

Reply to the comments of referee 2:

We thank the referee for the constructive review of our paper. Please find our replies below

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 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.

Reply: In response to these comments and the comments from referee 1, the paper has been significantly streamlined, but we prefer to keep several of the general parts in the paper, since these aspects have to our knowledge not been discussed in the isotope literature.

Specific comments:

p.12042, l.15: define TTL

OK

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.

Reply: This will be rewritten accordingly.

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

Reply: This will be rewritten in the process of shortening this introduction.

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

Reply: We agree that this is unclear and it will be reworded.

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

Reply: What we mean to say is that there are no oscillations of large variations in a single dataset, and we agree that wording can be improved here.

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?

Reply: The increase of CH₄ had almost leveled off during this period, so the trend was in fact considerably smaller than for CO₂.

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"

Reply: This point was highlighted also in the review of referee 1, and the terminology and explanation will be refined. We refer to mixing of air masses with different chemical characteristics across mixing barriers like the polar vortex.

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

Reply: The reason of this example is to show that values of $f < 0.5$ can actually occur. This will be discussed more clearly in the revised manuscript.

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

Reply: The entire section will be shortened and the equation will be removed in this process.

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

Reply: This will be added

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

OK

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

OK

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

Reply: Will be spelled out

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

Reply: Reference will be added.

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 (A2) and integrate it.

Reply: This section will be thoroughly revised following the recommendations from the other referee. Appendix A will be removed in this process.

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"

OK

p.12058, eq. (18): insert space before r-squared p.12063, l.13: remove "and" after "consequently"

OK