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Tracing atmospheric selenium from sources to deposition by combining chemical speciation analyses with modelling

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The atmosphere is an important reservoir for the essential trace element selenium (Se). An estimated amount of 11.9-14.6 Gg(Se) [1] is annually deposited on terrestrial environments, which serves as a source of Se to ecosystems and food chains. The fate of Se that is deposited to surface environments strongly depends on its chemical speciation. However, the factors determining Se speciation in atmospheric deposition remain poorly understood.

Here, we applied different chemical measurements and modelling approaches to aerosol samples (n=134) taken at a weekly resolution over 5 years (2015-2019), as well as precipitation (n=26) and cloud water (n=56) samples taken during a field campaign of two months in 2019 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. Chemical analyses of aerosols and wet deposition included total concentrations of various elements, Se speciation obtained via an improved (ultra)sensitive method based on liquid chromatography and inductively coupled plasma tandem mass spectrometry (LC-ICP-MS/MS), as well as organic molecular composition of aerosols based on pyrolysis-gas chromatography mass spectrometry (Py-GC-MS). These analyses were combined with modelling of Se source contributions using the atmospheric aerosol-chemistry-climate model SOCOL-AERv2, air parcel backward trajectories and Lagrangian moisture source analyses.

The main Se species, selenate (Se^{VI}), is primarily linked to continental sources, while a newly detected organic Se species has been identified as a biomarker for marine-biogenic sources. We show that the combination of speciation with other chemical proxies such as organic markers of emissions sources, as well as trajectory modelling are crucial to disentangle Se source contributions and atmospheric processes. These results represent a new step towards an improved prediction of Se inputs from the atmosphere to terrestrial environments.