Organics in the mix: how important are they for the uncertainty in global aerosol-climate effects?

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Secondary organic aerosol (SOA) is one of the most challenging problems to solve in global aerosol-climate science, but how does it rank alongside other uncertainties in global models? Here, we perform a comprehensive uncertainty analysis of a global model to try to put SOA uncertainty in context. We quantify the uncertainty in modelled concentrations of CCN on a global scale due to 28 parameters related to aerosol and precursor gas emissions, aerosol processes and model structures. The unique approach, using emulation and variance analysis, enables the CCN variance in every grid box of the model to be decomposed into contributions from each parameter, including parameter interactions. The global production of biogenic SOA was perturbed between 5 and 360 Tg a^{-1}, and anthropogenic SOA between 3 and 160 Tg a^{-1}. The anthropogenic SOA uncertainty is ranked 7th out of 28 and accounts for about 5% of the global mean CCN uncertainty, somewhat higher than for anthropogenic SO_2 (although the latter emissions are assumed to be known to within ±50%, much better than for SOA). Biogenic SOA is only about half as important. Given the large assumed SOA uncertainty, these results suggest a rather low importance of SOA production to global CCN uncertainty. These results are based on the assumption that secondary organics do not contribute to nucleation, only to particle growth. However, the latest measurements suggest biogenic secondary organics need to be included in the nucleation rate expression. When such a mechanism is used in the model, we obtain better agreement with the seasonal cycle of particle concentration measurements and a much greater contribution of SOA uncertainty to the overall uncertainty in CCN. The sensitivity of CCN to SOA therefore depends on the extent to which organics control nucleation.

Changes in amino acid nitrogen isotopic composition patterns during phytoplankton degradation

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The isotopic composition of organic nitrogen (N) in marine, estuarine and lacustrine sediments is often used to reconstruct paleoenvironmental conditions. Bacteria-mediated degradation and sedimentary diagenesis can modify the isotopic composition of the organic matter (OM). In order to study the biogeochemical mechanisms behind N isotope shifts in bulk organic N during early diagenesis, and to verify the robustness of algal amino acid (AA) $d^{15}N$ patterns during partial degradation, as well as the integrity of amino acid-based degradation indicators, we conducted incubation experiments, in which we simulated the decay of algal OM (*Fragilaria crotonensis*) under controlled oxic and anoxic conditions. During progressing decomposition, we monitored the concentration and N isotopic composition of bulk OM and of particulate amino acids (AAs).

Particulate N concentrations decreased during the 300 day experimental period from 4 mg L^{-1} to 1 mg L^{-1} in the oxic and to 2 mg L^{-1} in the anoxic set-up. In both experimental settings, the $\delta^{15}N$ values of the particulate N increased during simulated degradation. The $d^{15}N$ shift was more pronounced in the oxic (3.5‰) than in the anoxic (1.9‰) incubation. The total particulate AA concentrations decreased by 55% in the anoxic and by 46% in the oxic environment. Bacterial buildup, most pronounced in the initial degradation phase and under oxic conditions, was indicated by an increase in the concentration of D-glutamic acid (D-Glx), which is unique to bacteria. After two days of incubation, most AAs were enriched in ^{15}N relative to the fresh bulk diatom biomass, independent of the redox conditions. In the course of the experiments, however, both the AA composition and N isotopic composition showed trends that were distinctive for oxic versus anoxic degradation. Overall, our findings show that bacterial degradation and biosynthesis during early sedimentary diagenesis overprint initial diatom $\delta^{15}N$-AA patterns and contribute to the alteration of bulk N isotope signatures.

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