High-precision measurement of $^{86}Kr/^{82}Kr$ and $^{136}Xe/^{129}Xe$ in air to study convective mixing in polar firn

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Abstract

Trapped gases in polar ice cores have provided unique records of past atmospheric greenhouse gases. To establish the phasing between the gas records and climatic records kept in the surrounding ice matrix, the age difference between air and ice must be known. This age difference arises because the air is trapped at the base of the ~100-m-thick porous firn layer (consolidated snow) and is thus younger than the ice. Unfortunately, the age difference is not well constrained for the past and thus obscures the phasing of temperature and gases. One constraint on firn thickness may come from isotopic fractionation of nitrogen and other inert gases in trapped air. Gravitational and thermal fractionation in the firn layer produces a signal in trapped air that is proportional to the firn thickness and top-to-bottom temperature gradient. However, ice-core $\delta^{15}N$ ($^{15}N/^{14}N$) and $\delta^{40}Ar$ ($^{40}Ar/^{36}Ar$) records from the Antarctic interior (Vostok, Dome Fuji, Dome C) for glacial periods have shown unexpectedly small gravitational fractionation, such that derived firn thickness is thinner by 30-40 m (~30 % of firn thickness) than expected from glaciological models. The discrepancy may be explained if a near-surface "convective zone", where air convection eliminates isotopic fractionation, was extremely well developed during glacial times, but unfortunately there has been no way to estimate the past thickness of it.

Heavy noble gases in the trapped air may solve this problem. Because Kr and Xe have smaller diffusivities than N_2 and Ar, they should be less gravitationary fractionated in deep firn, if strong convective mixing existed. We have developed a method to measure $^{86}Kr/^{82}Kr$ and $^{136}Xe/^{129}Xe$ in air with 1σ uncertainty of less than 0.02 and 0.03 ‰, respectively, and applied it to air samples withdrawn from the firn at a near-zero-accumulation site in the Megadunes region, Antarctica. This site is suitable for verifying the theory because it has a thick convective zone (~20m). When normalized to unit mass difference, $\delta^{86}Kr$ and $\delta^{136}Xe$ are indeed found to be smaller than $\delta^{40}Ar$ by 0.017 ± 0.005 and 0.022 ± 0.005 ‰, respectively, though exact interpretation requires futher measurements to rule out a possible small thermal signal. When applied to ice core analyses, this method should allow us to deduce the past convective zone thickness in the firn at the central Antarctic sites, thereby leading to a better constraint on the phase relationship between changes of greenhouse gases and climate records from the ice cores.