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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fractional release factors of long-lived halogenated organic compounds in the tropical stratosphere" *by* J. C. Laube et al.

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General author response

We would like to thank the referee for his constructive critique which helped to improve this manuscript significantly. The issues brought up in the general comment are addressed in the following responses to the specific comments. Special attention was given to the limitations of the data set with respect to the possible influence of temporal and spatial changes in the text ("The limitations of these correlations lie in the temporal and spatial span of the available data set. . .") as well as in the conclusions ("For further refinements studies are needed to assess the influence of temporal and spatial stratospheric variations such as the quasi-biennial oscillation.")

Referee comments

P 20284 L 11 imprecise could be understood as following from instrumental errors etc.

P 20284 L 9ff Moreover ... These statements are too strong in my opinion. ODP and FRFs are used typically as an approximate representation of ozone photochemistry.

Author response

The phrase (". . .revealed the latter to be imprecise measures. . .") was reformulated to "revealed the limitations of these measures". The word "thus" was removed from the last sentence of the abstract in order to disconnect it from the previous statement. Nevertheless in our opinion the data set could indeed be used to improve future ozone level and climate projections.

Referee comment

P 20284 L 15 enhance it's not the halocarbon itself which take part in catalytic cycles

Author response

The sentence was expanded (". . .via the products of their chemical degradation.").

Referee comment

P 20284 L 15 paragraph The argument is too general. The paragraph can be combined with the following one and formulated in a more concise way.

Author response

We do not agree with the referee here and do not wish to shorten the explanation of the importance of the presented work.

Referee comment

P 20284 L 21 very few, ff. This is not true when taking into account the great number of satellite observations by HALOE, MIPAS, CRISTA etc.. Even the number of different

CFCs observed by the satellites is not really small. The special advantage of the in-situ WAS observations (much more completeness, high precision, vertical resolution) should be pointed out, but in view of their limited coverage in space and time.

Author response

The section was slightly modified in order to be fairer with the satellite community. ("Very few non-satellite measurements. . ."; "Satellite instruments provide higher spatial and temporal coverage but have only limited precision. . .").

Referee comment

P 20285 L 6ff paragraph the equation and definition should be given first. The FRFs should be put in the context of ODP and GWP estimation here, see first paragraph on page 20287.

Author response

The definition was already given first and still is. We do not agree with the referee, that changing the position of the formula would improve the manuscript. In order to explain the importance of FRFs to the reader as soon as possible two sentences ("Global Warming Potentials (e.g. Daniel et al., 1995) and the. . .") were moved and follow just after the definition.

Referee comment

P 20287 L 9 These FRFs ... Please specify how this can be accomplished

Author response

A reference was added for clarification (Newman et al., 2007).

Referee comment

P 20287 L 12 parameterization what do you mean by parameterization of CTMs, and how can CTMs predict future ozone??

Author response

The phrase "parameterisation and evaluation" was replaced by "validation". The example of CTMs and the corresponding references were removed. A further reference and a sentence connected to it were added as a different example: "Douglass et al. (2008) for instance showed that those model simulations that include realistic mean age and FRF distributions predict longer atmospheric lifetimes for chlorofluorocarbons."

Referee comment

P 20288 L 12 The whole air do you mean "all" samples from both platforms?

Author response

Yes. The first word was changed to "All" for clarification.

Referee comment

P 20288 L 18 procedure Can you specify the differences to Schauffler and how this could influence the results? The cited references give the impression that Engel et al. is not the whole story. As I understand you apply the age spectrum according Engel et al. and the entry mixing ratios according Laube. Perhaps a simple formula would help to clarify.

Author response

Engel et al. is the whole story. A passage was added for clarification: "We calculated FRFs using Eq. (1). The unknown quantity in this equation was . Here, a procedure similar to those of Schauffler et al. (2003) and Newman et al. (2006) was used to derive it." Three references (Laube et al. (2008), Hall and Plumb (1994) and Volk et al. (1997) were removed to clarify, that the calculations were identical to those in Engel et al. (2002). The simple formula for the parameterisation of the age spectrum was added to the text ("As the corresponding age spectra cannot be measured directly they were derived from the mean ages via a parameterisation according to Engel et

al. (2002) using 0.7 as a fixed ratio of the squared spectral width to the mean age."). The other formula (i.e. the calculation of corrected stratospheric entry mixing ratios) is not novel and can be found in this reference. In addition, differences to the method of Schauffler et al. were specified and evaluated ("The parameterisation of the age spectra is different to the one used in Schauffler et al. (2003). We exemplarily recalculated FRFs for flight B42 using two different ratios (0.5 and 1.25) and found all corresponding changes to be below 0.02. Thus we consider the exact parameterisation to have little influence on the FRFs.").

Referee comment

P 20288 L 22 SF6 analysis could/should be part of the section analysis. What about the error of mean age? Is it assumed to be zero and therefore no error bar is shown for mean age in the figures? Does the fitting procedure include the error of mean age?

Author response

The part of the sentence dealing with analysis was moved as requested. The fitting procedure was performed without considering any error bars. For the revised version the following was added to the error bar section: "Uncertainties of the mean ages were derived for the flights B42 and B43 only. First the influence of addition and subtraction of the 1 σ SF6 measurement uncertainty on the mean age was calculated. Second, the mean ages were calculated using different widths of the age spectrum (0.5 and 1.25). The error bars were calculated as the sum of those both errors that had the same algebraic sign and can also be found in the figures."

Referee comment

P20290 L 6ff error analysis Could you give some idea what the main contributions to the errors are?

Author response

The analytical precisions vary from compound to compound and also between differ-

ent measurement days and instruments (i. e. the NOAA instruments). In addition the uncertainties originating from interhemispheric mixing ratio gradients are highly dependent on the individual temporal tropospheric trends of the compounds. In general, the measurement uncertainty contributes most to the error bars except for compounds with strong interhemispheric gradients such as HCFC-22. The only exceptions to this are CCl4 and CH3CCl3 on flight B42. These were the only samples having an additional sample instability error which then was the dominant contributor to FRF uncertainties.

Referee comment

P20290 L 16 But transport... The different correlations are caused by the different photochemical "dose" air parcels experience on different paths through the stratosphere even having the same transit time. Just the existence of transport barriers or the fact that air parcels experience chemistry is not sufficient to explain the differences.

Author response

In our opinion the photochemical dose is already included in the phrase "chemical processes".

Referee comment

P 20288 L 18 analysis Age spectra as derived by empirical studies or by model studies may include some contribution stemming not from irreversible mixing processes but from averaging over air masses of different characteristic. In your sample the tropical balloon flights are the ones where the difference between tropics and mid latitudes is most pronounced (as expected). One may ask how representative these two data sets for the tropical stratosphere are on a global scale.

Author response

The requested discussion of this issue was added to the text ("The limitations of these correlations lie in the temporal and spatial span of the available data set.") as well as to the conclusions ("For further refinements studies are needed to assess the influence of

temporal and spatial stratospheric variations such as the quasi-biennial oscillation.").

Referee comment

P20290 L 23 The correlation. . .What about the QBO? Schoeberl et al., JGR 2008, have analysed QBO amplitude for various substances. As for N2O, also for the shorter lived CFCs QBO would influence transport and to some extent the correlation. The data set presented spans only a small part of the qbo cycle and could therefore include some systematic deviation.

Author response

The requested discussion of this issue was added to the revised version. ("The limitations of these correlations lie in the temporal and spatial span of the available data set. Schoeberl et al. (2008) presented evidence for an influence of the annual oscillation (AO) as well as the quasi-biennial oscillation (QBO) on stratospheric trace gas distributions in the tropical stratosphere from satellite observations. This could potentially change correlations depending on the phase of the respective oscillations. Further studies are necessary to quantify these impacts on long-lived halocarbons in the tropics as well as in higher latitudes."). Another sentence was added to the conclusions ("For further refinements studies are needed to assess the influence of temporal and spatial stratospheric variations such as the quasi-biennial oscillation.").

Referee comment

P20291 L 26 polynomials First, it would be helpful to state also in the text, and not just in the table caption, that the fit functions were derived using data between 22S and 22N only. Second, it is worth to mention (and to explain) that through the fit procedure negative and meaningless FRFs are produced.

Author response

The respective statement was and still is stated in the text as well as in the table caption. As for the second part of the comment: We give validity ranges for the derived

polynomials in Table 2. Within these ranges none of the functions gives negative values. We agree with the referee concerning the fact that negative FRF values can occur. This was and still is already mentioned and explained in the caption of Figure 1 ("Small negative values that occur for low ages and FRFs are caused by atmospheric variability and measurement uncertainties).

Referee comment

P20292 L 4 agreement Inspecting the correlations, data of the Geophysica flights at mean age > 1.5 a obviously are not belonging to the tropical domain. It may be better to leave these data completely out for constructing the correlations.

Author response

The following sentences were added: "The six tropical Geophysica samples with mean ages above 1 year were collected on a survey south between 16 and 22 °S at altitudes between 18 and 20 km and seem to follow the higher-latitudinal correlations. Considering the high mean ages for the collection altitudes these air masses are very likely to be mainly of extra-tropical origin. Thus the respective samples were not considered for the calculation of tropical FRF-mean-age correlations." Calculations, polynomials, validity ranges and figures were changed accordingly.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20283, 2009.