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Spatial sensitivities of human health risk to intercontinental and high-altitude pollution

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HIGHLIGHTS

- The GEOS-Chem adjoint is used to quantify health risk from intercontinental pollution.
- Sensitivities of human health risk to aircraft pollution are calculated.
- >90% of aircraft emissions-related human PM exposure is due to NO_x.
- Aircraft NO_x creates half of aircraft-attributable surface sulfate.
- 95% of US aviation emissions-related health risk is incurred outside the US.

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ABSTRACT

We perform the first long-term (>1 year) continuous adjoint simulations with a global atmospheric chemistry–transport model focusing on population exposure to fine particulate matter ($PM_{2.5}$) and associated risk of early death. Sensitivities relevant to intercontinental and high-altitude PM pollution are calculated with particular application to aircraft emissions. Specifically, the sensitivities of premature mortality risk in different regions to NO_x, SO_x, CO, VOC and primary $PM_{2.5}$ emissions as a function of location are computed. We apply the resultant sensitivity matrices to aircraft emissions, finding that NO_x emissions are responsible for 93% of population exposure to aircraft-attributable $PM_{2.5}$. Aircraft NO_x accounts for all of aircraft-attributable nitrate exposure (as expected) and 53% of aircraft-attributable sulfate exposure due to the strong "oxidative coupling" between aircraft NO_x emissions and non-aviation SO_2 emissions in terms of sulfate formation. Of the health risk-weighted human $PM_{2.5}$ exposure attributable to aviation, 73% occurs in Asia, followed by 18% in Europe. 95% of the air quality impacts of aircraft emissions in the US are incurred outside the US. We also assess the impact of uncertainty or changes in (non-aviation) ammonia emissions on aviation-attributable $PM_{2.5}$ exposure by calculating second-order sensitivities. We note the potential application of the sensitivity matrices as a rapid policy analysis tool in aviation environmental policy contexts.

1. Introduction

Civil aviation represents the main anthropogenic source of high altitude emissions. While emissions from aircraft at cruise have long been studied in terms of their climate impacts (Penner et al., 1999; Lee et al., 2010), until recently only aircraft landing and takeoff (LTO) emissions, commonly defined as emissions below 3000 feet above ground level, have been considered in terms of their potential to impact surface air quality and human health (Ratliff et al., 2009; Woody et al., 2011; Mahashabde et al., 2011). Although >90% of aircraft NO_x and SO_x emissions occur above 3000 ft (Wilkerson et al., 2010), it has previously been assumed that these emissions do not have an impact on surface air quality (Brasseur et al., 1998). For this reason aviation's impact on air quality has primarily been considered a local and regional issue.

In contrast, recent studies by Barrett et al. (2010, 2012) have found that non-LTO emissions dominate LTO emissions in terms of their impacts on surface air quality and human health. Barrett et al. (2010) focused on the impact of cruise emissions on surface fine particulate matter ($PM_{2.5}$) concentrations, and resultant human exposure and premature mortality risk. The primary mechanisms identified by which cruise emissions impact surface air quality include: (i) the transport of direct PM precursors (SO_2 and NO_x) from

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cruise altitudes to the surface in dry subsiding regions of the atmosphere and (ii) aviation NO_x increasing the oxidative capacity of the atmosphere, which results in an increase in oxidation of non-aviation PM precursors to sulfate and nitrate aerosol. [This has also been explored by Unger et al. (2006) and Leibensperger et al. (2011) in a general intercontinental pollution context.] The studies also found a significant intercontinental component of air pollution associated with aviation emissions, which are deposited in high-speed westerlies at ~10 km altitude. For example, while civil aviation over India and China combined accounts for 10% of global aviation fuel burn, their combined share of global aviation-attributable premature mortalities was calculated to be 35% (Barrett et al., 2010). Pollution associated with aircraft emissions is therefore an intercontinental issue, but the breakdown of regional sources and their intercontinental impacts has not been investigated.

Aside from in an aviation context, intercontinental pollution has been extensively studied (UN ECE, 2010). In terms of understanding the relationship between sources of PM pollution and the resultant exposure, the development of source–receptor (S–R) matrices is of interest, particularly in a policy assessment context (Liu et al., 2009a). Human PM exposure has also been related to premature mortality risk (Liu et al., 2009b) in this context, drawing on an increasing quantitative evidence base associating long-term fine PM exposure with increased risk of cardiopulmonary diseases or lung cancer (Ostro, 2004; Pope and Dockery, 2006; USEPA, 2006; Lewtas, 2007; Pope et al., 2002; Laden et al., 2006). Forward chemistry–transport modeling has been used to create S–R matrices for intercontinental PM transport (Liu et al., 2009a), resultant human exposure and health risk (Liu et al., 2009b), either by perturbing emissions regions in-turn or tagging emissions by region (UN ECE, 2010). Such forward modeling approaches have the advantage of producing many disaggregate outputs (PM concentration fields globally) based on few aggregate inputs (a computationally constrained number of tagged tracers or separate simulations for each aggregate source region). This approach is well matched to cases where the impact of, for example, a specific country's emissions on many other countries is sought. However, in other applications the opposite property is needed. For example, if all contributions (multiple sources) to human PM exposure in one country (i.e. one aggregated receptor) are required, it may be impractical to conduct forward simulations for all possible sources that contribute to PM in the receptor country.

In this paper we develop an adjoint approach to tackling the problem of understanding the relationship between sources and receptors of intercontinental PM air pollution. This approach results in the sensitivity of human PM exposure in selected receptor regions to PM and PM precursor emissions globally. The resultant sensitivity matrices can be multiplied by PM or PM precursor emissions fields to estimate human PM exposure or premature mortality risk. We apply this to the case of global civil aviation emissions to quantify the impact of PM emissions around the world, both on the surface and at high altitudes, on human PM exposure and premature mortality risk in selected regions. The method is also used to elucidate the cross-coupling between PM emissions species – in particular the extent to which aircraft NO_x emissions enhance sulfate formation. The resulting sensitivity matrices can also be used to determine emissions reductions – both by species and spatially – that most effectively reduce human PM exposure, and as a rapid policy assessment tool to assess the health impacts of emissions changes.

2. Methodology

The adjoint method is widely used in atmospheric science for inverse modeling and data assimilation, but less often for sensitivity assessment. We are not aware of a prior long-term (~1 year), global scale continuous sensitivity assessment based on the adjoint method (as distinct from applications to data assimilation). A long-term study with a global domain is especially important for capturing the effects of intercontinental pollution, as exemplified by the case of aviation emissions.

2.1. GEOS-Chem and the GEOS-Chem adjoint

We use GEOS-Chem, a global tropospheric chemical transport model (CTM). The adjoint of GEOS-Chem was developed following the development of its forward model (Henze et al., 2007; Singh et al., 2009), and it has been used to conduct data assimilation and sensitivity studies that relate emissions to atmospheric composition (Kopacz et al., 2011; Walker et al., 2012; Jiang et al., 2011; Henze et al., 2009). These sensitivity studies span a few days to a few weeks, in some cases using the average of multiple week-long sensitivity calculations to approximate yearly average source–receptor relationships (Henze et al., 2012). (This may be due to the relatively local focus of the studies, the computational intensity of the adjoint method, and/or numerical issues resolved in our application of the GEOS-Chem adjoint to long-term simulations.) This study extends the length of a single adjoint simulation to over one-year to capture intercontinental pollution mechanisms.

GEOS-Chem uses assimilated meteorology data from the Goddard Earth Observing System of the NASA Global Modeling and Assimilation Office (Bey et al., 2001; Liu et al., 2001). With the assimilated data, this study uses the standard NO_x–O_x–hydrocarbon–aerosol chemistry mechanism in the model, as originally described in Wang et al. (1998) and since updated. This tropospheric chemistry mechanism includes the gas-phase chemistry of about 90 chemical species. The gas-phase chemistry is solved by Kinetic PreProcessor (KPP) (Damian et al., 2002), and sulfate–nitrate–ammonium thermodynamic equation is calculated by MARS-A, an inorganic aerosol thermodynamic equilibrium module (Park et al., 2004; Binkowski and Roselle, 2003; Zhang et al., 2000). Stratospheric chemistry is modeled by the LINearized OZone model (LINOZ), which implements the first order Taylor expansion of the relationship between ozone mixing ratio, temperature, and overhead ozone column (McLinden et al., 2000). Simulations of GEOS-Chem, including the chemistry and transport relevant to aircraft emissions, i.e. NO_x–O_x–hydrocarbon–aerosol chemistry and intercontinental transport, have been evaluated with networks of observations (Bey et al., 2001; Park et al., 2003, 2004; Fiore et al., 2009; Wu et al., 2009) and most recently in terms of vertical transport in an aircraft emissions context (Barrett et al., 2012). Aircraft emissions from the AEDT 2006 inventory (Wilkerson et al., 2010) were applied.

2.2. Definition of sensitivities

A sensitivity metric contains two parts: the quantity of interest and the source to which sensitivities are calculated. The objective function, J, discussed in this paper is averaged over time and space as

$$J = \frac{1}{\mathrm{PPL}_r V_r T_r} \int_{T_r} \int_{V_r} \mathrm{ppl}(S)\mathrm{pm}(S,t)\mathrm{d}v\mathrm{d}t. \quad (1)$$

This objective function shows a population-weighted exposure to $PM_{2.5}$ concentration. In Eq. (1), pm is the concentration of $PM_{2.5}$ and ppl is the number of people exposed to the $PM_{2.5}$ at (receptor) location S, and time t. The unit of the objective function, J, is $\mu g\ m^{-3}$, PPL_r is the total population in the domain of the objective function,