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Global distribution and climate forcing of solid and aqueous sulfate aerosols: effect of the hysteresis of particle phase transitions

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The partitioning between solid and aqueous phases of tropospheric sulfate-ammonium particles is simulated with a global 3D chemical transport model (CTM). The simulation explicitly accounts for the hysteresis of particle phase transitions by transporting aqueous sulfate and three solid sulfate forms (viz. ammonium sulfate, letovicite, and ammonium bisulfate). We find that the solids mass fraction on a sulfate basis is 0.34, partitioned as 93% ammonium sulfate, 6% letovicite, and 1% ammonium bisulfate. The fraction increases with altitude from 0.10-0.30 in the boundary layer to 0.60-0.80 in the upper troposphere. Omission of the hysteresis effect in the CTM by assuming that particle phase follows the lower side of the hysteresis loop increases the solids mass fraction from 0.34 to 0.56. An upper-side assumption decreases the fraction to 0.17. For the anthropogenic sulfate, the base-case simulation finds that solid particles contribute 41% of the global burden, 26% of the clear-sky optical thickness, 31% of the clear-sky SDCF, and 37% of the full-sky sulfate direct climate forcing (SDCF), a trend that reflects the correlation of solid particles with clear skies. A change to the model, omitting hysteresis by assuming that all particles are aqueous, results in an overestimate of the SDCF by +8% compared to the base case. A converse assumption that crystallization occurs at the deliquescence relative humidity underestimates the SDCF by -8%. A case that assumes that aqueous particles occur whenever the ambi-

ent relative humidity exceeds the crystallization relative humidity biases the SDCF by +5%. A case that includes hysteresis but omits the difference in the fraction of radiation backscattered to space by aqueous compared to solid particles changes the SDCF by +15%.