



The Effect of the Interannual Variability of the OH Sink on the Interannual Variability of the Atmospheric Methane Mixing Ratio and Carbon Stable Isotope Composition

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The atmospheric hydroxyl radical concentration (OH) varies due to changes in the incoming UV radiation, in the abundance of atmospheric species involved in the production, recycling and destruction of OH molecules and due to climate variability. Variability in carbon monoxide emissions from biomass burning induced by El Niño Southern Oscillation are particularly important. Although the OH sink accounts for the oxidation of approximately 90% of atmospheric CH_4 , the effect of the variability in the distribution and strength of the OH sink on the interannual variability of atmospheric methane (CH_4) mixing ratio and stable carbon isotope composition ($\delta^{13}\text{C-CH}_4$) has often been ignored. To show this effect we simulated the atmospheric signals of CH_4 in a three-dimensional atmospheric transport model (TM3). ERA Interim reanalysis data provided the atmospheric transport and temperature variability from 1990 to 2010. We performed simulations using time dependent OH concentration estimations from an atmospheric chemistry transport model and an atmospheric chemistry climate model. The models assumed a different set of reactions and algorithms which caused a very different strength and distribution of the OH concentration. Methane emissions were based on published bottom-up estimates including inventories, upscaled estimations and modeled fluxes. The simulations also included modeled concentrations of atomic chlorine (Cl) and excited oxygen atoms (O(1D)). The isotopic signal of the sources and the fractionation factors of the sinks were based on literature values, however the isotopic signal from wetlands and enteric fermentation processes followed a linear relationship with a map of C4 plant fraction. The same set of CH_4 emissions and stratospheric reactants was used in all simulations. Two simulations were done per OH field: one in which the CH_4 sources were allowed to vary interannually, and a second where the sources were climatological. The simulated mixing ratios and isotopic compositions at global reference stations were used to construct more robust indicators such as global and zonal means and interhemispheric differences. We also compared the model CH_4 mixing ratio to satellite observations, for the period 2003 to 2004 with SCIAMACHY and from 2009 to 2010 with GOSAT. The interannual variability of the different OH fields imprinted an interannual variation of the atmospheric CH_4 mixing ratio with a magnitude of ± 10 ppb, which is comparable to the effect of all sources combined. Meanwhile its effect on the interannual variability of $\delta^{13}\text{C-CH}_4$ was minor (< 10%). The interannual variability of the mixing ratio interhemispheric difference is dominated by the sources because the OH sink is concentrated in the tropics, thus its interannual variability affects both hemispheres. Meanwhile, although the OH plays an important role in the establishment of an interhemispheric gradient of $\delta^{13}\text{C-CH}_4$, the interannual variation of this gradient is negligibly affected by the choice of OH field. Overall the study showed that the variability of the OH sink plays a significant role in the interannual variability of the atmospheric methane mixing ratio, and must be considered to improve our understanding of the recent trends in the global methane budget.