

Rethinking organic aerosols: Semivolatile emissions and photochemical aging

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Organic particulate matter is a dynamic system coupling phase partitioning with multiple generations of chemical reactions in both the gas and condensed phases. We have recently shown that essentially all primary organic aerosol sources in fact emit material over a wide range of volatility, most of which evaporates as particles dilute to ambient conditions. Likewise, traditional secondary organic aerosol sources (oxidation of volatile terpenes and aromatics) generate reaction products with a wide range of volatilities, most of which remain vapors under ambient conditions.

Recent observations have also established that ambient organic aerosol are frequently highly oxidized¹ and relatively non-volatile² (OOA). This sets a target for experiments and chemical mechanisms, which most current experiments and mechanisms miss. They miss because chemistry in relatively short timescale experiments is either too slow or otherwise not representative of true ambient conditions.

We shall explore possible missing chemistry, starting with the hypothesis that gas-phase oxidation is the principle driver generating ambient OOA. We shall describe a series of experiments designed to test this hypothesis for various organic aerosol sources, ranging from traditional SOA sources such as α -pinene + ozone to evaporated diesel emissions. In each case we follow aerosol formation (whether by emission followed by dilution to ambient conditions or chemical formation at typical ambient levels) with exposure to OH radicals. By tracking the evolving mass, volatility, and mass-spectra of the organic aerosol, we are able to test the ability of OH radicals to generate OOA.

[1] Q. Zhang *et al.* (2007) Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced northern hemisphere midlatitudes. *Geophys. Res. Lett.*, **34**. [2] Huffman *et al.* (2009) Chemically-resolved aerosol volatility measurements from two megacity field studies. *Atm. Chem. Phys. Discuss.* **9**, 2645-2697.

A model for “cold-snap” episodes occurring during the Mesozoic

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The Mesozoic, perhaps representing the longest period of warmth during the Phanerozoic Earth history has been repeatedly affected by short-lived cold interludes lasting less than a million of years in length. The evidence for these cooling events includes glendonite abundance, stable oxygen isotope records and faunal migration. While plausible mechanisms have been proposed for the two largest ice age event of the Phanerozoic, convincing explanations for these Mesozoic cold snaps are still lacking. Here, we investigate the climate-carbon cycle behavior during these events with a particular focus on the Middle Late Jurassic Transition using a general circulation model with coupled components for atmosphere, ocean, cryosphere and biogeochemical cycles of C, O and P. We force our climate-carbon model with geological evidences of the timing of the evolution of carbonate production. We show that the general drawdown of carbonate platforms is a powerful mechanism capable of explaining a fast atmospheric CO₂ decrease and a moderate sea level drop associated with ice-sheet buildup. Temporary nature of the carbonate drawdown explains the relative short time of these cold events.