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Multisite high resolution measurements of carbon monoxide along Greenland ice cores: evidence for in-situ production and potential for atmospheric reconstruction

Xavier Faïn (1), Jérôme Chappellaz (1), Rachael Rhodes (2), Christopher Stowasser (3), Thomas Blunier (3), Joseph McConnell (4), Edward Brook (2), Thibault Desbois (5), and Daniele Romanini (5)

(1) UJF – Grenoble 1/CNRS, UMR5183, Laboratoire de Glaciologie et Geophysique de l'Environnement (LGGE), Grenoble, France, (2) College of Earth, Ocean and Atmospheric Sciences, Oregon State University, Corvallis, OR, USA, (3) Center for Ice and Climate, Niels Bohr Institute, University of Copenhagen, Copenhagen, Denmark, (4) Division of Hydrologic Sciences, Desert Research Institute, Reno, NV, USA, (5) UJF – Grenoble 1/CNRS, UMR5588, Laboratoire Interdisciplinaire de Physique (LIPhy), France

Carbon monoxide (CO) is the principal sink for hydroxyl radicals (OH) in the troposphere. Consequently, changes in atmospheric CO levels can considerably perturb the oxidizing capacity of the atmosphere, affecting mixing ratios of a host of chemical species oxidized by OH, including methane. In addition, CO variations (and changes in its stable isotopic composition) are expected to be good tracers of changes in biomass burning emissions. Investigating past mixing ratios of carbon monoxide is thus a promising approach towards reducing uncertainty related to the past oxidative capacity of the atmosphere and biogeochemical cycling of methane. Recent developments in optical spectrometry (Optical Feedback Cavity Enhanced Absorption Spectrometry, OFCEAS), combined with continuous flow analysis (CFA) systems, allow efficient, precise measurements of CO concentrations in ice cores. Coupling our OFCEAS spectrometer with the CFA melter operated at DRI (Reno, USA) provided the first continuous CO measurements along the NEEM (Greenland) core covering the last 1800 yr at an unprecedented resolution. Although the most recent section of this record (i.e. since 1700 AD) agreed with existing discrete CO measurements from the Eurocore ice core and the deep NEEM firn, it was difficult to interpret in terms of atmospheric CO variation due to high frequency, high amplitudes spikes related to in-situ production (Faïn et al., Climate of the Past Discussion).
During a recent 8-week analytical campaign, three different ice archives from Greenland were melted on the DRI CFA and analyzed continuously for CO with the OFCEAS spectrometer: (i) the D4 core (spanning the last 170 yr), (ii) the NEEM core (extending the existing record from 200 AD to 800 BC), and (iii) the Tunu core (spanning the last 1800 yr). Although in-situ production of CO is observed at all sites, these new records reveal different CO patterns and trends. This multisite approach allows us to better characterize the processes involved in CO in-situ production by evaluating the influence of site-specific factors such as surface accumulation rate (10, 22 and 41 cm ice yr-1 for Tunu, NEEM, and D4 respectively), surface temperature, or aerosols loading (with e.g., median black carbon concentration ranging from 0.9 to 2.3 ng g-1 among the investigated sites). However, a quantitative understanding of the past evolution of atmospheric CO above Greenland remains challenging due to the existence of these artifacts.