## The hydrological cycle of iodine in Atacama: Natural and anthropogenic sources revelead by iodine-129 isotopes

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The Atacama region in northern Chile hosts the driest desert on Earth and is the world's premier source of natural iodine, which is mostly concentrated in the nitrate-rich "caliche" soils. These deposits are unique due to their mineralogical features and extension, with iodine concentrations in excess of 500 ppm. However, little is known about the regional setting and distribution of iodine in Atacama.

We present a comprehensive survey of iodine concentrations and isotopic ratios (129 I/I) of different reservoirs including rocks, soils and waters. Nitrate deposits were found to have iodine concentrations significantly higher than mean values in the earth crust (~700 ppm). In case of fluids, the highest concentrations were measured in groundwater below nitrates in the Central Depression (3.5-10 ppm) and in geothermal fluids in the volcanic arc (1-3 ppm).

In most solid reservoirs, <sup>129</sup>I/I values are below the pre-anthropogenic input ratio of 1500 x 10<sup>-15</sup>, demonstrating that anthropogenic additions are absent in them. Regarding fluids, we identify two main groups. The first one includes the natural waters, which have <sup>129</sup>I/I ratios substantially above the pre-anthropogenic threshold value of 1500 x 10<sup>-15</sup>, indicating the likely presence of anthropogenic <sup>129</sup>I. There is a second group, where the natural waters have <sup>129</sup>I/I ratios lower than 1500 x 10<sup>-15</sup>, which vary between ~215 and ~1300 x 10<sup>-15</sup>.

Geochemical mixing models reveal that the measured <sup>129</sup>I/I ratios are in agreement with multiple sources of iodine, including variable contributions from old organic sources (i.e., marine sedimentary rocks) and younger fluids such as volcanic fluids, pre-anthropogenic and anthropogenic meteoric waters. Our results show that the large variation observed in the isotopic ratios is indicative of significant mixing and circulation of fluids of meteoric, sedimentary and volcanic origin along the Chilean continental margin over scales of millions of years.