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Insights into the atmospheric cycle of arsenic: Linking elemental speciation, organic composition with atmospheric transport in a 5-year time series of aerosol measurements

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Most studies on arsenic (As) in the environment have primarily focused on contamination of groundwater and soils. The atmosphere is, however, also an important environmental compartment to investigate because atmospheric deposition is a source of this toxic element to terrestrial and marine environments, affecting ecosystems and human health. While previous global atmospheric As budgets estimated that the majority of atmospheric As is of anthropogenic origin, recent studies suggested important contributions of biogenic sources. However, these biogenic contributions and their origins (e.g., marine versus terrestrial emissions) remain poorly characterized.

For many other elements isotopes can be used as source tracers to better understand atmospheric contributions but this is not possible for As as it is monoisotopic. However, as anthropogenic and biogenic activities emit specific elemental species and organic compounds, the combination of these can be used as distinct atmospheric markers of emissions and (bio)chemical transformations during atmospheric transport. Furthermore, atmospheric As speciation is not only important for identifying emission sources and atmospheric transformations but it also determines the fate of As that is deposited to surface environments, e.g., its mobility, bioavailability for plant uptake and/or toxicity.

In this study, we investigated the concentration and speciation of As in a unique series of aerosol samples (n=134) collected weekly between 2015 and 2020 at Pic du Midi Observatory (French Pyrenees; 2877 m.a.s.l.). This high-altitude site enables the investigation of local and long-range elemental transport from both marine and continental sources. Arsenic species (i.e., As oxyanions and methylated As) were quantified by HPLC-ICP-MS/MS after extractions with water (water soluble fraction) and nitric acid (total species). In addition to inorganic geochemical data (various element concentrations determined by ICP-MS/MS), we analysed the organic composition (using pyrolysis-GC/MS) and estimated the dominant sources and atmospheric transport patterns of sampled aerosols using air parcel backward trajectories using three-dimensional wind fields from the atmospheric reanalysis dataset ERA5.

Based on the combination of these analyses, we will present new insights into seasonal variability of atmospheric As sources and cycling, which are crucial to better understand and predict As toxicity in atmospheric deposition that functions as an As source to surface environments.