

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

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Abstract

We present model experiments similar in design to that of Tans^[1], implemented in the 3D AC-GCM model EMAC^[2,3] adapted for simulating methane isotope kinetic chemistry on decadal scale. We investigate atmospheric distributions of CH₄ and its potential ²H/¹H, ¹³C/¹²C and ¹⁴C isotope enrichments resulting only from photochemical sinks of methane up to the middle atmosphere (about 80km). Several emission/boundary conditions/sink distributions are being probed. Our results suggest that differences between the simplified (box-model) and full-fledged (AC-GCM) approaches may be important when dealing with isotope-resolved budget of CH₄ in the present and the past.

EMAC Model setup and experiments

We employ the ECHAM5/MESy Atmospheric Chemistry general circulation model (EMAC^[2,3]). To handle the isotope chemistry, the isotope tagging technique^[4] for the kinetic chemistry sub-model MECCA^[4] is applied (kinetic scheme and isotope effects are listed on the right). The setup includes several CH₄ counterparts whose emission/sinks are simulated in the same dynamic setup. The kinetic effects in sink reactions are simulated explicitly for each CH₄ counterpart-isotopologue. We apply additional diagnostic to infer strengths of each photochemical sink pathway. Presented here are the results from simulation period of 75 years (equilibration, phase 1). The model is initialised without CH₄ in the atmosphere. The details of the model/ experiment setup are given in Table 1.

Table 1: Model experiments with EMAC simulated in this study			
Abb. Description / Ref. with REF	Sinks, Isotope, isotope effects (KIE)	Emissions	Tracer counterparts
REF Reference setup, studying effective ¹³ C/ ¹² C, ² H/ ¹ H, ¹⁴ C isotope fractionation/eqilibration	OH, O(1D), Cl, hv monthly averages for 2000	see notes 1*	
ZERO Studying effective fractionation explicitly	REF + MOD KIEs for ¹⁴ C, based on ¹³ C/ ¹² C KIEs for O(D) and O(1D) are set to 0% (100 μMε is kept)	REF + emission signatures for O(D) and O(1D)	CH4Z
MOD Studying the importance of OH/hv diurnal cycle (yielding same monthly averages)	OH, O(1D) and hv sinks are diurnally modulated (REF)		CH4m, CH4n, CH4m
NUDGE Studying the effects of nudging CH4 mixing/isotope ratios at the surface (similar to FI abundance ^[5])	CH4 is prescribed (nudged) at the surface		CH4n
SCD Studying the effect of applying OH climatology from Spivakovsky (20y)	REF + alternative OH climatology ^[6] is used (original)		CH4d0, CH4d1
TCS Studying the effect of applying OH climatology from Spivakovsky (TandCH)	SCD + zeros in the original OH climatology ^[6] are removed		CH4c, CH4c
TANS Reiterating experiment ^[1] Phase 1 (equilibration) in AC-GCM	Only OH sink	4.5.*	CH4L, CH4c

* Simulated CH₄ flux from EMAC ESOMD eval. sim. ¹⁴C is used, -220 Tg(CH₄)/yr
 * Following emission signatures are used: 0°C=50‰ [VP06], 0‰=-20‰ [VSM06], Δ¹⁴C=100 [pMC]
 * Emissions from REF at latitudes north of 30° are used, total flux is scaled to yield 250 Tg/yr
 * See Fig 1

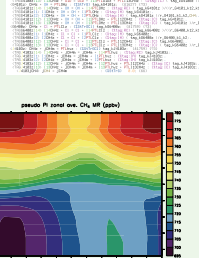


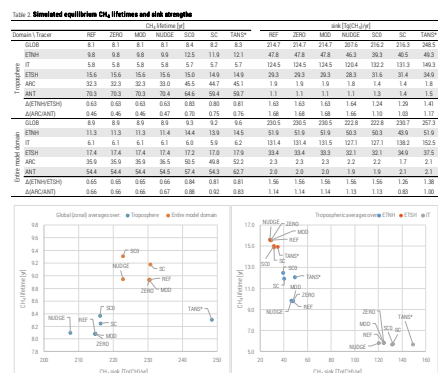
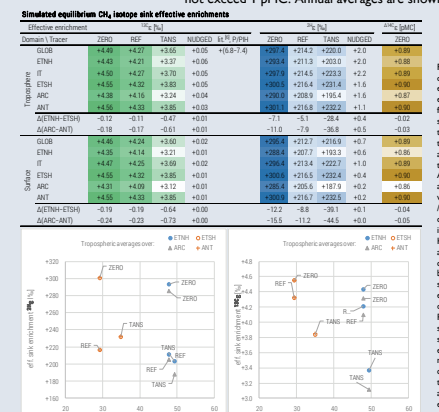
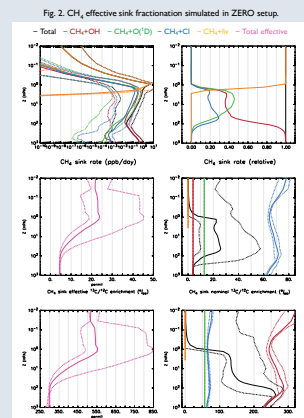
Fig. 2. Pseudo-preindustrial (PI) surface CH₄ mixing ratios used in NUDGE setup of this study.

Questions

- Estimates obtained with simplified atmospheric box-models (e.g. hemispheric boxes) are quantitatively consistent with hemispherically average (homogeneous) CH₄ 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. How different may be sinks simulated in the box-model and in an AC-GCM? The situation may be complicated by the non-linear convection of photochemistry and mixing/transport acting between source regions and a regarded location. Therefore, with this study we are interested in the following aspects:
 - Local CH₄ lifetimes are not homogenous in the troposphere. How does that affect CH₄ local/global equilibration times?
 - How does the local isotope enrichment of CH₄ (resulting from sink fractionation processes) correspond to that?
 - Is there a distinct relationship between CH₄ lifetime, sink strengths and isotope enrichment? In which domain?
 - Does a more realistic atmospheric model indicate importance of the abovementioned issues? How important are these for present-day and paleoclimate records of isotope CH₄?

Sink isotope fractionation in CH₄

Fig. 2 shows effective (resulting from chemistry, transport and mixing in the atmosphere) and nominal (corresponding to KIE) sink fractionations simulated in ZERO setup. For sinks and nominal fractionations, the decomposition by sink pathway is presented. Annual zonal averages are shown. Two features deserve attention: Tropospheric effective enrichment is primarily determined by the reactions with OH radicals. Stratospheric sinks are dominated by photolysis (has no/unknown KIE), so above 1 hPa average fractionation does not vary significantly with altitude. Between 100hPa and 1hPa, sinks via O(D) and Cl radicals compete with the OH channel, which results in increase in ¹³C/¹²C and ²H/¹H effective fractionation by some 20‰ and 250‰, respectively.



Correspondence between sink fluxes and lifetimes

Table 2 lists sink strengths and corresponding CH₄ lifetimes simulated in each setup. Larger (dis)equilibrium between sources and sinks results in (in)dependence of the local lifetimes on the local sink (averages), as contrasted by all setups in tropics(TT) and ET, respectively. It is noteworthy that global/lifetime does correlate with sink strengths but not with effective enrichments in the same domains. Similarly, hemispheric ratios of lifetimes and sink correlate less than may be provided by a model employing hemispheric boxes. Such may be problematic for models using isotope-resolved CH₄, and its lifetimes as inversion constraints, for example^[9].

Fig. 4. Simulated CH₄ lifetimes as a function of sink strengths (annual averages over given domains are shown).

Equilibration times

Fig. 5 Shows the time series of CH₄ mixing and isotope ratios reduced to the "equilibrium" value (average of the last simulated year) in REF, TANS and NUDGE in various domains. The numbers quoted indicate the diagnosed time period required for simulated CH₄ to reach the "equilibrium" value within 1% from scratch. Generally, remote SH needs several years more to equilibrate (cf. GRIP and Vostok sites in TANS). Despite that nudged CH₄ typically needs about 1–2 yrs to equilibrate, the isotope ratios need 3–5 times longer, which is greater than in setups employing CH₄ emissions.

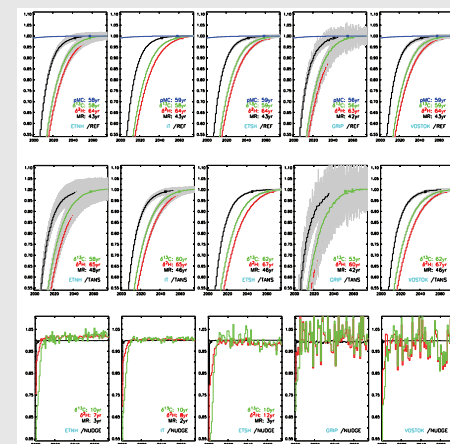


Fig. 5. Time series of CH₄ mixing/isotope ratios equilibrating at given locations/domains simulated in selected setups. Except monthly averages shown in grey colour, values are annual averages.

References

- Tans, P., Glob. Biogeochem. C., 11, 77-81, 1997.
- Jöckel, P., et al., Geosci. Model Dev., 3, 717-752, 2010.
- Gromov, S., et al., Geosci. Model Dev., 3, 337-364, 2010.
- Sander, R., et al., Atmos. Chem. Phys., 5, 445-450, 2005.
- Spivakovsky, et al., J. Geophys. Res. Atm., 105, 8931–8980, 2000.
- Jöckel, P., et al., Geosci. Model Dev., 9, 1153-1200, 2015.
- Schaefer and Whiticar, Glob. Biogeochem. C., 22, GB1001, 2008.
- Fischer, H., et al., Nature, 452, 864-867, 2008.

S. Gromov acknowledges the support of the German Federal Ministry of Education and Research (BMBF) within the framework of PALMOD project (see also www.palmod.de)

