EGU2020-11366
https://doi.org/10.5194/egusphere-egu2020-11366
EGU General Assembly 2020

Global Survey of Aerosol Acidity from Polluted to Remote Locations: Measurements and Comparisons with Global Models

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The inorganic composition of aerosol impacts numerous chemical and physical processes and properties. However, many chemical transport models show large variability in both the concentration of the inorganic aerosols and their precursors (up to 3 orders of magnitude differences) and the inorganic aerosol composition. Different models predict very different properties (e.g., aerosol liquid water concentration and aerosol acidity) and outcomes (e.g., heterogeneous uptake of gases or aerosols' direct and indirect impacts on climate). Here, we use airborne observations from campaigns conducted around the world to investigate how the inorganic fine aerosol (PM_1) composition, and one of its key parameters, aerosol acidity, changes from polluted regions (Mexico City, Los Angeles, Northeastern US, and Seoul) to remote ocean basins (the Atmospheric Tomography campaigns 1 and 2) in order to provide constraints for the chemical transport models. I find that the empirical ammonium balance with major ions (ammonium balance = mol NH_4 / (2×mol SO_4 + mol NO_3)) rapidly decreases from ~1 at the highest inorganic PM_1 concentration to 0 at the lowest inorganic PM_1. The data indicate a robust trend for ammonium balance vs inorganic PM_1 at all altitude levels in the troposphere, suggesting that NH_3 emissions and subsequent neutralization of H_2SO_4 becomes negligible in the most remote (lowest inorganic PM_1) regions. Further, a robust trend for PM_1 pH (calculated with E-AIM) vs inorganic PM_1 is observed at all levels for these campaigns, as well, decreasing from a pH of ~3 to a pH of ~ –1

from the highest to lowest inorganic PM_1. The data overall implies very low NH_3 (and NH_4^+) throughout most of the atmosphere, contrary to predictions of some models, implying different physical properties than predicted in models. We compare these trends of ammonium balance and pH vs inorganic PM_1 against 9 chemical transport models (CTMs), and we find that the CTMs show large variability for both the ammonium balance and pH vs inorganic PM_1, compared to observations. Generally, we find a high bias in the ammonium balance and pH, likely due to too much NH_3 in model (possibly too high NH_3 emissions over oceans or too long lifetime) and inclusion of externally mixed seasalt into the submicron pH calculation. These results overall would imply different aerosol properties in the models than observed, impacting the chemistry, optical properties, and cloud properties.

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How to cite: Nault, B. A., Campuzano-Jost, P., Jo, D., Day, D., Bahreini, R., Bian, H., Chin, M., Clegg, S., Colarco, P., Kodros, J., Lopez-Hilfiker, F., Marais, E., Middlebrook, A., Neuman, A., Nowak, J., Pierce, J., Thornton, J., Tsigaridis, K., and Jimenez, J. and the ATom Science Team: Global Survey of Aerosol Acidity from Polluted to Remote Locations: Measurements and Comparisons with Global Models, EGU General Assembly 2020, Online, 4–8 May 2020, EGU2020-11366, https://doi.org/10.5194/egusphere-egu2020-11366, 2020

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