

***Interactive comment on* “The isotopic composition of methane in the stratosphere: high-altitude balloon sample measurements” by T. Röckmann et al.**

Anonymous Referee #2

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The manuscript by Röckmann et al. presents a new data set of the isotopic composition of methane from stratospheric balloon samples. This new data set significantly extends and augments the previously available observations and thus opens new opportunities to study the related kinetic isotope effects. The manuscript discusses in detail the relevant transport and chemistry related processes.

The manuscript is rather long and the presentation style is somewhat unusual for a scientific paper, reminding me more of a lecture script. Many aspects of relevant processes are discussed, often with idealized examples. While in general there is nothing wrong with this presentation style, in many cases it was not clear to me what can be

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learned from the discussion and how the idealized examples apply to the data set. This is also reflected by the fact that the Abstract and Conclusions of the manuscript contain very little concrete and quantitative results, but rather summarize the discussion. On the other hand, relatively little discussion is devoted to the measurements and data analysis itself.

In summary I believe that the manuscript can be significantly shortened, focusing more on the discussion of the new data set, its quantitative analysis and a discussion of the new results that can be derived from this data set.

Specific comments:

p.12042, l.15: define TTL

p.12042, l.26: I have difficulties with the statement "homogenized globally". This is, as you later acknowledge, an idealized picture and will not hold in the presence of mixing barriers, the polar vortex and the edge of the tropical pipe being just two examples.

p.12043, l.8: what is the meaning of "thermodynamic" here?

p.12043, l.15: the word "disturbed" seems slightly out of place here as for an undisturbed dynamical situation you would get "disturbed" mixing.

p.12047, l.19: what exactly is the meaning of a "single line" in this context?

p.12048, l.4: I don't understand the statement that CH₄ has only a small trend. Is not the observed CH₄ trend over the past decades in the same order of magnitude as the CO₂ trend? In what sense is this trend small?

Section 5.2: I'm having difficulties to understand the fundamental difference between diffusive mixing and two-end-member mixing. Is two-end-member-mixing really "a second type of mixing" (p.12054, l.1), or can you derive diffusive mixing as a limiting case from the two-end-member mixing? I.e., is it possible to understand the relation of the $f=0.5$ for the diffusive mixing to your results shown in Fig.4? Are the results of Fig.4 just

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an illustrative example, or can we learn something quantitative from it for the analysis of your data set?

p.12054, eq.(12): what is "m"?

p.12058, l.7: There are chemical sources of N₂O in the stratosphere, although in most cases they can be neglected

p.12058, eq.(16) and eq.(18): please give units

p.12059, eq.(19): please give appropriate units for the parameters k

p.12059, eq.(20): what is VPOB and VSMOW?

p.12065, l.17: Can you give a reference for the statement that models have a poor representation of transport in the tropical stratosphere?

Section 7: I found most of the discussion in Section 7 rather confusing. It is explained where the limitations of different approaches are, but it did not become clear to me if any robust result can be derived from the data. How critical do the results depend on the assumed sink strength (eq. 21)?

Appendix A: I'm not sure if I fully understood what has been done here, but I believe much of the reasoning can be simplified: It is basic text-book knowledge that the mass of air (per unit area) between two pressure surfaces is constant, so I don't see why you need to introduce (A_2) and integrate it.

Technical corrections:

p.12040, l.19: remove bracket after "O(1D)"

p.12044, l.5: insert closing bracket "Geophysik"

p.12047, l.4: include reference to the relevant section

p.12048, l.21: remove bracket after "samples"

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p.12058, eq. (18): insert space before r-squared

p.12063, l.13: remove "and" after "consequently"

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12039, 2011.

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