Intercontinental influence of NOx and CO emissions on particulate matter air quality

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ABSTRACT

Anthropogenic emissions of nitrogen oxides ($NO_x \equiv NO + NO_2$) and carbon monoxide (CO) affect particulate matter (PM) air quality on an intercontinental scale by changing background concentrations of oxidants (OH, ozone, H_2O_2) and thus increasing the oxidation rate of sulfur dioxide (SO_2) to sulfate and NO_x to nitrate. We conduct sensitivity simulations with the GEOS–Chem chemical transport model and find that these intercontinental influences of NO_x and CO emissions on PM can be greater than those from SO_2 emissions (a direct PM precursor). The intercontinental impact of oxidant precursors is greatest in receptor regions with high domestic SO_2, NO_x, and ammonia emissions and hence already high levels of PM. US NO_x and CO emissions increase annual mean PM in northern Europe and eastern China by up to 0.25 μg m^{-3}. The increase in Europe is mostly as sulfate, whereas in China it is mostly as nitrate. East Asian NO_x and CO emissions have a weaker intercontinental influence ($\sim$0.2 μg m^{-3} in northern Europe, $\sim$0.1 μg m^{-3} in the eastern US). These intercontinental effects of NO_x and CO emissions on PM depend in a complex way on the chemical environment of receptor regions. Intercomparison of results from different models would be of great interest.

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1. Introduction

Developed countries worldwide regulate domestic sources of particulate matter (PM) to meet air quality goals designed to protect public health and visibility. These regulations may involve neighboring countries to address transboundary transport. However, PM can also be transported on intercontinental scales with significant implications for air quality in the receptor continent (Park et al., 2006; Liu et al., 2009a). Here we show that this intercontinental influence on PM does not only involve emissions of PM and its direct precursors from the source continent, but also emissions of nitrogen oxides ($NO_x \equiv NO + NO_2$) and CO that affect PM through global perturbations to oxidant concentrations.

There has been much recent interest in quantifying the intercontinental transport of PM pollution (Jaffe et al., 1999; Park et al., 2004; Heald et al., 2006; Chin et al., 2007; Liu et al., 2009b). Intercontinental transport of desert dust and of plumes from large forest fires has long been recognized as a contributor to surface PM (Prospero, 1999; Forster et al., 2001; Husar et al., 2001; Fairlie et al., 2007). Surface, aircraft, and satellite observations have identified episodic transport of anthropogenic PM across the Pacific (Jaffe et al., 1999, 2003; Yu et al., 2008). Global models indicate annual mean sulfate enhancements in US surface air of up to 0.2 μg m^{-3} from anthropogenic Asian SO_2 emissions (Park et al., 2004; Heald et al., 2006; Chin et al., 2007). Direct intercontinental transport of anthropogenic organic and nitrate aerosols appears to be far less important based on both observational and modeling evidence (Heald et al., 2006; Peltier et al., 2008; van Donkelaar et al., 2008).

Sulfate, nitrate, and organic aerosol form in the atmosphere by oxidation of their precursor gases SO_2, NO_x, and volatile organic compounds (VOCs). Oxidant levels affect the rate of aerosol production. Previous global model studies have found that global changes in anthropogenic emissions of oxidant precursors (NO_x, CO, VOCs) affect surface PM concentrations and radiative forcing by perturbing background concentrations of the oxidants OH, H_2O_2, and ozone (Unger et al., 2006; Rae et al., 2007; Kloster et al., 2008; Shindell et al., 2008, 2009). A recent study by Barrett et al. (2010) found that NO_x emissions from aircraft at cruising altitudes enhance surface sulfate PM production by increasing background oxidant levels such that surface sulfate concentrations increase linearly with aircraft NO_x emissions.

Current understanding of intercontinental pollution influence on PM air quality has been assessed by the Task Force on Hemispheric Transport of Air Pollutants (TF-HTAP, 2007) of the United Nations Environmental Program (UNEP). The assessment presents

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global multi-model estimates of intercontinental source–receptor relationships for PM and its precursors. These estimates can be made with either of two methods. In the first, PM or precursors from a given region are "tagged" and tracked as they undergo transport and chemical evolution. This method is applicable only for linear problems. In the second, more general method, sensitivity simulations with perturbed emissions in the source continent are compared to a control simulation. The TF-HTAP (2007) multi-model assessment used the second method but did not investigate the role of oxidant precursor emissions in contributing to intercontinental PM influences. We do so here.

2. Model simulations

We conducted detailed simulations of coupled tropospheric ozone–NO_x–VOC–aerosol chemistry with the GEOS–Chem chemical transport model (version 8-01-01; http://geos-chem.org) driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS)-4. The model has a horizontal resolution of $2°$ latitude $\times 2.5°$ longitude and 30 vertical levels. GEOS–Chem simulates gas-phase oxidant chemistry together with the mass concentrations of the major aerosol types including sulfate–nitrate–ammonium (SNA), black carbon, primary organic carbon, secondary organic carbon, fine and coarse mode sea salt, and dust in four size classes (Park et al., 2003, 2006; Alexander et al., 2005; Fairlie et al., 2007; Liao et al., 2007). Gas-phase and aerosol chemistry are coupled by in-cloud SO_2 oxidation, gas–aerosol thermodynamic partitioning of SNA and secondary organic aerosol (SOA), aerosol effects on photolysis rates (Martin et al., 2003), and heterogeneous chemistry (Jacob, 2000).

The model forms sulfate aerosol from SO_2 through gas-phase oxidation by OH and in-cloud oxidation by H_2O_2 and ozone (Park
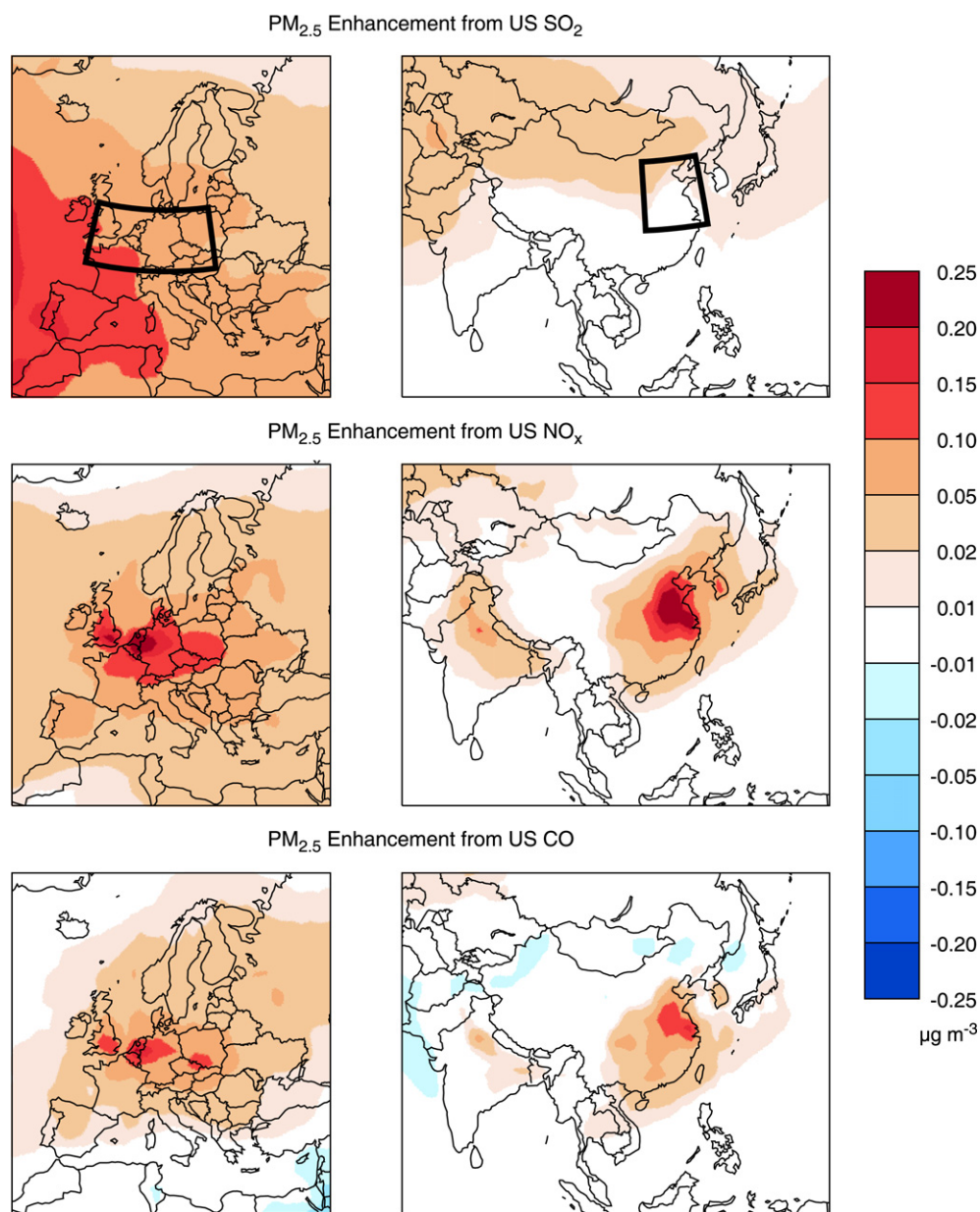
Fig. 1. Annual mean enhancements of surface PM concentrations in Europe and Asia from US anthropogenic emissions of SO_2 (top), NO_x (middle), and CO (bottom). Values are differences between the control GEOS–Chem simulation and a sensitivity simulation with US anthropogenic emissions shut off. Boxes in the top panels outline receptor regions in northern Europe and eastern China for which speciation and seasonality are shown in Fig. 2.

et al., 2004). The MARS-A aerosol thermodynamic equilibrium model is used to calculate SNA aerosol formation (Binkowski and Roselle, 2003). Nitric acid is formed by the gas-phase reaction of NO_2 with OH and ozone, the latter leading to formation of N_2O_5 which hydrolyzes to HNO_3 in aqueous aerosol (Evans and Jacob, 2005). SOA is formed by oxidation of VOCs by ozone and OH, following the work of Chung and Seinfeld (2002), as implemented in GEOS–Chem by Liao et al. (2007).

Successive versions of GEOS–Chem have been extensively evaluated with surface, aircraft, and satellite observations of tropospheric oxidants, aerosols, and related species. Recent worldwide evaluations with satellite data are presented by Zhang et al. (2010) for ozone, Kopacz et al. (2010) for CO, and van Donkelaar et al. (2010) for aerosol. Comparisons to OH and H_2O_2 vertical profiles measured on aircraft missions have been presented by Hudman et al. (2007) and Mao et al. (2010). Air quality relevant evaluations with surface data for ozone and aerosols have been presented for North America and China (Choi et al., 2009; Wang et al., 2009). In addition, a number of GEOS–Chem studies have evaluated the model with observations specifically in the context of intercontinental influences on surface PM (Park et al., 2004; Heald et al., 2006; van Donkelaar et al., 2008) and surface ozone (Fiore et al., 2009; Zhang et al., 2009a). GEOS–Chem results contributed to the TF-HTAP (2007) multi-model assessment were within the ranges of results from other models.

We conducted a control simulation for year 2000 and six sensitivity simulations where we removed individually anthropogenic emissions of CO, NO_x, and SO_2 from the contiguous US and East Asia. Anthropogenic emissions include fuel and industry, not open fires. We define East Asia as the emission inventory domain of Streets et al. (2003), which extends from Pakistan to Japan in the west–east direction and from Indonesia to Mongolia in the south–north direction. Each simulation used meteorological data for 2000–2001. The first year (2000) was used for model initialization and the second year (2001) for analysis. The model does not allow for aerosols or ozone to affect meteorology. Changes in PM concentrations are thus solely due to atmospheric chemistry.

Anthropogenic emissions of NO_x and SO_2 are from the EDGAR 3.2 FT inventory for 2000 (Olivier and Berdowski, 2001). These include 4.8 Tg N a^{-1} and 7.5 Tg S a^{-1} from the contiguous US and 10 Tg N a^{-1} and 27 Tg S a^{-1} from East Asia. US anthropogenic emissions of CO are from the EPA National Emissions Inventory 1999 (NEI99; http://www.epa.gov/ttn/chief/net/1999inventory.html) and amount to 82 Tg CO a^{-1}. East Asian fossil anthropogenic emissions of CO are from Streets et al. (2006), and amount to 250 Tg CO a^{-1}. Ammonia emissions are from Bouwman et al. (1997). Additional source information for the model is available from van Donkelaar et al. (2008).

3. Intercontinental PM enhancements

3.1. Effect of US emissions

Fig. 1 shows the annual mean PM enhancements in Europe and Asia from US anthropogenic sources of SO_2, NO_x, and CO as calculated by GEOS–Chem. PM enhancements are diagnosed as the differences in surface air concentrations of dry sulfate, nitrate, ammonium, and organic aerosol between the control simulation and a simulation with the corresponding emissions shut off. Fig. 2 shows the SNA speciation and seasonality of this enhancement for receptor regions (outlined as boxes in Fig. 1). Intercontinental enhancements in organic aerosol concentrations are small (<5 ng m^{-3}) and will not be discussed further.

The intercontinental effects of SO_2 emissions arise from the direct transport of SO_2 and sulfate, and decrease rapidly with
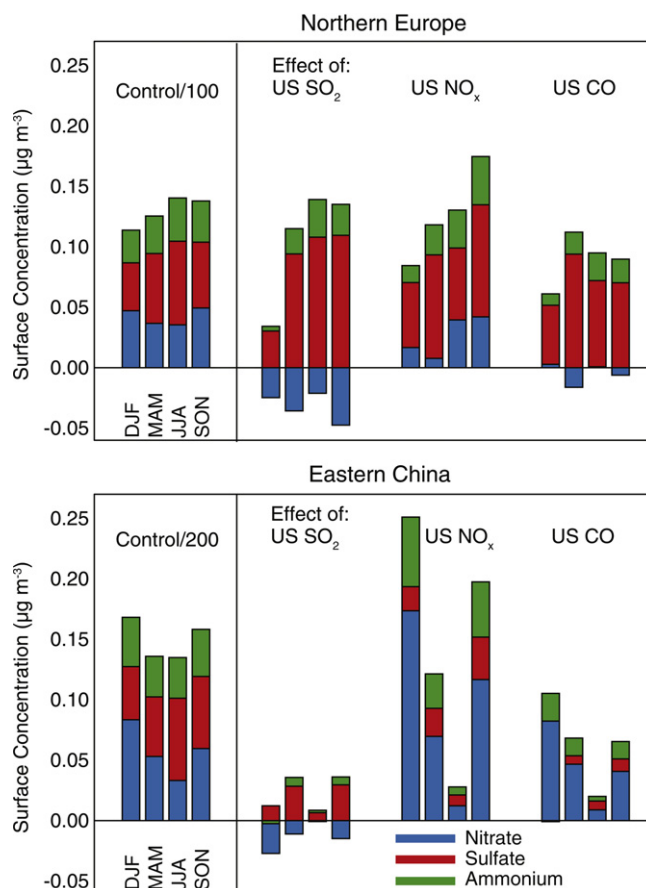

Fig. 2. Speciation of sulfate–nitrate–ammonium (SNA) PM concentrations in surface air for the receptor regions of Fig. 1 (left panels) and corresponding intercontinental enhancements from US anthropogenic emissions of SO_2, NO_x, and CO (right panels). Values are seasonal means from the GEOS–Chem model. Seasonal concentrations have been divided by 100 (Northern Europe) and 200 (Eastern China) to fit on scale.

distance downwind due to wet and dry removal. US SO_2 emissions thus mainly influence western Europe, by up to 0.2 $\mu g\ m^{-3}$ on an annual mean basis. This enhancement is comparable to the results of Park et al. (2004), but larger than those of Chin et al. (2007) and Liu et al. (2009b). Increases in sulfate are partly offset by decreases in nitrate due to competition for ammonium (West et al., 1999; Park et al., 2004). US SO_2 emissions actually induce a net decrease in PM over eastern China in winter because the replacement of $2NH_4NO_3$ by $(NH_4)_2SO_4$ results in net loss of PM mass.

We find that US NO_x emissions enhance European and Asian PM concentrations by up to 0.25 $\mu g\ m^{-3}$ on an annual basis. The patterns in Fig. 1 show that this is not due to direct intercontinental transport of nitrate and its precursors, but to an increase in background oxidant levels that promotes formation of sulfate and nitrate from local sources of SO_2 and NO_x in the receptor regions. US NO_x emissions increase annual mean tropospheric ozone by 3.3%, OH by 3.3%, and H_2O_2 by 0.2% in the Northern Hemisphere, with larger effects in Europe, which is directly downwind.

Fig. 2 shows that the intercontinental PM enhancement from US NO_x emissions is mostly driven by sulfate in northern Europe but by nitrate in eastern China. Conversion of SO_2 to sulfate in the model is mostly by in-cloud oxidation by H_2O_2 and ozone. The process by H_2O_2 is faster than that by ozone, and in the presence of excess H_2O_2, SO_2 oxidation is insensitive to increases in oxidants. At the high latitudes of northern Europe, H_2O_2 production is slow so that SO_2 in-cloud oxidation is H_2O_2-limited over SO_2 source regions for most of the year; thus increases of ozone and H_2O_2 result in

increased sulfate. The sulfate increase is largest in fall due to a combination of extensive cloud cover and significant enhancement of oxidants. Cloud cover is also extensive in winter but the increase in oxidants is then less. In eastern China, by contrast to northern Europe, H_2O_2-limited conditions prevail only in winter, and cloud cover is then infrequent due to the winter monsoon.

The large enhancement of nitrate PM in eastern China compared to northern Europe in Fig. 2 reflects the regional presence of excess ammonia for NH_4NO_3 formation. We find that increases in total inorganic nitrate (gas-phase HNO_3 plus aerosol nitrate) are comparable in the two regions, but northern Europe has less excess ammonia so that this inorganic nitrate remains in the gas-phase as HNO_3. The effect of increased oxidants on nitrate PM in eastern China is largest in winter, when cold temperatures promote nitrate partitioning into the aerosol and when oxidation of NO_x is relatively slow. In summer, oxidation of NO_x by OH in the region is sufficiently fast that the effect of added oxidants is small. This is not the case for

northern Europe where OH concentrations are much lower because of the higher latitude.

CO emissions in the US decrease annual mean tropospheric OH by 1.9% in the Northern Hemisphere, but increase ozone by 1.3% and H_2O_2 by 3.4%. The effect on PM is qualitatively similar to that of US NO_x emissions, but not as large due to the decrease in OH concentrations. Speciation and seasonal patterns in Fig. 2 are similar to those for the effect of US NO_x emissions. The decrease in OH, affecting NO_x oxidation, results in some negative effects on nitrate in northern Europe. Nitrate concentrations still increase over China in seasons outside of summer, when oxidation of NO_x by ozone is a significant nitrate formation pathway (Dentener and Crutzen, 1993).

3.2. Effect of Asian emissions

Fig. 3 shows the intercontinental increases in PM from Asian emissions. The intercontinental influence of Asian SO_2 emissions
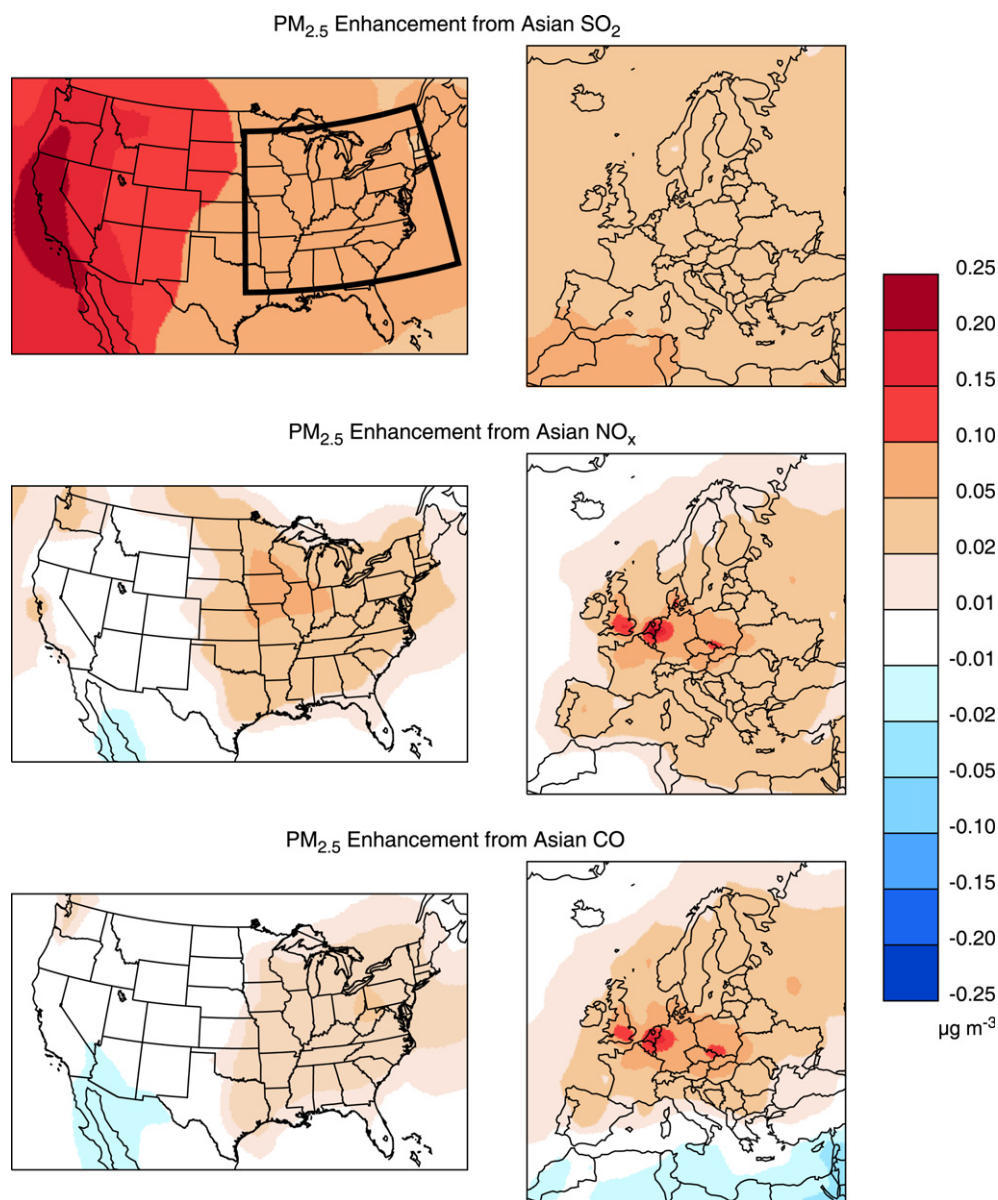
Fig. 3. Annual mean enhancements of surface PM concentrations in Europe and the United States from Asian anthropogenic emissions of SO_2 (top), NO_x (middle), and CO (bottom). Values are differences between the control GEOS−Chem simulation and a sensitivity simulation with Asian anthropogenic sources shut off. Boxes in the top panels outline the receptor region in the United States for which speciation and seasonality are shown in Fig. 4.

are strongest in the western US where subsidence from the free troposphere brings Asian outflow to the surface. The enhancement of 0.1–0.25 $\mu g\ m^{-3}$ in this region is comparable to the results of Park et al. (2004), Heald et al. (2006), and Chin et al. (2007). The effect on Europe is weaker and more uniform than that of US SO_2 emissions, which are closer upwind (Fig. 1). Increases in sulfate cause lower levels of nitrate due to competition for ammonium, similar to the effects of US SO_2 emissions previously discussed.

The intercontinental influence of Asian NO_x on PM shown in Fig. 3 is weaker than that of US emissions (Fig. 1). Asian NO_x emissions increase annual mean tropospheric ozone by 8.8%, OH by 12.7%, and H_2O_2 by 1.1%. These increases are much larger than those from US emissions (Section 3.1), but the effect on PM is also contingent on emissions in the receptor continent. Thus the effect on the US is relatively weak, up to 0.1 $\mu g\ m^{-3}$ over the central US where excess ammonia from agricultural emissions promotes ammonium nitrate formation. The effect on northern Europe (up to 0.2 $\mu g\ m^{-3}$) is also weaker than for US emissions, which are closer upwind (Fig. 1). Fig. 4 shows that sulfate accounts for most of the overall increase in PM over the eastern US and northern Europe. The increase is weakest in summer when Asian NO_x emissions actually cause H_2O_2 concentrations to decrease over the US and Europe. This reflects the large Asian source of NO_x and complicated effects of NO_x emissions on H_2O_2. On the one hand, NO_x leads to ozone production and from there to H_2O_2 production. On the other hand, NO_x increases OH, which is a major H_2O_2 sink.

Asian CO emissions decrease annual mean tropospheric OH by 5.6% and increase ozone by 3.1% and H_2O_2 by 9.4%. Figs. 3 and 4 show that Asian CO affects northern European PM in a very similar manner to US CO emissions (Figs. 1 and 2) and with a similar magnitude as Asian NO_x. Similar to the effect of US CO emissions, Asian CO emissions increase in-cloud sulfate production by H_2O_2 over Europe causing sulfate enhancements in all seasons. Changes in sulfate in the US are largely driven by increased production by H_2O_2. A reduction in gas-phase production of sulfate by OH detracts from increased aqueous production. Nitrate concentrations decrease in both northern Europe and the US because of the lower OH concentrations.

4. Discussion

Anthropogenic emissions of NO_x and CO perturb sulfate and nitrate PM concentrations on intercontinental scales by affecting the background concentrations of oxidants and hence the oxidation rates of SO_2 and NO_x emitted in receptor regions. NO_x emissions cause OH, ozone, and H_2O_2 to increase on the hemispheric scale although H_2O_2 can decrease in some regions and seasons. CO emissions cause OH to decrease, but ozone and H_2O_2 concentrations to increase. CO increases H_2O_2 formation in part by decreasing OH, a major sink of H_2O_2 and in part by decreasing the OH/HO_2 ratio and thus promoting H_2O_2 formation. Most of the intercontinental effect on sulfate is through increases in ozone and H_2O_2, which drive faster SO_2 in-cloud oxidation under H_2O_2-limited conditions. Most of the effect of nitrate is through the increase in ozone in winter and the shoulder seasons, when NO_x oxidation is otherwise slow and the cold temperatures promote nitrate fractionation into the aerosol.

The intercontinental PM enhancement from NO_x and CO emissions tends to peak in receptor regions with the highest domestic sources of SO_2, NO_x, and NH_3, and hence with the highest PM concentrations. It is thus of particular policy relevance. US emissions of NO_x and CO increase PM by over 0.25 $\mu g\ m^{-3}$ (annual mean) in polluted regions of northern Europe and eastern China, a much larger effect than that of US SO_2 emissions. PM concentrations in the US are less sensitive to intercontinental NO_x and CO emissions because domestic PM sources are lower. The effect of Asian NO_x and CO on PM in the eastern US is still ~0.1 $\mu g\ m^{-3}$, comparable to the effect of Asian SO_2 emissions. Northern Europe is more sensitive to US than to Asian emissions of NO_x and CO because of the greater upwind proximity.

Our study used emission inventories for 2000. Emissions have changed over the past decade and also have some uncertainty. The more recent EPA NEI 2005 inventory for the US (http://www.epa.gov/ttnchie1/net/2005inventory.html) includes 2% more NO_x, 20% less CO, and 57% less SO_2 than used in this work, while the East Asian 2006 inventory of Zhang et al. (2009b) includes 12% more NO_x, 19% more CO, and 14% less SO_2. These differences arise from both actual trends and improved emission accounting. The emission changes would tend to increase the importance of NO_x and CO vs. SO_2 in driving intercontinental influence on PM.

The intercontinental influence of NO_x and CO emissions on PM through changes in background oxidant fields depends in a complex way on the chemical environment in the receptor region including (1) the relative importance of OH, ozone, and H_2O_2 as oxidants for SO_2 and NO_x; (2) cloudiness; (3) H_2O_2 limitation of in-cloud SO_2 oxidation by H_2O_2; and (4) availability of ammonia for nitrate PM formation. All of these factors have substantial uncertainty in models, and the corresponding errors cannot easily be quantified using a single model. The intercontinental influence on oxidant levels is itself uncertain. The HTAP multi-model inter-comparison of Fiore et al. (2009) shows good agreement between
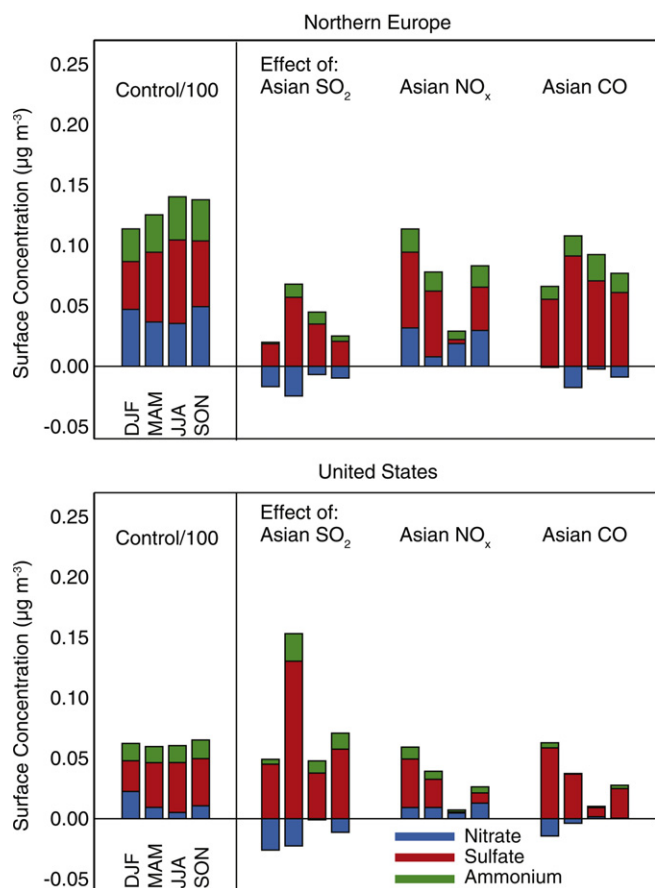

Fig. 4. Speciation of sulfate–nitrate–ammonium (SNA) PM concentrations in surface air for the receptor regions of Figs. 1 and 2 (left panels) and corresponding intercontinental enhancements from Asian anthropogenic emissions of SO_2, NO_x, and CO (right panels). Values are seasonal means from the GEOS–Chem model. Seasonal concentrations from the control simulation have been divided by 100 to fit on scale.

models for intercontinental influences on ozone, but no such evaluations have been conducted for OH and H_2O_2. In view of the potential for significant impacts presented in this paper, it would be of great interest to carry out a multi-model intercomparison of intercontinental influences of NO_x and CO emissions on PM.

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