Atmospheric Methane and its Isotopic Composition in a Changing Climate: A Modeling Study

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Methane is after carbon dioxide (CO_2) the second most important anthropogenic greenhouse gas (GHG) and implies, due to its relative short lifetime, an attractive mitigation potential. However, variations in methane (CH_4) growth rates are still poorly understood, with uncertainties in both, sources and sinks of CH_4.

We present a comprehensive modeling study using the ECHAM/MESSy Atmospheric Chemistry (EMAC) model [Jöckel et al. 2010]. The study covers four aspects of atmospheric CH_4, namely its sinks, sources, global distribution and climate effect, to provide an improved understanding of the interactions of atmospheric CH_4. This includes, first, the sink processes of CH_4, which are studied by means of the main tropospheric sink reaction partner, the hydroxyl radical (OH). We analyzed 16 simulations concerning the CH_4 lifetime, deriving an average of 8.11 a. Our results indicate that the CH_4 lifetime is not constant and strongly relates to OH abundance and also temperature, which are determined by the model set-up and climate scenario. As a second step, focusing on the sources of CH_4, an inverse optimized emission inventory derived by the fixed-lag Kalman Filter is presented. In our analysis it becomes evident that the uncertainty in the sink of CH_4 limits the certainty of estimated CH_4 emissions. The third part investigates the global distribution of CH_4, which involves the analysis of mixing and transport of CH_4 from specific sources. For this, the model is extended concerning the simulation of CH_4 isotopologues. It is now possible to track the isotopic signature of CH_4 from its source to the end products, which is expected to improve the estimation of CH_4 sources by simultaneously accounting for CH_4 sinks. Lastly, we investigate the oxidation products of CH_4. In order to do so we re-evaluate the assumption that two H_2O molecules are produced per oxidized CH_4 molecule. Our analysis comprises three different approaches, focusing primarily on the tropical region. They agree that the yield of H_2O from CH_4 oxidation is smaller than two in the lower stratosphere and upper mesosphere and potentially attains a value above two in between. We conclude that assuming a constant chemical yield of H_2O from CH_4 oxidation neglects vertical variations in chemical kinetics as well as the chemical loss of H_2O.

In summary, we emphasize the decisive linkage of sources and sinks of CH_4 with respect to their uncertainties and features first steps towards a comprehensive global simulation of CH_4 and its isotopologues for further analysis of these.

Key words: methane, lifetime, hydroxyl radical, emissions, Chemistry-Climate modeling

References

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