A holistic approach of primary Ice Nucleating Particles in immersion freezing from organic aerosols and mineral dust simulated by TM4-ECPL and EC-EARTH 3

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Aerosol-cloud interactions are among the most important sources of uncertainty in climate projections. Atmospheric particles that enable heterogeneous ice formation are known as Ice Nucleating Particles (INP) and affect clouds' lifetime, electrification and radiative properties as well as precipitation rates. INP concentrations differ substantially between terrestrial and marine environments, with those over sea being in general lower. The simulated ice crystal concentrations in mixed-phase cloud (MPC) are affected by uncertainties in the INP concentrations, leading to discrepancies in the modelled top-of-atmosphere radiative flux, and thereby in the climate sensitivity of the model. Mineral dust and more specifically its K-rich feldspar and quartz content, is thought to be globally the most important type of INP in MPC. Chemistry-transport and Earth System models can simulate INP based on dust mineralogy and experimentally-derived immersion freezing parameterizations. Dust mineralogy depends upon regional variations in the mineralogy of dust sources as well as for the differences in size-resolved mineralogy between the emitted dust and the parent soil. Additionally, over oceans particles ejected into the atmosphere during bubble bursting through the sea surface microlayer, which is enriched in organic matter, are considered as the major precursors of INP and also terrestrial bioaerosols such as fungi and bacteria that have been shown to have INP activity are transported over the ocean and contribute to the INP in the marine environment.

In the present study, we further develop the global 3-D chemistry transport model TM4-ECPL to account for INP concentrations in the atmosphere using state of the art parameterizations. TM4-ECPL simulates INPs concentrations derived from K-rich feldspar and quartz particles using two different soil mineralogy atlases (Claquin et al. (1999) and Journet et al. (2014)) based on brittle fragmentation theory (Kok, 2011) and INP from marine and terrestrial bioaerosols. The simulated distribution of INP concentrations globally agrees with currently available ambient measurements (BACCHUS and Wex et al., 2019 databases). The analysis highlights how different soil mineral composition datasets impact the ice initiation and to what extent a better quantification of dust mineralogy may improve ice nucleation representation in modelling. Also, it is found that INP from desert dust dominates the concentration of INP over the entire Northern Hemisphere, terrestrial bio-aerosols contribute to INP concentration mainly close to the emission sources, while marine organics are important contributors to primary ice crystals over remote oceans depending on marine biota, which varies seasonally. Finally, to understand the impact of these findings on climate and their feedback on clouds, we applied these well documented INP parameterizations derived from dust and marine organic aerosols to Earth-System Model (EC-EARTH-3) providing early results in radiative forcing variation.