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Isotopic mass independent signature of black crusts: a proxy for atmospheric aerosols formation in the Paris area (France).

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In view of the negative forcing of the sulfate aerosols on climate, a more accurate understanding of the formation of these particles is crucial. Indeed, despite the knowledge of their effects, uncertainties remain regarding the formation of sulfate aerosols, particularly the oxidation processes of S-bearing gases. Since the discovery of oxygen and sulfur mass independent fractionation (O- and S-MIF) processes on Earth, the sulfate isotopic composition became essential to investigate the atmospheric composition evolution and its consequences on the climate and the biosphere. Large amount of S-bearing compounds (SO_2 mainly) is released into the atmosphere by anthropogenic and natural sources. Their oxidation in the atmosphere generates sulfate aerosols, H2SO4, which precipitate on the earth surface mainly as acid rain. One consequence of this precipitation is the formation of black crust on buildings made of carbonate stones. Indeed the chemical alteration of $CaCO_3$ by H2SO4 leads to gypsum ($CaSO4{\cdot}2H_2O$) concretions on building walls. Associated to other particles, gypsum forms black-crusts. Therefore, black crusts acts as 'sulfate aerosol traps', meaning that their isotopic composition reveals the composition and thus the source and formation processes of sulfate aerosols in the atmosphere in a specific region.
In this study we collected 37 black crusts on a 300km NW-SE profile centered on Paris (France). In our samples, sulfate represent 40wt.% and other particles 60wt.% of the black crusts. After sulfate extraction from each samples we measured their O- and S-isotopes composition. Variations of about 10‰ in δ18O and δ34S are observed and both O-MIF (Δ17O from 0 to 1.4‰ and S-MIF (Δ33S from 0 to -0.3‰ compositions have been measured.
In regards to these compositions we can discuss the source and formation (oxidation pathways) of the sulfate aerosols in troposphere above the Paris region that covers urban, rural and coastal environments. Furthermore, this study shows for the first time O- and S-MIF signature in black crusts. Finally, we demonstrate that black crusts can be considered as a good 'sulfate aerosols traps', which can be widely used to study the sulfate aerosol formation, fate and sink in the troposphere.