

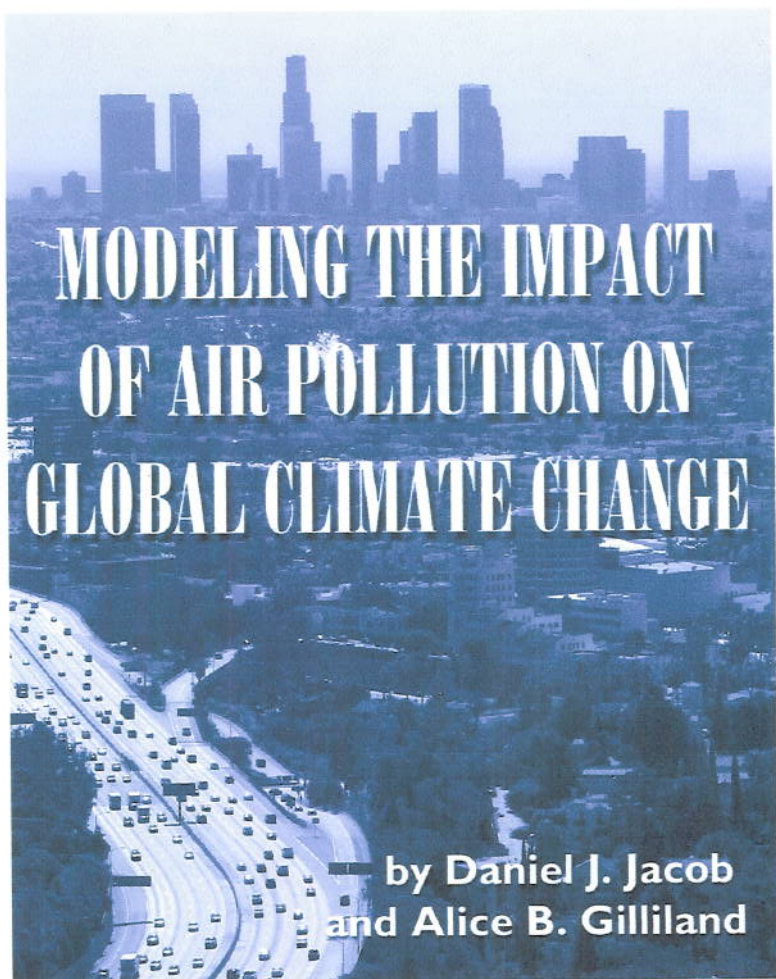
Tropospheric ozone (O₃) and aerosols have major effects on climate and are the two air pollutants of most concern in the developed world. O₃ is a major greenhouse gas (GHG) and light-absorbing aerosols such as black carbon (BC) also contribute to global warming. In contrast, light-scattering aerosols such as sulfate have a cooling effect. Aerosols further affect climate by serving as cloud condensation nuclei (CCN) and modifying the radiative and precipitation properties of clouds. In this article, we review current understanding of the climatic effects of air pollutants and examine the implications for U.S. emissions control strategies directed at air quality.

The climate of the Earth is determined by input of energy from the Sun as solar radiation, loss of energy to space as terrestrial infrared radiation, and internal processes redistributing energy within the Earth system. Our fundamental understanding of global climate is built on general circulation models (GCMs) that solve the equations describing the transport of momentum, energy, and water in the atmosphere and surface reservoirs (i.e., oceans and land). The main uncertainties in these models relate to the simulation of processes internal to the Earth system, such as clouds, land-atmosphere exchange, and ocean-atmosphere coupling. The global energy balance at the top of the atmosphere between incoming solar radiation and outgoing terrestrial radiation is understood more thoroughly. When this energy balance is disrupted, the climate responds by warming or cooling. Such an imbalance is called radiative forcing.¹ Remarkably, a common result in all GCMs is that the ultimate (equilibrium) global change in surface temperature (ΔT_s), resulting from a perturbation proportional to the initial global radiative forcing (ΔF) from that perturbation:

$$\Delta T_s = \lambda \Delta F \quad (1)$$

where λ is the climate sensitivity parameter and varies from 0.3 to 1.4 K m² W⁻¹ depending on the GCM. All GCMs agree that eq 1 is correct to within 30% for a wide range of different forcings, even though the GCMs differ considerably in their estimates for λ , reflecting uncertainty in the simulation of internal climate processes.² The relative ease of radiative forcing calculations, combined with the general reliability of eq 1, has made radiative forcing the standard metric for research and policy directed at climate change.³ To the best of our current understanding, radiative forcings from different agents can be added or subtracted following eq 1 to assess their overall combined impacts on climate change.

Figure 1 illustrates the radiative forcing of climate since pre-industrial times, separating the contributions from different forcing agents.⁴ The best estimates for forcing by BC (+0.8 W m⁻²) and O₃ (+0.5 W m⁻²) are comparable in magnitude to carbon dioxide (CO₂; +1.4 W m⁻²). Anthropogenic aerosols other than BC have negative radiative forcings, with



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an estimated total of -2.3 W m⁻², but with large uncertainties.⁴ Methane (CH₄), another large radiative forcing agent (+0.7 W m⁻²), is a precursor for background tropospheric O₃ and has received recent attention as a lever for improving O₃ air quality.⁵⁻⁷ Thus, we see that air quality management decisions could have a large impact on climate change. Note that O₃ and aerosols are inhomogeneously distributed because of their short lifetimes, and the resulting regional radiative forcings over polluted regions can be considerably larger than the global radiative forcings presented in Figure 1. The consequences of these regional forcings are discussed below.

CLIMATE EFFECTS OF AEROSOLS

Aerosols affect climate by scattering and absorbing solar radiation; this is called the direct radiative effect. In addition, they affect the formation and precipitation tendency of clouds; these indirect effects encompass a number of processes (see Table 1). BC is an efficient absorber of solar radiation and, thus, has a positive radiative forcing (Figure 1).

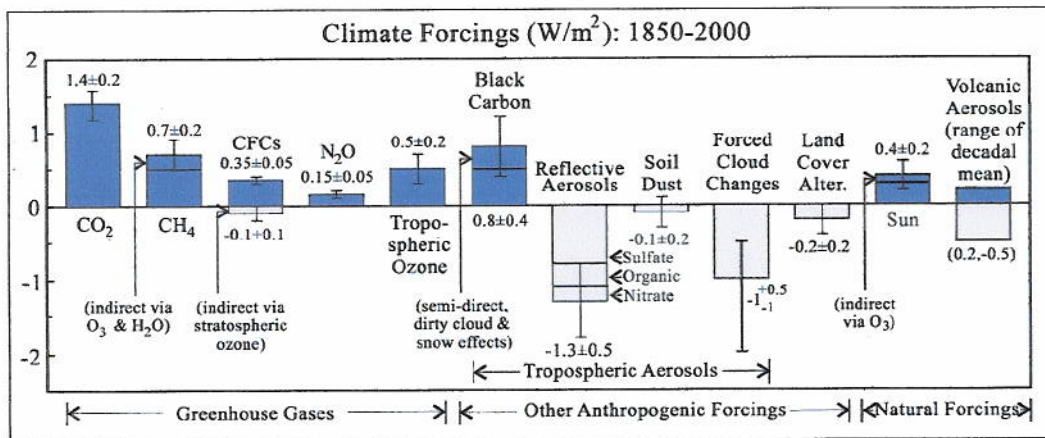


Figure 1. Radiative forcings since 1850 due to changes in GHGs, aerosols (including indirect effects; e.g., "forced cloud changes"), land use, solar activity, and volcanoes. From Hansen and Sato.⁴

Other aerosols (e.g., sulfate, nitrate, organics) are weak absorbers, but efficient scatterers of solar radiation, and the resulting reflection of radiation back to space causes negative radiative forcing (i.e., a cooling effect). The indirect effects of aerosols can cause either cooling or warming (Table 1), though the overall effect is expected to be cooling.

Calculations of aerosol radiative forcings generally rely on global three-dimensional aerosol simulations using GCMs or chemical transport models (CTMs), the latter being driven by archived meteorological variables. These models attempt to include the best understanding of aerosol processes, as allowed by computational tractability. Radiative forcing since pre-industrial times is calculated from the difference between a present-day simulation and a simulation including no

The indirect effects of aerosols can cause either cooling or warming, though the overall effect is expected to be cooling.

anthropogenic sources. Recent studies using different GCMs and CTMs yield best estimates of overall aerosol radiative forcing in the range of -1 to -2.5 W m⁻², with extreme estimates ranging from 0 to -4.5 W m⁻².⁸ The principal anthropogenic aerosol forcing agent is sulfate because of its abundance, hygroscopicity, and nucleation properties (the latter being critical for the first and second indirect radiative effects of Table 1). Aerosol nitrate and organics are less important, although there are large uncertainties relating to nitrate partitioning in the aerosol phase and the anthropogenic sources of organic particles.

An alternate approach for estimating the overall radiative forcing by anthropogenic aerosols has been to use inverse models, in which observed trends of climate variables (e.g., temperature, ocean heat content) are compared to GCM-simulated values for these variables as a function of different

GCM state variables, including aerosol forcing, the climate sensitivity parameter, and ocean turnover. Fitting the GCM simulation to the observed climate variables, using standard optimal estimation methods, then yields best estimates of the state variables as constrained by both observations and our understanding of processes.⁹ Simply stated, the inverse approach involves determining

what compensating aerosol forcing is necessary to reconcile observed climate trends with our knowledge of greenhouse forcing. Different inverse studies conducted over the past five years yield best estimates of aerosol forcing in the range of -0.8 to -1.4 W m⁻².⁸ The inverse models show a narrower range of uncertainty than the process-based estimates, but this could reflect inadequate error characterization in the optimal estimation process.⁸

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Table 1. Overview of the different aerosol indirect effects.³

Effect	Cloud Type	Description	Sign of the Radiative Forcing
First indirect aerosol effect (cloud albedo or Twomey effect)	All clouds	For the same cloud water or ice content more but smaller cloud particles reflect more solar radiation	Negative
Second indirect aerosol effect (cloud lifetime or Albrecht effect)	Warm clouds	Smaller cloud droplets decrease the precipitation efficiency thereby prolonging cloud lifetime	Negative
Semi-direct effect	Warm clouds	Absorption of solar radiation by soot leads to an evaporation of cloud droplets	Positive
Glaciation indirect effect	Mixed-phase clouds	An increase in ice nuclei increases the precipitation efficiency	Positive
Thermodynamic effect	Mixed-phase clouds	Smaller cloud droplets inhibit freezing causing supercooled droplets to extend to colder temperatures	Unknown
Surface energy budget effect	All clouds	The aerosol induced increase in cloud optical thickness decreases the amount of solar radiation reaching the surface, changing the surface energy budget	Negative

Radiative forcing by aerosols is highly variable in space and time, unlike the forcing by long-lived GHGs; and the global radiative forcing metric in eq 1 does not capture the effect of this variability on regional climate. Aerosol forcing is largely confined to the Northern Hemisphere, and differences in observed temperature trends between the two hemispheres have in fact been used to constrain aerosol radiative forcing in inverse model studies.⁹ Regional aerosol forcing over populated continents can exceed -10 W m^{-2} and can swamp the effect of GHGs, but such regional forcing is difficult to interpret quantitatively. Equation 1 cannot be applied regionally because the effect of radiative forcing is compensated to some extent by lateral transport of heat. One GCM study suggests that anthropogenic carbonaceous aerosols increase annual mean temperatures in the United States by 0.5–2.5 K.¹⁰ Another GCM study suggests that BC aerosol emissions in Asia are responsible for observed trends in the summer monsoon over the past decades.¹¹ Large ensembles of GCM simulations are needed to establish confidence in the robustness of such results.³ Improved predictions will ultimately require high-resolution regional climate models (RCMs) nested within GCMs and accounting for the effects of aerosols on regional energy and precipitation budgets.

CLIMATE EFFECTS OF TROPOSPHERIC OZONE

Tropospheric O_3 is the third most important anthropogenic GHG after CO_2 and CH_4 . As for GHGs in general, the radiative forcing is driven by O_3 in the middle and upper troposphere where the temperature contrast with the surface is large. O_3 in surface air has a negligible greenhouse effect because it re-emits radiation at the same temperature as the surface. The climate impact of O_3 pollution thus depends critically on the venting of anthropogenic O_3 and its precursors out of the boundary layer to high altitudes. Models and observations find that this venting is highly efficient, resulting in pervasive anthropogenic influence on tropospheric O_3 over the scale of the Northern Hemisphere.^{2,12,13}

Estimates of radiative forcing by O_3 since pre-industrial times have generally relied on GCM/CTM simulations of tropospheric chemistry, where the anthropogenic O_3 enhancement is calculated as the difference between a present-day simulation and one with no anthropogenic emissions. An intercomparison of nine such GCMs and CTMs² found

a high degree of consistency across models, with increases in the global tropospheric burden of 30–50% since pre-industrial times, corresponding to global radiative forcings in the range $0.2\text{--}0.5 \text{ W m}^{-2}$. However, these models all overestimate observed surface O_3 concentrations for the 1850–1950 period,^{14,17} suggesting that they underestimate human influence. The historical observations would imply a present-day forcing of 0.8 W m^{-2} , but calibration errors in these observations cannot be excluded.¹⁵ A better understanding of natural tropospheric O_3 is needed.

O_3 has a lifetime on the order of weeks in the middle and upper troposphere. Its radiative forcing is less heterogeneous than that of aerosols, but there are still important hemispheric and seasonal variations. In a GCM study that examined the specific climatic signatures of the rise in tropospheric O_3 since pre-industrial times, O_3 was found to be approximately 30% less effective as a global warming agent than CO_2 , but more effective in the Northern Hemisphere.¹⁸ This study found that the rise in tropospheric O_3 since pre-industrial times increased global mean surface air temperatures by 0.3 K (0.4 K in the Northern Hemisphere, 0.2 K in the Southern Hemisphere), with particularly large warmings (up to 1 K) downwind of the industrial continents.

CLIMATE EFFECTS OF METHANE

CH_4 is the second most important anthropogenic GHG after CO_2 , and it is also a major source of background tropospheric O_3 . An economic analysis⁷ recommends CH_4 emissions controls as a cost-effective approach to meet both air quality and climate stabilization objectives. The main sink for CH_4 is oxidation by the hydroxyl (OH) radical, resulting in a lifetime of approximately 10 years. This lifetime is long enough that CH_4 is globally well mixed. Major anthropogenic sources of CH_4 include livestock, natural gas exploitation, coal mining, and landfills.² Changes in CH_4 can also result from changes in the OH abundance; and, here again, air pollutants are involved. OH increases with increasing



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nitrogen oxides (NO_x) and O_3 , and decreases with increasing carbon monoxide (CO) and volatile organic compounds (VOCs). Global tropospheric chemistry models find little net sensitivity of OH to anthropogenic emissions since pre-industrial times (less than 10%) because increases due to rises in NO_x and O_3 have been compensated by decreases due to rises in CO and CH_4 .^{14,16} However, observational analyses based on long-term time series of halocarbons imply larger variations.² For example, one study finds that global OH concentrations increased by 15% during the 1980s and decreased by 25% during the 1990s.¹⁹ Better understanding is needed of the sensitivity of global OH to trends in emissions of air pollutants as the implications for future CH_4 levels could be large.

PERSPECTIVES FOR THE FUTURE

Future radiative forcing estimates for the 21st Century have been made using global scenarios of future emissions generated from a range of economic story lines.² These projections are considered credible for long-lived GHGs, and they forecast in particular a rise in CH_4 emissions by 10–30% between now and 2030. However, they do not account for the evolution of air quality policies and are therefore less useful for projection of air pollutant emissions. For global BC aerosol emissions, a 10–30% decrease has been projected between now and 2030.²⁰ Over the next decades one may expect substantial decreases in emission from North America and Europe, possibly compensated by increases in the developing world. In the United States, the new Clean Air Interstate

Rule (CAIR) and previous control requirements project that NO_x and sulfur dioxide (SO_2) emissions should decrease by approximately 40% over the next two decades, and CO and VOC emissions by 20%. As discussed above, CO has important climate implications as a sink for OH radicals.²¹ The projected decreases in VOC and CO emissions should help to mitigate climate change. Decreases in NO_x emissions are thought to be climate-neutral.⁵ Decreases in SO_x emissions, while necessary for improving air quality, would amplify warming trends. No air quality management policies are yet in place in the United States to control CH_4 , though such controls would help to mitigate the warming effect.

As shown in Figure 1, the total radiative forcing from air pollutants over the past 100 years has been of a magnitude comparable to that from CO_2 . As the 21st Century progresses, the overall forcing of climate will become increasingly dominated by CO_2 . This is because of the long lifetime of CO_2 in the atmosphere (~100 years) and the prospect for sustained increases in CO_2 emissions.² However, the real question to be asked is whether air pollution control policies could have a large climate impact over the next few decades by potentially enforcing large decreases in the concentrations of aerosols, O_3 , CO, and CH_4 . Decreasing BC, O_3 , and CH_4 would clearly be beneficial. Decreasing SO_2 would be deleterious, and reliable assessment of this effect requires improved understanding of aerosol radiative forcing. A large negative radiative forcing by aerosols in the present-day atmosphere would imply, in turn, a very large sensitivity of climate to greenhouse forcing, which may have been hidden so far by compensating aerosol effects.⁸ If this were the case, reducing SO_2 emissions could expose us to a very large greenhouse warming. There is a compelling need to reduce SO_2 emissions because of the threat to public health and ecosystems, but the climatic implications need to be assessed. It may be that an aggressive schedule of GHG emission reductions will be required as the compensating effect of SO_2 emissions wanes.

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