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Relationship between atmospheric methane lifetime, isotope budget and effective sink enrichments simulated in AC-GCM EMAC

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In his note adamant for interpreting paleoclimate isotope-resolved CH_4 records, Tans [1] has emphasised the large disparity in the timescales of abundance and isotope ratio changes in the atmospheric CH_4. Derived using a simple two-box model, quantitatively this result is consistent for hemispherically average (homogeneous) CH_4 emitted and removed by yet homogenous and invariable sources and sinks. However, neither the abundance of methane nor its sources and sink rate (determined largely by OH and temperature) are spatiotemporally even. The situation is further complicated by non-linear convolution of photochemistry and mixing/transport acting between source regions and a regarded location. Compared to about 10 years on average in the troposphere, local CH_4 lifetime varies from 15 months (near the surface in tropics) to hundreds of years at high latitudes in winter. How does the local isotope enrichment of CH_4 (resulting from sink fractionation processes) correspond to that? Will using a realistic atmospheric model indicate importance of the abovementioned issues, and for which paleoclimate records?

Inspired by these questions, we designed a similar to [1] experiment implemented, however, in the 3D AC-GCM model EMAC [2; 3] which resolves $^{13}C/^{12}C$ and $^{2}H/^{1}H$ isotope chemistry, $^{14}CH_4$ abundance and methane photochemical sinks including reactions with OH, $O(^1D)$, Cl with respective kinetic isotope effects up to the middle atmosphere (about 80 km). We simulate long-term equilibration of CH_4 abundance and isotope ratios for several emission magnitudes/distributions and OH fields, subsequently perturbed by the pulse change in source strengths or isotope signatures. The resulting sensitivities of effective $^{13}C/^{12}C$ and $^{2}H/^{1}H$ enrichments in atmospheric methane ($^{13C}\varepsilon$ and $^{2H}\varepsilon$, respectively) are important for gauging the isotope signatures of CH_4 sources derived for present and from paleo-records of CH_4. The simulated hemispheric difference in $^{13C}\varepsilon$ correspond to that of [1] when averages are used, however differences in local values ($e.g.$ between the N and S poles) may reach double of that. We find that surface ε values can be parametrically derived using local and average tropospheric CH_4 mixing ratios, however not lifetimes. Importantly, the effective enrichment signal is lost if the lower boundary condition (so-called "nudging") is used instead of surface CH_4 emissions in the model. Such will likely lead to wrong estimates of the isotope signatures of CH_4 sources in inverse modelling approaches. Some conclusions and quantitative estimates of $^{2H}\varepsilon$ are presented in addition.

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