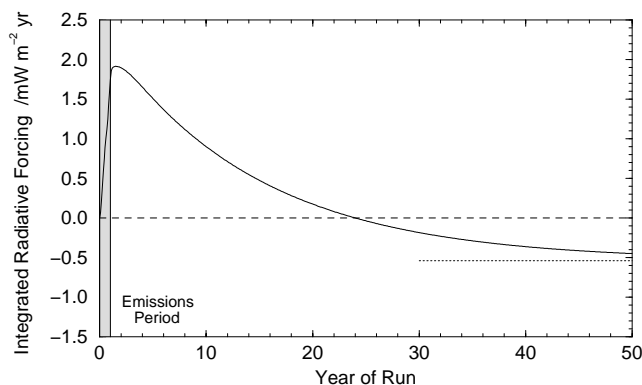For all surface emissions studied here, the primary mode dominates the RF, with a small net cooling in each case.

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.

	ΔCH$_4$	ΔO$_3$	ΔO$_3$ (Tg-yr)[a]		R.F. (mW m^{-2} yr)		
Perturbation	Total (Tg-yr)	Total (Tg-yr)	Short-lived Modes	Primary Mode	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.
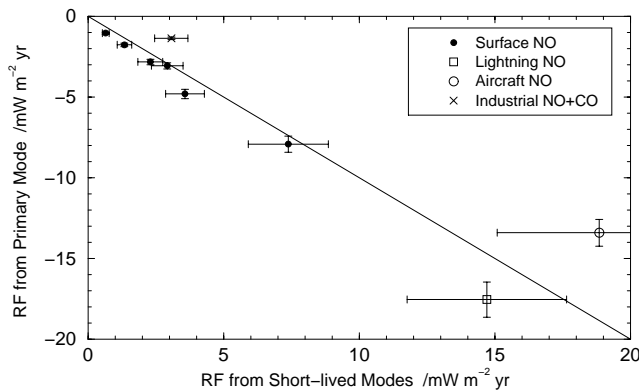

Figure 3. A scatter plot of the integrated radiative forcing (RF) from CH$_4$ and O$_3$ in the primary mode against that of O$_3$ in short-lived modes for model runs with additional one-year emissions of 0.5 TgN from a variety of sources (see Table 1). Above the zero-forcing line, the warming effects 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 $\pm20\%$. 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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