Atmospheric History of Carbonyl Sulfide During the 20th century from Antarctic and Greenland Firn Air Measurements

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Carbonyl sulfide (COS) was measured in firn air from multiple field campaigns carried out in Greenland (at Summit in 2006 and 2013) and in Antarctica (at Mega Dunes in 2003 and at South Pole in 2001, 2008, and 2015). Firn air samples were collected in glass flasks and measured at NOAA GMD and UC Irvine laboratories 2 to 6 months after collection. A Monte Carlo-based Bayesian inversion algorithm is used to recover a Northern Hemisphere and a Southern Hemisphere atmospheric history consistent with data from Summit, Greenland and Antarctica, respectively. The Antarctic record constrains atmospheric COS variability during the entire 20th century, while the Greenland record is highly uncertain prior to 1950. The records show that atmospheric COS levels generally increased through the first 70–80 years of 20th century, peaked in the 1970s and 1980s, then declined to near present-day levels. During the rise period, COS levels in the Northern Hemisphere were higher than the Southern Hemisphere except early on, suggesting the source of the increase was in the Northern Hemisphere and likely anthropogenic. There are differences in both the timing and the intensity of the COS peaks in the two hemispheres, with the Southern Hemisphere lagging the north by about ten years and leveling off at lower mixing ratios. The COS peak over Greenland was 550–600 ppt, which is about 20% higher than the atmospheric levels at Summit today. The COS peak over Antarctica was 530–550 ppt, which is about 10–15% higher than the present-day levels. A six-box coupled ocean-atmosphere model is used to explore the implications of the firn air-based COS records on the atmospheric COS budget. Results suggest changes in anthropogenic emissions alone do not explain the atmospheric variability of COS during the 20th century.

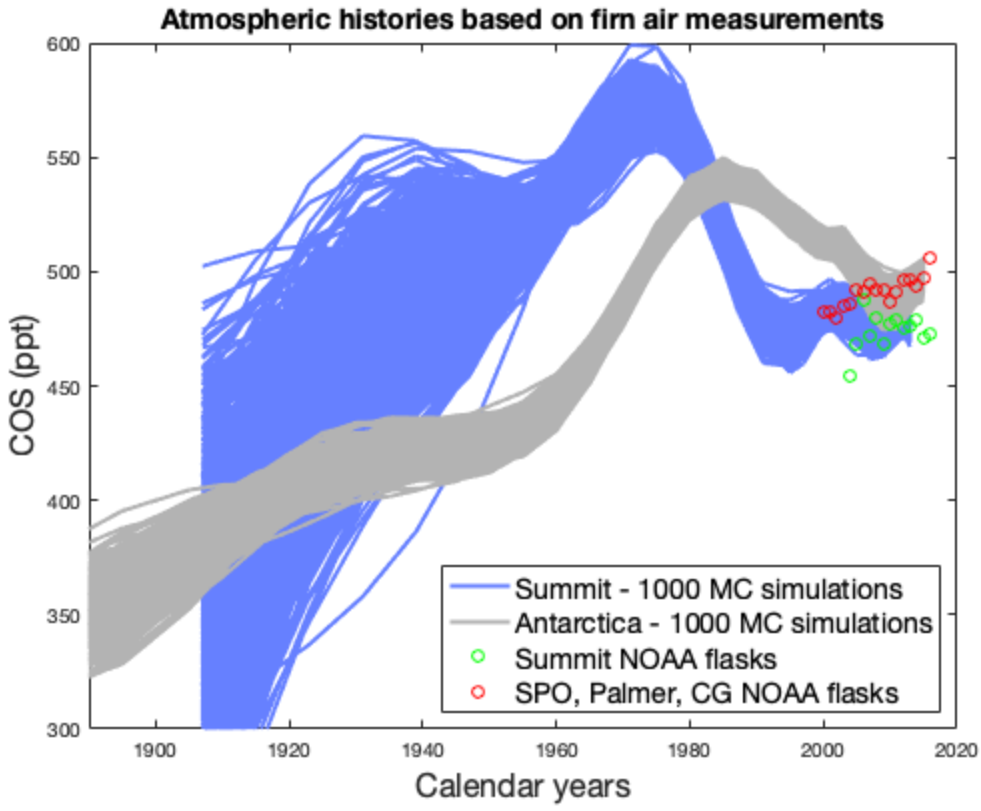

Figure 1. COS atmospheric histories based on firn air measurements from Greenland and Antarctica.