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## Impact of atmospheric pollution inputs and climate change on dissolved inorganic carbon fluxes in karst aquifers: evidences from a 36 years past monitoring of karstic watersheds.

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Atmospheric pollution is known to modify the soil  $CO_2$  consumption associated with carbonate bedrock weathering. To evidence the long term feedbacks of atmospheric pollution and climate change on this chemical reaction, we investigated the inorganic carbon fluxes monitored weekly from 1979 to 2006 in a small forested karstic watershed in the Pyrénées Mountains, characterized by a large precipitation variability, a 0.025 °C air temperature increase per year and a low agricultural pressure.

The yearly average concentrations of [Ca + Mg] and dissolved inorganic carbon increases of about 0.057 meq.L-1.yr-1 and the 0.1 meq.L-1.yr-1, respectively. The gap relative to the 1:2 relationship between [Ca + Mg] and HCO<sub>3</sub> (in mmole. L-1), noted Delta-HCO<sub>3</sub>, was founded to be driven by the atmospheric pollution inputs, producing strong acids that inhibit the consumption of carbon from soil during the carbonate dissolution processes. In addition, atmospheric temperature increase is correlated with the [Ca + Mg] change, whereas the decrease of the atmospheric acid inputs observed since the seventies, is linked with a + 0.0022 meq.L-1.yr-1 increase in Delta-HCO<sub>3</sub>.

Similar trends in Delta-HCO<sub>3</sub> change were found over other karstic watersheds monitored more recently in the framework of the SNO KARST, one the observatory networks from the OZCAR Research Infrastructure, highlighting that Delta-HCO<sub>3</sub> changes over time were partially controlled by atmospheric pollution inputs. The re-interpretation of hydrochemical databases using this Delta-HCO<sub>3</sub> indicator enables to evaluate better the impact of atmospheric pollution load and climate change on surface waters. In an indirect way, the dephasing between atmospheric loads recorded in precipitation and Delta-HCO<sub>3</sub> observed in groundwater could be a new tracer method to estimate groundwater residence times.