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East Asian SO₂ pollution plume over Europe – Part 2: Evolution and potential impact

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Abstract. We report on the first observation-based case study of an aged East Asian anthropogenic SO₂ pollution plume over Europe. Our airborne measurements in that plume detected highly elevated SO₂ mole fractions (up to 900 pmol/mol) between about 5000 and 7000 m altitude. Here, we focus on investigations of the origin, dispersion, evolution, conversion, and potential impact of the observed excess SO2. In particular, we investigate SO₂ conversion to gas-phase sulfuric acid and sulfuric acid aerosols. Our FLEXPART and LAGRANTO model simulations, along with additional trace gas measurements, suggest that the plume originated from East Asian fossil fuel combustion sources and, 8-7 days prior to its arrival over Europe, ascended over the coast region of central East Asia to 9000 m altitude, probably in a cyclonic system with an associated warm conveyor belt. During this initial plume ascent a substantial fraction of the initially available SO₂ must have escaped from removal by cloud processes. Hereafter, while mostly descending slowly, the plume experienced advection across the North Pacific, North America and the North Atlantic. During its upper troposphere travel, clouds were absent in and above the plume and OH-induced gas-phase conversion of SO₂ to gas-phase sulfuric acid (GSA) was operative, followed by GSA nucleation and condensation leading to sulfuric acid aerosol formation and growth. Our AEROFOR model simulations indicate that numerous large sulfuric acid aerosol particles were formed, which at least tempora\-rily, caused substantial horizontal visibility degradation, and which have the potential to act as water vapor condensation nuclei in liquid water cloud formation, already at water vapor supersaturations as low as about 0.1%. Our AEROFOR model simulations also indicate that those fossil fuel combustion generated soot particles, which have survived cloud induced removal during the initial plume ascent, have experienced extensive $\rm H_2SO_4/H_2O$ -coating, during upper troposphere plume travel. This coating may have dramatically altered the morphology and markedly increased the light absorption efficiency of soot particles.

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