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Modelled radiative effects of sea spray aerosol using a source function encapsulating wave state

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Sea spray aerosol particles have significant effects on global climate by scattering solar radiation (direct effect) and modifying cloud properties (indirect effect). Sea spray consists mainly of sea salt, but in biologically active regions, major fraction of sea spray may come in the form of primary marine organic matter (PMOM). Traditionally, sea spray flux has been parameterized in global models in terms of wind speed, and organic fraction of sea spray in terms of chlorophyll-a concentration. In this study, we have incorporated recently developed parameterizations for the sea spray aerosol source flux into the global aerosol-climate model ECHAM-HAMMOZ. The parameterizations encapsulate the wave state via Reynolds number, and predict the organic fraction of the sea spray aerosol source flux. The model was then used to investigate the direct and indirect effects of sea spray aerosol particles. We compared simulated sea spray concentrations with in-situ measurements from Mace Head (North Atlantic), Point Reyes (North Pacific), and Amsterdam Island (Southern Indian Ocean). Aerosol optical depth (AOD) was compared with satellite measurements from PARASOL.

Modelled annual mean global emissions of sea salt and PMOM were 805 Tg yr^{-1} (uncertainty range of 378-1233 Tg yr^{-1}) and 1.1 Tg yr^{-1} (0.5-1.8 Tg yr^{-1}), respectively. Sea salt emissions were considerably lower than the majority of previous estimates, but PMOM was in the range of previous studies.

The model captured sea salt concentrations fairly well in the smaller size ranges at Mace Head (annual normalized mean bias of -13% for particles with vacuum aerodynamic diameter D_{va} <1 μm), Point Reyes (-29% for particles with aerodynamic diameter D_a <2.5 μm) and Amsterdam Island (-52% for particles with D_a <1 μm), but larger particles were clearly overestimated (899% for particles with 2.5 μm <D_a <10 μm) at Amsterdam Island. Organic matter concentrations were underestimated at Mace Head and at Amsterdam Island, and the overestimation of organic matter in Point Reyes was explained by very large continental contribution at that site. This indicates that the parameterization for the organic fraction of sea spray is biased low.

Modelled AOD was on average lower than the measured by PARASOL (mean over oceans 0.10 and 0.16, respectively), but the new sea spray source function improved the AOD compared to model default sea spray source function (normalized mean errors of 35% and 41%, respectively). In the remote area around the Amsterdam Island, measured monthly-mean AOD fell within the uncertainty of the model simulations in 90% of the simulated months.

Effective radiative forcing (total radiative effect at the top of the atmosphere) of sea spray was -0.2 W m^{-2}. Contrasting previous global modeling studies, the global mean indirect effect of sea spray was positive (0.3 W m^{-2}). This was caused by large sea salt particles preventing condensation of sulfuric acid and water on smaller sulfate particles. Direct effect of sea spray was -0.5 W m^{-2} which is in the range of previous estimates. Although sea salt had a positive indirect effect, the indirect effect of PMOM was negative (-0.07 W m^{-2}) due to its high cloud-activation efficiency.