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Histories of atmospheric methane and its carbon and hydrogen isotopic ratios over the last 250 years obtained from an ice core and firn air

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We analyzed air extracted from an ice core and firn air samples for carbon and hydrogen isotopic ratios (δ13C and δD) as well as the CH4 concentration. The ice core was drilled at G15 site at Antarctica, and the firn air samples were collected at YM85, Dome Fuji (DF), Antarctica and North GRIP (NGRIP), Greenland. In firn layers, we observed gradual decreases of the CH4 concentration, δ13C and δD with increasing depth, and rapid decreases were observable below the firn-ice transition layers at the YM85 and NGRIP sites. A one-dimensional diffusion model for firn air was used to reconstruct atmospheric histories of δ13C and δD. Diffusivity in the firn was set so that the model well reproduces the depth profile of the CO_2 concentration at each site and scaling factors were multiplied for the CH4 and its isotopologues. An atmospheric history of the CH4 concentration at Antarctica was prepared according to previous studies and given to the model. Since no reliable atmospheric history of the CH4 concentration at Greenland over the last century is available, we prepared five scenarios of the CH4 concentration with different interpolar gradients. Effective air ages at the respective sampling depths were determined by referring to the atmospheric CH4 histories. Corrections for diffusion and gravitational effects were applied to the isotopic data. Finally, we reconstructed the atmospheric histories of δ13C and δD over the last 250 and 50 years at Antarctica and Greenland, respectively. At Antarctica, the δ13C and δD values had remained unchanged until around 1950 and then increased rapidly by about 2.1 and 8‰ to the present levels. The reconstructed δ13C and δD histories at Greenland showed similar secular trends, but their values were lower than those obtained at the Antarctica over the whole period. The interpolar differences of δ13C and δD would be ascribed to the fact that large portions of biogenic CH4 sources with depleted δ13C and δD signatures exist in the northern hemisphere. We discuss secular changes of the interpolar differences of the atmospheric CH4 and its isotopes as well as trends of CH4 sources that could reproduce the atmospheric histories reconstructed from our data.