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Evidence for an increase in the oxidative capacity of the atmosphere in the late twentieth century

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The hydroxyl radical, OH, is the dominant sink for the majority of trace gases in the troposphere. Thus, it plays a major role in controlling atmospheric chemical composition. However despite this importance there remains much uncertainty as to whether concentrations of OH have changed in the background atmosphere in recent decades. It has previously been reported that recent levels of OH in the troposphere (1997-2008) are well buffered against changes in atmospheric composition (Montzka et al., 2011).

We present two independent records that suggest that there was a significant increase in concentrations of the OH radical in the northern hemisphere during the last two decades of the twentieth century. Measurements from Greenland firn air of the changing ratios of n-butane, iso-butane, n-pentane and iso-pentane were compared using a photochemical clock method. Using these changing ratios we calculate an increase in the chemical processing of the air (i.e. [OH].t) between 1980 and 2000. Assuming t to be constant this provides a semi-quantitative historic record of OH concentrations.

Furthermore, measurements of three alkyl nitrates (also from Greenland firn air), secondary oxidation products of the alkanes, suggest an increase in the [NO]/[HO_2] ratio in the background atmosphere. This could be indicative of increasing NO_x concentrations during this period, which would be consistent with increasing [OH]. These two records are further corroborated by comparison with the long term trend in increasing ozone mixing ratios from background European sites.

Knowledge of historic changes to the oxidative capacity of the atmosphere is fundamental to understanding the atmospheric records of trace gases and to determining historic trace gas emissions using top-down approaches. The results presented here have profound implications for our understanding of atmospheric composition in the past, the present and for predicting the future evolution of the atmosphere.