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Variations in tropospheric oxidant levels during the Holocene and late Glacial

Content

The reconstruction of past changes in the temperature, composition and dynamics of the atmosphere is of great interest, also with regard to predictions of future climate change. There are discrepancies between models and climate proxies on temperature variations over the Holocene, and also the temporal evolution of key greenhouse gases like CH4 is not understood. Variations of atmospheric oxidants in the past atmosphere are particularly poorly constrained because they are not stable in any paleo-climate archive. We present measurements of the clumped isotope composition of atmospheric O2 (abundance of 18O18O denoted by Δ36) extracted from a Greenland ice core covering the Holocene and late glacial periods, and in the present atmosphere. The data provide new constraints on upper tropospheric temperatures and oxidant levels in the past. In the glacial period Δ36 was higher than in the Late Holocene, because of the lower temperatures and oxidant levels during this period. Δ36 shows pronounced millennial-scale variability over the Holocene, with Mid Holocene Δ36 values being lower than in the Late Holocene, and even lower compared to present-day air. Simulations of Δ36 in the 3D atmospheric chemistry model EMAC are used to investigate the contribution of changing temperatures, oxidant precursors and atmospheric transport on oxidant levels in the different climate states. Our data suggest a maximum in oxidant levels during the Mid Holocene, consistent with the well-established Mid Holocene CH4 minimum. However, an increase in oxidants alone cannot account for the unexpectedly low Δ36 values in the Mid Holocene. Middle and upper tropospheric temperatures must also have been warmer than today. The Δ36 data imply that a number of key atmospheric processes must have varied considerably over the Holocene, which is generally considered a climatically stable period.

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