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Source-receptor relationships between East Asian sulfur dioxide emissions and Northern Hemisphere sulfate concentrations

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Abstract. We analyze the effect of varying East Asian (EA) sulfur emissions on sulfate concentrations in the Northern Hemisphere, using a global coupled oxidant-aerosol model (MOZART-2). We conduct a base and five sensitivity simulations, in which sulfur emissions from each continent are tagged, to establish the source-receptor (S-R) relationship between EA sulfur emissions and sulfate concentrations over source and downwind regions. We find that from west to east across the North Pacific, EA sulfate contributes approximately 80%–20% of sulfate at the surface, but at least 50% at 500 hPa. Surface sulfate concentrations are dominated by local anthropogenic sources. Of the sulfate produced from sources other than local anthropogenic emissions (defined here as "background" sulfate), EA sources account for approximately 30%–50% (over the Western US) and 10%–20% (over the Eastern US). The surface concentrations of sulfate from EA sources over the Western US are highest in MAM (up to 0.15 μg/m^3), and lowest in DJF (less than 0.06 μg/m^3). Reducing EA SO_2 emissions will significantly decrease the spatial extent of the EA sulfate influence (represented by the areas where at least 0.1 μg m^{-3} of sulfate originates from EA) over the North Pacific both at the surface and at 500 hPa in all seasons, but the extent of influence is insensitive to emission increases, particularly in DJF and JJA. We find that EA sulfate concentrations over most downwind regions respond nearly linearly to changes in EA SO_2 emissions, but sulfate concentrations over the EA source region increase more slowly than SO_2 emissions, particularly at the surface and in winter, due to limited availability of oxidants (in particular of H_2O_2, which oxidizes SO_2 to sulfate in the aqueous phase). We find that similar estimates of the S-R relationship for trans-Pacific transport of EA sulfate would be obtained using either sensitivity (i.e., varying emissions from a region to examine the effects on downwind concentrations) or tagging techniques. Our findings suggest that future changes in EA sulfur emissions may cause little change in the sulfate-induced health impact over downwind continents. However, SO_2 emission reductions may significantly reduce the sulfate concentrations and the resulting negative radiative forcing over the North Pacific and the United States, thus providing a warming tendency.

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