A Consistent Picture of Inter-Annual Variations in Tropospheric OH during 1998-2006 as Inferred from Observations of Methyl Chloroform, Methane, and Other Trace Gases

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Because OH plays a central role in the photochemistry of the global atmosphere, understanding the variability of global OH on inter-annual and multi-decadal time scales is critical for projecting the chemical and physical state of the atmosphere in the 21st century. Here we analyze NOAA observations of methyl chloroform (CH_3CCl_3) during 1998-2006, a time when atmospheric gradients, emissions, and inter-annual variability in emissions were substantially reduced compared to earlier years. During this unique period, inter-annual variability in CH_3CCl_3 loss frequency and, therefore, mean tropospheric OH burdens should be more directly discernable from the atmospheric methyl chloroform observations than has been possible previously.

The NOAA measurements of CH_3CCl_3, obtained from a global flask-sampling network, show inter-annual changes in the exponential loss frequency of methyl chloroform of <5% (mean = 2.4 ± 1.5%) during 1998-2004. This result implies a similar inter-annual variability in tropospheric OH, which is much smaller than has been inferred from an analysis of CH_3CCl_3 observations in the 1980s and early 1990s. The smaller inter-annual changes suggested for OH in recent years are much more consistent with methane observations, which suggest only small variability in the tropospheric OH burden from year to year. In addition to presenting the implications for OH based on CH_3CCl_3 and CH_4, results for other chemicals whose main sink is oxidation by OH will also be discussed. Some coherence is apparent in tropospheric OH variability inferred from CH_3CCl_3, CH_4, CH_3Cl, and C_2Cl_4, which suggests that a substantial portion of the variability observed for all these gases since 1998 is indeed controlled by OH oxidation rates.

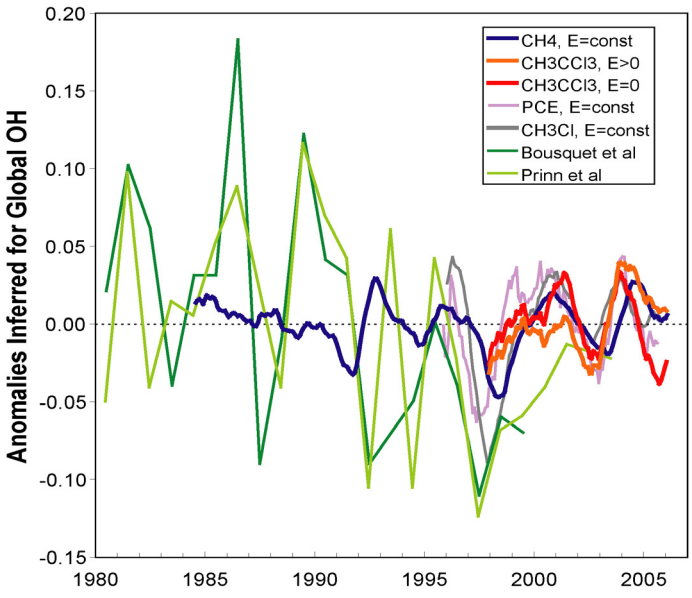

Figure 1. Variations in the hydroxyl radical concentration inferred over time from measurements of CH_4 (dark blue), CH_3CCl_3 (red and orange lines=NOAA; green lines from AGAGE data analyzed by Prinn et al. [2005] or Bousquet et al. [2005]), CH_3Cl (gray, NOAA), and C_2Cl_4 (pink, NOAA). Where emissions are assumed constant (E=const), variations in actual emissions are expressed as variability in OH. Time-dependent CH_3CCl_3 emissions are included in the analyses by Prinn et al. (2005) and Bousquet et al. (2005), and NOAA data (E>0, orange line). These latter studies incorporated a range of models from simple 1-box simulations (NOAA) to sophisticated 3-D models that included inter-annually varying meteorology.