## Natural and anthropogenic iodine in natural waters from the Atacama Desert: Sources and cycling of iodine in a continental margin

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The Atacama region in northern Chile hosts the driest desert on Earth and is the world's premier iodine production province. The origin of iodine enrichment in Atacama is controversial, and recent studies have provided evidence that groundwater has played a key role in the remobilization, transport and deposition of iodine (e.g., nitrate-iodine and supergene Cu deposits), although little data is available for natural waters in the region. In order to constrain the natural and potentially anthropogenic iodine inputs in the Atacama Desert and their association with geological and climatic processes, we explore the halogen chemistry and iodine isotopic signature of natural waters in the region.

Iodine and bromine concentrations of surface and ground waters vary from 0.25  $\mu$ M to 48  $\mu$ M and from 1.28  $\mu$ M to 87.38  $\mu$ M, respectively, showing a strong iodine enrichment when compared to seawater concentrations (I = ~0.4  $\mu$ M, Br = ~600  $\mu$ M). In contrast, no bromine enrichment is detected, suggesting the influence of an organic-rich source. On the other hand, the <sup>129</sup>I/I ratios are indicative of different natural and anthropogenic sources for iodine in the studied area. Samples with the lowest isotopic ratios (<sup>129</sup>I/I from ~215 to ~1000 x 10<sup>-15</sup>) strongly suggest mixing between an organic-rich deep fluid and pre-anthropogenic meteoric water, while samples with higher values (>1500 x 10<sup>-15</sup>) indicate the input of anthropogenic iodine in meteoric water and/or seawater.

Considering the geological, hydrological and climatic conditions of the Atacama Desert, we propose two different contributions to explain the anthropogenic <sup>129</sup>I signal. The first source is associated with <sup>129</sup>I releases during nuclear weapon tests carried out in the Pacific Ocean (<sup>129</sup>I/I = ~12000 x 10<sup>-15</sup>); and the second contribution is related to <sup>129</sup>I releases in reprocessing nuclear plants in the Northern Hemisphere (<sup>129</sup>I/I = ~93700 x 10<sup>-15</sup>). Both sources reflect rapid redistribution of <sup>129</sup>I on a global scale as previous studies have suggested. Our results strongly point towards a long-lived continental iodine cycle in the hyperarid margin of western South America, which is driven by local hydrological conditions, producing iodine enrichment and subsequent remobilization and transport in surficial reservoirs.