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Experimental determination for kinetic fractionation during solid condensation at low temperature and theoretical framework

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Isotopic fractionation of water enables climate reconstruction from ice cores. The use of different heavy isotopes of the water such as H218O, H217O or HDO gives information about local temperature but also temperature and humidity of water vapour sources. Quantification of these parameters relies on the good knowledge of equilibrium and kinetic isotopic fractionation at each step of the water cycle. The strongest limitation when interpreting water isotopes in remote Antarctic ice cores is the formulation of the isotopic fractionation at solid condensation (vapour to ice). This classical formulation also implies a good knowledge of coefficients for equilibrium fractionation and water vapour diffusion in air as well as supersaturation in clouds. The uncertainties lying on these different parameters make the formulation of isotopic fractionation at solid condensation only empirical.

Here, we make use (1) of recent development in the measurements of water isotopes in the water vapour through infra-red spectroscopy and (2) of the possibility to measure accurately 17O-excess of water to test the classical formulation and parameterization of isotopic fractionation at solid condensation. A first experiment involving very strong supersaturation evidences a strong kinetic effect on 17O-excess on solid condensation, similar to d-excess. It also shows the limits of the classical formulation of water isotopic fractionation during solid condensation estimation at very low temperature. A second experiment performed in a cloud chamber in controlled conditions uses CRDS instruments to depict the spatial variability of water vapour isotopic composition due to diffusion (kinetic effect) during solid condensation. These experiments are in agreement with a new theoretical model that we present for the competition between diffusions of different isotopes.