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Volcanic ash chemical aging from multiple observational constraints for the Pinatubo eruption

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Condensation of sulfuric acid formed from the co-injected sulfur dioxide on volcanic ash particles, so-called chemical aging, increases the particle size and changes their microphysical and optical properties. The larger aged particles have a higher removal rate, which reduces their lifetime. On the other hand, the aging increases the scattering cross-section, and therefore the ash optical depth is increasing due to aging. The uptake of sulfuric acid by volcanic ash delays the formation of new sulfate particles depending on the level of aging, which is characterized by the number of sulfuric acid layers coating a single ash particle (i.e., monolayers). Both the formation of sulfate aerosols and sulfuric acid uptake by ash particles affect the development of a volcanic plume and its radiative impact.

We employ the ECHAM5/MESSy atmospheric chemistry general circulation model (EMAC) to simulate the chemical aging of volcanic ash in the 1991 Pinatubo eruption volcanic plume. We emit 17Mt of SO₂ and 75Mt of fine ash. Two aerosol modes represent ash size distribution: accumulation and coarse with 0.23 and 3.4 μm median radii, respectively. We allow the sulfuric acid to condense on the ash particles and assume different levels of aging (from not aged to highly aged). We use independent observations for sulfur dioxide, volcanic ash mass, volcanic ash optical depth, and plume coverage area from the Advanced Very-High-Resolution Radiometer (AVHRR) observations and total optical depth from the Stratospheric Aerosol and Gas Experiment II (SAGE II). We constrain the number of monolayers on ash particles by testing simulated ash surface area and optical depth calculated within a fully coupled online stratospheric-tropospheric chemistry model against observations. The level of volcanic ash aging strongly affects the surface area of the volcanic ash plume, ranging from 3x10⁶ km² to 6x10⁶ km², compared to 3.8x10⁶ km² from AVHRR retrievals. The volcanic ash optical depth, averaged over the volcanic plume area, ranges between 2 and 3.6. Using five monolayer coating assumption allows us to better reproduce the observed SO₂ mass, its decay rate, total plume surface area, and ash optical depth. Most of the coarse ash particles are removed within a week after the eruption reducing the amount of sulfuric acid within the volcanic plume. The smaller particles have much longer residence time and continue to uptake sulfuric acid for more than three months.

