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Change in the Northern Hemisphere atmospheric carbon monoxide budget evaluated from CO stable isotopes in Greenland NEEM firn air

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The importance and interest for reconstructing past atmospheric CO arises from its significant role on the chemistry of the troposphere as CO is the major sink for hydroxyl radicals (OH). A previous study (1) has provided a reconstruction of atmospheric CO from Berkner Island (Antarctica) roughly covering the last century, which is important for understanding the past CO budget in the Southern Hemisphere. However, there is limited information about the 20th century CO evolution in the Northern Hemisphere, and none yet from firn air.

We present here a combined NH record of [CO] and its carbon and oxygen stable isotopic ratios, based on measurements in Stony Brook University on firn air samples collected in July 2008 at the NEEM ice core site in Greenland. Our [CO] results indicate a similar trend as those from other laboratories involved in the NEEM project (2). Besides [CO] data, we also present for the first time δ^{13}C and δ^{18}O of CO in NH high-latitude firn air. These isotopic ratios are a proxy for CO source strengths since different sources can have distinct isotopic signatures. δ^{13}C(CO) is a good tracer for CH_4 oxidation, which is depleted by about 20‰ in ^{13}C compared with other CO sources. δ^{18}O(CO) is an excellent tracer for CO from combustion processes, which is enriched by about 10-15‰ in ^{18}O compared with CO from OH-oxidation sources.

The smooth trends with depth of both δ^{13}C and δ^{18}O suggest that there is no contamination in our samples. In addition to the seasonal imprint in the first 10-20 m, our isotopic ratio data indicate isotopically heavier δ^{13}C and δ^{18}O below about 55 m of depth, with a maximum centered around 65 m. The amplitude of this enrichment with depth is larger than expected from gravitational fractionation, and thus reflects isotope diffusion gradients in the NEEM firn and potential atmospheric changes of the CO isotopic ratios. We will show scenarios of changes in atmospheric CO isotopic ratios, deduced from a firn air diffusion model, and able to reproduce the firn air profile. These scenarios will be discussed with respect to the relative contribution of CH_4 oxidation, non-methane hydrocarbon oxidation, biomass burning and fossil fuel combustion, to the concomitant changing [CO] in the NH.

1. Assonov S.S., Brenninkmeijer C.A.M., Jockel P.J., Mulvaney R., Bernard S. and Chappellaz J., Evidence for a CO increase in the SH during the 20th century based on firn air samples from Berkner Island, Antarctica, Atmospheric Chemistry And Physics 7, 295-308, 2007.

2. V.V. Petrenko, P. Novelli, D.M. Etheridge, I. Levin, Z. Wang, T. Blunier, P. Lang, L.P. Steele, F. Vogel, J. Chappellaz, and the other members of the NEEM firn carbon monoxide Team, Records of Northern Hemisphere atmospheric carbon monoxide and hydrogen back to about 1960 AD from Greenland firn air, EGU 2009 Meeting abstract