Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 4, 183-214, 2004
www.atmos-chem-phys.net/4/183/2004/

Effects of the physical state of tropospheric ammonium-sulfate-nitrate particles on global aerosol direct radiative forcing

S. T. Martin[1], H.-M. Hung[1], R. J. Park[1,2], D. J. Jacob[1,2], R. J. D. Spurr[3], K. V. Chance[3], and M. Chin[4,5]
[1]Division of Engineering and Applied Sciences, Harvard University, USA
[2]Department of Earth and Planetary Sciences, Harvard University, USA
[3]Harvard-Smithsonian Center for Astrophysics, USA
[4]School of Earth and Atmospheric Sciences, Georgia Institute of Technology, USA
[5]Laboratory for Atmospheres, NASA Goddard Space Flight Center, USA

Abstract. The effect of aqueous versus crystalline sulfate-nitrate-ammonium tropospheric particles on global aerosol direct radiative forcing is assessed. A global three-dimensional chemical transport model predicts sulfate, nitrate, and ammonium aerosol mass. An aerosol thermodynamics model is called twice, once for the upper side (US) and once for lower side (LS) of the hysteresis loop of particle phase. On the LS, the sulfate mass budget is 40% solid ammonium sulfate, 12% letovicite, 11% ammonium bisulfate, and 37% aqueous. The LS nitrate mass budget is 26% solid ammonium nitrate, 7% aqueous, and 67% gas-phase nitric acid release due to increased volatility upon crystallization. The LS ammonium budget is 45% solid ammonium sulfate, 10% letovicite, 6% ammonium bisulfate, 4% ammonium nitrate, 7% ammonia release due to increased volatility, and 28% aqueous. LS aerosol water mass partitions as 22% effloresced to the gas-phase and 78% remaining as aerosol mass. The predicted US/LS global fields of aerosol mass are employed in a Mie scattering model to generate global US/LS aerosol optical properties, including scattering efficiency, single scattering albedo, and asymmetry parameter. Global annual average LS optical depth and mass scattering efficiency are, respectively, 0.023 and 10.7 m^2 (g SO_4^{-2})$^{-1}$, which compare to US values of 0.030 and 13.9 m^2 (g SO_4^{-2})$^{-1}$. Radiative transport is computed, first for a base case having no aerosol and then for the two global fields corresponding to the US and LS of the hysteresis loop. Regional, global, seasonal, and annual averages of top-of-the-atmosphere aerosol radiative forcing on the LS and US (F_L and F_U, respectively, in W m^{-2}) are calculated. Including both anthropogenic and natural emissions, we obtain global annual averages of F_L=-0.750, F_U=-0.930, and $\Delta F_{U,L}$=24% for full sky calculations without clouds and F_L=-0.485, F_U=-0.605, and $\Delta F_{U,L}$=25% when clouds are included. Regionally, $\Delta F_{U,L}$=48% over the USA, 55% over Europe, and 34% over East Asia. Seasonally, $\Delta F_{U,L}$ varies from 18% in DJF to 75% in SON over the USA. The global annual average contribution from anthropogenic aerosol is F_L=-0.314 and F_U=-0.404, which yield normalized direct radiative forcings (G) of G_L=-205 W (g SO_4^{-2})$^{-1}$ and G_U=-264 W (g SO_4^{-2})$^{-1}$.

Final Revised Paper (PDF, 7087 KB) Discussion Paper (ACPD)

Citation: Martin, S. T., Hung, H.-M., Park, R. J., Jacob, D. J., Spurr, R. J. D., Chance, K. V., and Chin, M.: Effects of the physical state of tropospheric ammonium-sulfate-nitrate particles on global aerosol direct radiative forcing, Atmos. Chem. Phys., 4, 183-214, 2004. Bibtex EndNote Reference Manager