Importance of solubility for climate impacts of organic aerosol

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Introduction

Aerosol particles introduce high uncertainties to estimates of the Earth's radiative forcing. If exposed to a given relative humidity (RH), aerosol particles containing soluble material can absorb water and grow in size (hygroscopic growth). If RH is increased further beyond supersaturation (RH >100%) the particles can act as cloud condensation nuclei (CCN). Both of these processes impact the climate influence of aerosol particles. The amount of water that can be absorbed by aerosol particles depends on their chemical composition and can be represented with the single hygroscopic parameter κ (Petters and Kreidenweis, 2007).

Organic aerosols (OA) contribute a large fraction (20-90%) of atmospheric submicron particulate mass. On the other hand, they consist of thousands of compounds with different properties. The complexity of OA is a challenge for representing OA in large scale atmospheric models. If potential limited solubility of OA is not accounted for, the same κ value is expected in both the sub-and supersaturated regimes. The κ values for organic aerosols inferred from hygroscopic growth factor (GF) measurements in sub-saturation often differ from the corresponding κ values inferred from CCN measurements in super-saturation, κ_{HGF} being typically lower than κ_{CCN}. The underlying reasons for this behavior not fully resolved.

In this study, we combine climate model simulations with analysis of laboratory and field observations to investigate the importance of solubility for OA hygroscopicity and CCN activation.

Methods

We conducted a set of simulations with the Norwegian Earth System Model (NorESM) to test the sensitivity of climate predictions to the hygroscopicity of OA. Four different κ_{OA} values were selected for the organic fraction in NorESM (0.01, 0.05, 0.1, and 0.15), representing the range of values reported in literature. The simulations were made in two modes, offline mode (aerosols are not allow to affect meteorology) and online mode, both for present day (PD). Furthermore, we used solubility distributions estimated for isoprene (IP) and monoterpene (MT) SOA (Riipinen et al, 2015) for exploring the effect of limited solubility on the κ values reported for these SOA types based on laboratory and field data.

Results and Discussion

The results from NorESM suggest that the variation in the reported κ_{OA} values results in a significant influence on the predicted radiative forcing. Aerosol-cloud interactions dominate the sensitivity over the direct effect of hygroscopic aerosol particles on the radiative properties of the atmosphere. The sensitivity to κ_{OA} parameter also depends considerably on the ambient RH (see Fig. 1).

The analysis of the laboratory data (Pajunoja et al., 2015) in water can be a key property in explaining the observed differences between the hygrosopicity and CCN activation of IP- and MT-derived SOA at sub- (90 % RH) vs. supersaturated conditions. At RHs below 80 %, however, in particular for the MT SOA, additional processes, such as surface adsorption or considerable non-ideality of the aqueous and/or organic phases, are needed to explain the observed water uptake.
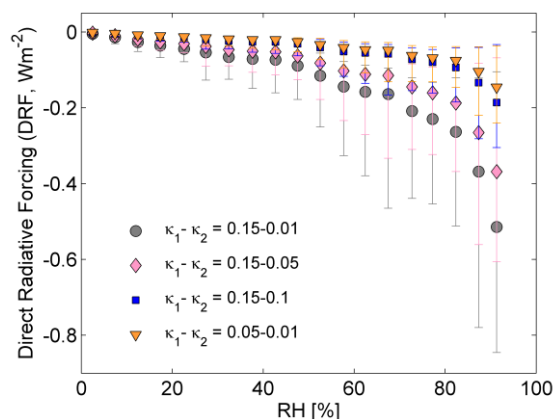

Figure 1. The impact of κ_{OA} on direct radiative forcing as a function of RH, calculated with NorESM. Error bars show 10 and 90 percentiles.

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