

# Interactions between Climate and Air Quality

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*We present two examples of air pollutant contributions to climate forcing. First, oxidation of the potent greenhouse gas methane produces tropospheric ozone, another greenhouse gas and the primary constituent of ground-level smog. Methane emission controls are thus a “win-win” strategy for jointly addressing air quality and climate goals, particularly given the availability of low-cost emission control options. Second is the “win-lose” case of aerosol sulfate, where decreases improve air quality but lead to additional warming due to decreased scattering of solar radiation. We highlight the potential for aerosols to change the hydrologic cycle and key aspects of how climate change may affect air quality, underscoring a need for evaluating chemistry-climate models with observed relationships between meteorology and air pollutants to build confidence in future projections.*

## 1. Introduction

Ground-level smog, detrimental to human health and vegetation, is pervasive in populated world regions. In the United States, over 150 million people live in counties exceeding air quality standards for ozone (O<sub>3</sub>) or particulate matter (aerosols), the two major smog constituents (U.S. EPA, 2008). These air pollutants also influence climate, with tropospheric O<sub>3</sub> the 3<sup>rd</sup> most important greenhouse gas after carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>), and aerosols exerting a net cooling influence (Forster et al., 2007).

The major precursors to O<sub>3</sub> that fuel rapid photochemical build-up of O<sub>3</sub> during regional air pollution episodes are non-methane volatile organic compounds (NMVOC) and nitrogen oxides (NO<sub>x</sub>), whereas the global burden of tropospheric O<sub>3</sub> is most sensitive to NO<sub>x</sub> and CH<sub>4</sub> (e.g., Fiore et al., 2002). As CH<sub>4</sub> and O<sub>3</sub> together are estimated to have contributed nearly half as much radiative forcing from 1750 to 2005 as CO<sub>2</sub> (Forster et al., 2007), controls on CH<sub>4</sub> emissions could help to slow greenhouse warming (Hansen et al., 2000). Since CH<sub>4</sub> oxidation (in the presence of NO<sub>x</sub>) contributes to the formation of tropospheric O<sub>3</sub> (Crutzen, 1973), including in surface air (Fiore et al., 2002), such controls would also de-

crease O<sub>3</sub> pollution. In contrast, decreasing tropospheric O<sub>3</sub> through NO<sub>x</sub> controls is relatively climate-neutral due to opposing influences on O<sub>3</sub> and CH<sub>4</sub> (e.g., Fuglestad et al 1999), and the forcing from pre-industrial to present is small for O<sub>3</sub> precursor emissions of NMVOC, carbon monoxide (CO) and NO<sub>x</sub> compared to CH<sub>4</sub> (Shindell et al., 2005).

Depending on composition, aerosols can heat or cool the atmosphere by absorbing or scattering solar radiation (“direct effects”). For example, the overall impact of absorption by black carbon is atmospheric warming, whereas sulfates cool by scattering solar radiation back to space. By interacting with the hydrologic cycle and changing cloud properties, aerosols also affect transmission of both solar and terrestrial radiation (“indirect effects”). The overall impact of aerosols is believed to be a cooling influence, estimated to offset ~75% of the positive radiative forcing from CO<sub>2</sub> from 1750 to 2005, though large uncertainties surround these estimates (Forster et al., 2007).

Since warm temperatures and stagnant air masses are conducive to O<sub>3</sub> pollution episodes, changes in climate will likely affect air quality. A variety of modeling approaches have been applied to project how local air quality will respond to climate change: sensitivity studies in which individual meteorological parameters are perturbed (e.g., Steiner et al., 2006); using observed historical correlations between meteorological variables and air quality indices to statistically downscale predictions of future meteorology from climate models (e.g., Holloway et al., 2008); and dynamical downscaling, which links a suite of climate and atmospheric chemistry models from global to regional scales (e.g., Hogrefe et al., 2004). In an analysis of studies examining how a warmer climate will influence air pollution at northern mid-latitudes, Jacob and Winner (2009) conclude that projected increases in temperature and stagnation will exacerbate O<sub>3</sub> pollution in urban areas, particularly in the northeastern United States and southern and central Europe, regions where climate models show consistency in predicted meteorological changes (Christenson et al., 2007). Studies of the aerosol response to climate change disagree in sign, reflecting discrepancies in model projections for changes in precipitation frequency and ventilation in many polluted regions (Christenson et al., 2007; Jacob and Winner, 2009 and references therein).

We present two examples of air pollutants influencing climate: (1) CH<sub>4</sub> and O<sub>3</sub>, and (2) sulfate and black carbon aerosols, including potential impacts of aerosols on precipitation. We then briefly review the key processes through which climate is expected to affect air quality, mainly focusing on the more widely studied O<sub>3</sub> response to climate change (Jacob and Winner, 2009). Finally, we suggest steps towards building confidence in model simulations of these processes.

## 2. Air pollutants influence climate: Methane, ozone, and aerosols

Methane is relatively well-mixed in the troposphere, reflecting its lifetime of approximately a decade. In contrast to the O<sub>3</sub> precursors currently regulated to abate O<sub>3</sub> pollution (NO<sub>x</sub>, NMVOC, and CO), the contribution from CH<sub>4</sub> to surface O<sub>3</sub> is fairly uniform globally, though largest in high-NO<sub>x</sub> polluted regions (Fiore et al., 2002; 2008). A multi-model study indicates that this result is robust, with a 20% decrease in global CH<sub>4</sub> abundances yielding roughly a 1.2 ppb decrease over populated regions in the northern hemisphere (Figure 1; Fiore et al., 2009). As CH<sub>4</sub> is not currently regulated for air quality, its contribution to surface O<sub>3</sub>, along with that from foreign (and natural) emissions of the other O<sub>3</sub> precursors, is typically considered part of the “background”. While combined reductions of NO<sub>x</sub>, NMVOC, and CO within a region are more effective than equivalent percentage reductions of CH<sub>4</sub> at decreasing surface O<sub>3</sub> within that same region, “background” surface O<sub>3</sub> responds roughly equivalently to foreign anthropogenic emission reductions of CH<sub>4</sub> and NO<sub>x</sub>+NMVOC+CO (Figure 2).

The availability of low-cost options suggests that CH<sub>4</sub> emission controls are feasible for jointly addressing climate and O<sub>3</sub> air quality goals (West and Fiore, 2005). Full-chemistry transient simulations (2005 to 2030) in the GFDL MOZART-2 global tropospheric chemistry model indicate that cost-effective CH<sub>4</sub> controls (at a marginal cost of ~\$15 per ton CO<sub>2</sub> equivalent) would offset the positive climate forcing from CH<sub>4</sub> and O<sub>3</sub> that would otherwise occur from increases in NO<sub>x</sub> and CH<sub>4</sub> emissions in the baseline CLE (Current Legislation) scenario (Figure 3a; Fiore et al., 2008). Cost-effective controls (scenario B in Figure 3) are sufficient to decrease the incidence of O<sub>3</sub> events above 70 ppb in the model to below the number of occurrences in 2005 over Europe in summer; over the United States, only the simulation where CH<sub>4</sub> is set to pre-industrial levels (CH<sub>4</sub>-700) achieves this result, reflecting differences in regional projections for NO<sub>x</sub> emissions under the baseline scenario (Figure 3b; Fiore et al., 2008). In all cases, however, CH<sub>4</sub> reductions decrease O<sub>3</sub> relative to the 2030 CLE baseline scenario.

Levy et al. (2008) find significant climate impacts by the year 2100 in the GFDL CM2.1 climate model due to decreasing emissions of sulfur dioxide (SO<sub>2</sub>; to 35% of 2000 levels by year 2100), the precursor of sulfate aerosol, and increasing emissions of black carbon (scaled to CO emission projections) according to the A1B “marker” scenario. In the second half of the 21<sup>st</sup> century, these projected changes in emissions of short-lived species contribute substantially to the total predicted surface temperature warming for the full A1B scenario: 0.2°C in the Southern Hemisphere, 0.4°C globally, and 0.6°C in the Northern Hemisphere. We consider only the direct radiative effect of aerosols, which has been shown to add linearly to the radiative effect of greenhouse gases (e.g., Gillett et al., 2004), with similar climate responses to their forcings (Levy et al., 2008). In Figure 4 we present the radiative forcing and surface temperature change in boreal summer between the 2090s and the 2000s due to the changes in emissions of short-lived gas-

es and aerosols. Note that the largest temperature changes occur over the continental United States, Southern Europe and the Mediterranean, which do not coincide with the regions of strongest emission changes and radiative forcing (Southern and Eastern Asia).

The equilibrium thermal and hydrological responses to the total aerosol effects (i.e., direct, semi-direct and indirect effects) are studied in a version of the GFDL AM2.1 atmosphere general circulation model (AGCM) that includes a prognostic treatment of aerosol-cloud interactions (Ming et al., 2007), coupled to a mixed layer ocean model (Ming and Ramaswamy, 2009). The pre-industrial to present-day increases in aerosols lead to a substantial reduction in the global mean surface temperature (1.9 K), with the strongest cooling over the Northern Hemisphere mid- and high latitudes (Figure 5a). This is accompanied by a significant reduction in precipitation north of the equator, and an increase to the south (Figure 5b). The combined response to aerosols and radiatively active gases (i.e., greenhouse gases) deviates considerably from the linear addition of the individual responses when aerosol indirect effects are included. The results indicate that the large shift in tropical precipitation is driven primarily by the spatially non-uniform aerosols.

### 3. Influence of climate change on surface O<sub>3</sub>

Observational analyses indicate that weather strongly modulates ambient surface O<sub>3</sub> levels from day to day, with many techniques developed over the past decades to remove this influence in order to evaluate the success of O<sub>3</sub> abatement strategies (e.g., Porter et al., 2001). The number of high-O<sub>3</sub> events can vary by as much as a factor of 10 from year to year, largely driven by fluctuations in meteorology (Leibensperger et al., 2008). Of all meteorological variables, temperature typically correlates most strongly with high-O<sub>3</sub> events (e.g., Jacob and Winner, 2009). This correlation largely reflects three key processes: ventilation of surface air, with higher temperatures associated with stagnant air (e.g., Jacob et al., 1993); local O<sub>3</sub> production chemistry, in particular the thermal dependence of PAN decomposition (e.g., Sillman and Samson, 1995); temperature-sensitive biogenic emissions, most notably isoprene (e.g., Guenther et al., 2006). Increases in other emissions such as wildfires and air-conditioning use in response to higher temperatures may further amplify the O<sub>3</sub> response. Other meteorological changes in a warmer world (e.g., convective activity, cloud distributions, humidity, mixing depths) might act as a negative feedback (e.g., Jacob and Winner, 2009 and references therein). Spatial variations in NO<sub>x</sub> and isoprene-emitting vegetation will influence the local sensitivity of O<sub>3</sub> to temperature (and other meteorological variables such as humidity), as has been shown to occur within broad U.S. regions (e.g., Lin et al 2001; Camalier et al. 2007) and on more local scales (e.g., Steiner et al., 2006).

To our knowledge, the ability of global chemistry-climate models to reproduce observed correlations between temperature (or any meteorological index) and air quality metrics has not been evaluated. For example, Figure 6 shows observed O<sub>3</sub>-temperature correlations at four U.S. sites, identified as “regionally representative” on the basis of seasonal variations in surface O<sub>3</sub> (Reidmiller et al., 2009). We suggest that the regional variations in slope reflect differences in the processes contributing to the O<sub>3</sub>-temperature relationship and that these types of relationships should be used to evaluate chemistry-climate models. A second test would be to examine how well models capture the relationship between a decreasing frequency of migratory cyclones and increasing high-O<sub>3</sub> events, as determined from a recent observational analysis for the northeastern U.S. (Leibensperger et al., 2008). The decreasing frequency of cyclones in this region is associated with a northward shift in storm tracks (Figure 7), a robust feature across climate models forced with increasing greenhouse gases (Christenson et al., 2007).

*Priorities for future work.* To build confidence in our understanding as represented in chemistry-climate models, observational constraints are crucial, not only for O<sub>3</sub> and temperature, but also for the processes driving their strong correlation. Critical to this effort is the availability of long-term, high quality measurements of relevant species. Complementing routine measurements of O<sub>3</sub>, PM, and weather variables with species such as formaldehyde (a proxy for isoprene), NO<sub>x</sub>, and PAN could help to determine the relative importance of the various climate-driven impacts on air quality. In particular, large uncertainties exist in our understanding of isoprene oxidation chemistry, especially in low-NO<sub>x</sub> regions (e.g., Horowitz et al., 2007).

Additional work is needed to place climate-driven changes in the context of potentially larger changes to air quality induced through trends in land use and anthropogenic emissions, both locally and globally (e.g., Avise et al., 2009; Chen et al., 2009). While global increases in humidity in a warmer world will likely decrease background O<sub>3</sub> in surface air (e.g., Murazaki and Hess, 2006), future increases in global anthropogenic emissions may extend the O<sub>3</sub> season (e.g., Fiore et al., 2002). A lengthening of the pollution season may also occur in a warmer climate independently of global emission changes (e.g., Racherla and Adams, 2006).

**Acknowledgments.** We thank S. Howard and J. Swall (EPA) for providing daily MDA8 O<sub>3</sub> and temperature from the CASTNet sites, and M. Evans (U Leeds), A. Steiner (U Michigan), and C. Wiedinmyer (NCAR) for insightful conversations.

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Question: Katarzyna Juda-Rezler

I would like to ask you for a comment about (future) precipitation role on air quality. In our simulation with coupled RCM-CTM for Poland, we obtained increased precipitation and decreased PM concentration. Could you comment on that?

Comment:

We have not yet analyzed the PM or precipitation response in our study in any detail and we are not sure that the modest summertime increase in PM just shown is statistically significant. We would not have been surprised to find just what you found. However, we do seem to have a slight increase in summertime PM<sub>2.5</sub> and a decrease in precipitation [not shown] over central Europe, though neither looks like it will be statistically significant. We are just beginning to examine the climate behavior of our new chemistry-climate model.

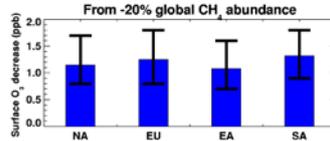
Question: K.Fedra

Does that not suggest we should look into adaptation, robustness, resilience rather than (NBR) impossible prediction?

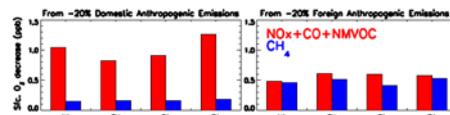
Comment:

The speaker's answer, speaking as a private citizen, is that his bet is on adaptation because of the political and technical complexity of identifying the long-term climate change signal, particularly the regional signal in the presence of short-term natural climate variability. The speaker would also note that, in his opinion, by far the biggest uncertainties are in our ability to project future emissions, rather than our ability to model the chemical-climate system.

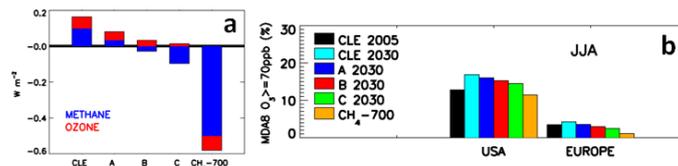
## FIGURES



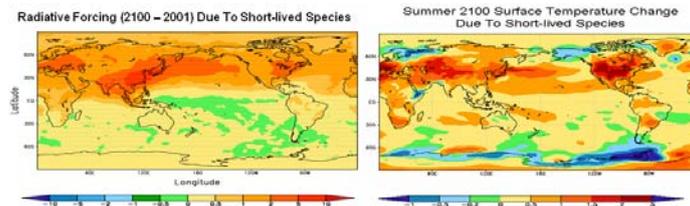
**Figure 1.** Decrease in surface  $O_3$  (ppb) resulting from a 20% decrease in global  $CH_4$  abundances (from 1760 to 1408 ppb) in 18 global or hemispheric chemical transport models over continental-scale source regions: North America (NA; 15-55°N, 60-125°W), Europe (EU; 25-65°N; 10°W-50°E), East Asia (EA; 15-50°N; 95-160°E) and South Asia (SA; 5-35°N; 50-95°E) (Fiore et al., 2009). Model ensemble mean (blue) and range across individual models (black lines) are shown.



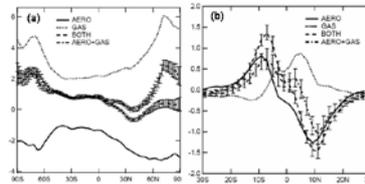
**Figure 2.** Model ensemble surface  $O_3$  decrease (ppb), annually and spatially averaged over the regions in Figure 1 from 20% decreases in anthropogenic emissions of  $NO_x+CO+NMVOC$  (red) versus 20% decreases in anthropogenic  $CH_4$  (blue). Influence of each source region on surface  $O_3$  within the same region (termed “domestic”; left panel). Sum of the  $O_3$  responses to emission changes within the 3 foreign source regions; these components are generally considered to be “background”  $O_3$  (right panel). Adapted from Figure 3 of Fiore et al. (2009).



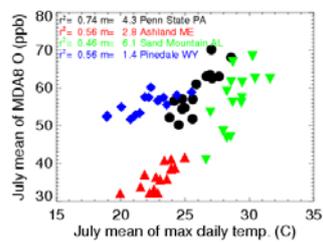
**Figure 3.** (a) Adjusted radiative forcing ( $W m^{-2}$ ) in 2030 versus 2005 due to changes in tropospheric  $CH_4$  (blue) and  $O_3$  (red) calculated with the GFDL AM2 radiative transfer model following Naik et al. (2007), and (b) percentage of model grid-cell days in the GFDL MOZART-2 model with daily maximum 8-hour average (MDA8)  $O_3 \geq 70$  ppb in summer (June-July-August) over the United States (62.5-127.5°W; 24-52°N) and Europe (10°W-50°E; 35-70°N), under the baseline emissions scenario (CLE; global emissions of  $CH_4$ ,  $NO_x$ , CO, and NMVOC change by +29%, +19%, -10% and +3%, respectively) and with decreases in anthropogenic  $CH_4$  emissions by 2030 of 75 (A), 125 (B; cost-effective with available technologies), and 180 (C; requires development of additional control technologies)  $Tg yr^{-1}$ , and in a simulation with pre-industrial  $CH_4$  concentrations (700 ppb). Non- $CH_4$   $O_3$  precursors follow the CLE scenario for 2030 in all simulations. Adapted from Table 4 and Figure 12 of Fiore et al., 2008.



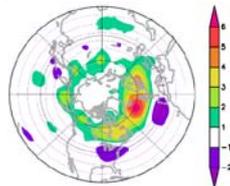
**Figure 4.** Radiative forcing ( $W m^{-2}$ ; left) and surface temperature change ( $^{\circ}C$ ; right) during boreal summer resulting from changes in short-lived species from the 2000s (2001-2010 average) to the 2090s (2091-2100 average) in the GFDL CM2.1 model following an SRES A1B emission scenario (Levy et al., 2008). Figure adapted from Levy et al., 2008.



**Figure 5.** Zonal mean differences (present – pre-industrial) in (a) surface temperature (K) and (b) precipitation ( $\text{mm day}^{-1}$ ) in response to aerosols (AERO), radiatively active gases (GAS) and both (BOTH). For reference is the arithmetic sum of AERO and GAS. Figure is from Ming and Ramaswamy (2009).



**Figure 6.** Regional variability in the relationship between July mean daily maximum 8-hour average (MDA8) O<sub>3</sub> (ppb) vs. July mean daily max temperature (°C) from 1989 to 2004 at selected U.S. CASTNet sites in the northeast (Penn State, PA), far northeast (Ashland, ME), southeast (Sand Mountain, AL), and western (Pinedale, WY) U.S.



**Figure 7.** Poleward shift in northern hemisphere summertime storm tracks in the GFDL CM2.1 climate model (Delworth et al., 2006), diagnosed from differencing the root mean square of 2-10 day bandpass filtered 500 hPa geopotential heights (m) between 100-year simulations corresponding to a pre-industrial control experiment and a  $1\% \text{ yr}^{-1}$  CO<sub>2</sub> increase to doubling.