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The Role of Surfactants on Cloud Formation: Surfactants in PM1 Aerosols from Urban to Remote Regions and Correlations with Cloud Occurrence

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Surface tension is a key parameter in the Köhler equation describing cloud droplet formation, but which has been ignored for many years, mostly because of the lack of information on the surfactants present in aerosols and their effects on the surface tension. For this reason, we have recently developed methods to extract the total surfactant fraction from aerosols (= sum of all the components affecting their surface tension), measure their concentrations, and determine the overall surface tension isotherm of the aerosols. This work presents the application of these methods to PM1 aerosols from different regions; Lyon, France (urban site, 55 samples), Rogoznica, Croatia (coastal site, 17 samples), and Pallas, Finland (remote site, 237 samples), and the first results on their surfactant properties and how much they might contribute to cloud formation.
The results show that, in spite of large differences in the concentration and size distribution of aerosols in these different regions, the average molar concentrations of anionic, cationic and non-ionic surfactants inside the PM1 particles displayed similar trends: non-ionic surfactants represented 60 to 70 % of the total surfactant concentration, anionic ones 20 to 35 %, while cationic surfactants were negligible. However, large differences were observed in the Critical Micelle Concentration (CMC), the key point of the surface tension isotherm: the CMC of surfactants in remote aerosols was nearly 10 times lower than that in urban aerosols (1.7 x 10-4 M and 9.3 x 10-4 M, respectively) evidencing very different molecular structures and the greater cloud-forming efficiency of the surfactants from remote regions.
In a second study, the potential role of surfactants on cloud formation was explored by comparing the analysis of the surfactants in PM1 aerosols (237 samples) with cloud occurrence over 8 months (257 clouds) at the remote Pallas Supersite of the Finnish Meteorological Institute in Finland. Statistical analyses (Canonical Correlation Analysis, CCA, and regression analyses) were applied to the data and revealed strong co-dependencies between the surfactant properties (ratio of concentration over CMC, C/CMC, quantifying the surfactant efficiency) and cloud frequency. As no such co-dependency was found between the surfactants and any of the other cloud-relevant variables (temperature, relative humidity, aerosol particle radius, and hygroscopic composition) these results suggested a direct, physical connection between the surfactants properties in PM1 and cloud properties. This connection was further confirmed by time-dependent analyses showing that, over 48h-periods, each increase in surfactant efficiency coincided with an increase in cloud frequency. These results are the first atmospheric evidence for a causality relationship between surfactants in PM1 aerosols and cloud formation.