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Cluster analysis of organic molecules in an alpine ice core: the transition from the pre-industrial to the industrial era

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The effects of atmospheric aerosol particles on the Earth's radiative balance are a major source of uncertainty in global climate models. A distinction and quantification between natural and anthropogenic atmospheric aerosol concentration and their sources has to be made to reduce this uncertainty. Therefore, the natural pre-industrial aerosol concentration of the atmosphere must be determined. Ice cores are climate archives that enable the reconstruction of past atmospheric composition changes.

For such a reconstruction, an ice core from the Swiss Alps, which covers the years from 1682-1985, was examined for secondary organic aerosol (SOA) compounds. A non-target analysis (NTA) was used to determine the chemical composition of small organic molecules in the ice. The analytical method of the melted ice samples is based on solid-phase extraction, liquid chromatography and high-resolution mass spectrometry. The result of the NTA showed more than 630 features statistically different from the blank. A hierarchical cluster analysis was performed, in which compounds with a similar trend over time were grouped (clustered) together. The cluster analysis separated the considered features into two main groups. The first cluster showed a good correlation with the dissolved organic carbon concentration (DOC) of non-fossil origin ($R = 0.75$) while the second main group correlated excellently with the fossil DOC ($R = 0.95$), attributed based on the radiocarbon content. This leads to the presumption that compounds represented in the first cluster originated from biogenic sources while compounds in the second cluster are anthropogenic emissions or SOA formed by anthropogenically emitted precursors. This hypothesis is supported by the temporal trend of the two groups. The potential biogenic compounds show a relative stable behavior throughout time. At the beginning of the 20th century a decrease of biogenic SOA is recorded. No compounds from the anthropogenic cluster were detected in pre-industrial times, they increase slowly from 1800 and more and more from 1900. Based on the division into the two main clusters, a detailed graphical evaluation of their chemical composition was performed. We show that the suspected biogenic cluster consists mainly of oxidation products of volatile organic compounds (VOC). The presumed anthropogenic cluster consists mainly of organosulfates, nitrooxy-organosulfate, aromatic compounds and mono- and

dinitroaromatics.