Reply to the comments of reviewer #3

Norbert Glatthor et al.

Reviewer comments are in black, while our replies are in blue.

The manuscript by Glatthor et al. describes the new MIPAS CH4 and N2O data products version 8 from IMK/IAA. The paper is well written and explains the retrieval algorithm and error sources in much detail.

5 I have the following comments.

Thank you for this favourable judgement.

General Comments:

- 1. The description of horizontal gradients in Sect. 3.2 should be clarified. Where does the information
- 10 on horizontal variability for an individual measurement come from? Are only 2D/3D temperature fields used? How exactly are the horizontal gradients of CH4 and N2O mixing ratios "jointly retrieved" (p. 6, l. 15)? Are there additional assumptions?

As stated in the manuscript, fitting of horizontal temperature gradients and of 3D apriori temperature fields is described in detail in Kiefer et al. (2021). The information on horizontal temperature variability

- 15 for an individual measurement comes from the 2D/3D temperature fields used and from retrieval of temperature gradients. The information on horizontal gradients of CH4 and N2O mixing ratios comes from the measurements only. Fitting of CH4 and N2O gradients in the V8-retrievals is performed in a similar way as for temperature gradients, i.e. meridional and longitudinal CH4- and N2O-gradients are retrieved for a range of ± 400 km around the tangent points along with retrieval of their mixing ratios at the tangent
- points. To make things clearer we will replace the sentence on P6, L8ff
 "Instead, we consider horizontal temperature inhomogeneities by inferring a 2D temperature distribution in the plane spanned by the lines of sight of the limb sequence under analysis directly from the retrieved V8 3D temperature fields (cf. Kiefer et al., 2021)."

by

²⁵ "Instead, we consider horizontal temperature inhomogeneities by use of the 2D/3D temperature field and the additional temperature gradient (upper range of the retrieval altitudes only) from the TLOS-retrieval

(see Kiefer et al. 2021).".

Further we will add the sentence

"The fitting of the horizontal CH4 and N2O gradients as unknowns is done by including the respective

- Jacobians in the fitting procedure. These gradients are assumed to be valid for a range of ±400km around the tangent points. No additional assumptions are made."
 after the sentence (P6, L14ff)
 "Instead, the horizontal gradients ..."
- 10 2. The different error contributions are explained in very much detail in the paper and the Supplement. Which of this error information is contained in the data products?

We will change the passage

"For each single CH4 and N2O profile, ..."

into

¹⁵ "For the general user, the individual error due to measurement noise is provided for each CH4 and N2O profile in the data base, along with total random, total systematic and total error. The latter three errors are mean error estimates, calculated for each typical atmospheric condition (northern/southern, polar/midlatitude/tropics, winter/spring/summer/autumn, day/night). In cases of multiplicative error components, these are adjusted to the actual profile. In total, ..."

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3. Is there any information about the consistency between the different data products, esp. FR vs. RR products? For example, Figs. 4, 5 and 8 show averages over the complete MIPAS time series, so this should include both FR and RR data. A possible systematic offset between FR and RR could have some impact on the results. From the error budget described in section 4.1 it seems that there are differences.

A consistency check - which we unfortunately missed to perform up to now - shows that at 10 km the CH4 (N2O) VMRs of the FR data set are 10-15% (7%) lower than those of the RR data set for latitudes higher than $\pm 50^{\circ}$. This is due to stronger oscillations of the FR-profiles in this altitude region. On the other hand, the tropical CH4 VMRs at 10 km are slightly higher for the FR data set than for the RR data set. At 15 km the tropical CH4 (N2O) VMRs of the FR data set are about 5% (2%) lower than those of

the RR data set. At the altitudes above, the agreement between FR and RR data becomes fairly well both for CH4 and N2O.

The inconsistencies between V8 FR- and RR data at 10 and 15 km also show, that the delta validation against V5 data should better be performed separately for the FR- and the RR-period. Therefore we

- 5 suggest to replace Fig.8 by an update containing V8-V5 differences for the FR- and the RR-period and to discuss them separately. Except for the altitudes of 10 km at high latitudes and of 15 km in the tropics, the new discussion of V8-V5 differences for the larger RR data sets will not change much. However, due to the inconsistencies described above, the differences between the FR data sets show some specific characteristics, especially at 10 km altitude at high latitudes and at 15 km in the tropics. Referring to
- 10 those differences between the RR- and FR-differences, we will also add some sentences as given above wrt the V8-V5 offset in Sect. 5.3 and in the conclusions.

4. The Supplement is very extensive (284 pages). I think for the paper all this information is not really required, especially because the references in the paper do not point to specific parts/pages in the Supple-

15 ment. I also expect that data users would prefer to have the relevant part of this information in the data product itself. However, as this is only a Supplement you may decide to keep it. We would prefer to keep it, because, as mentioned in Sect. 4.1, it contains the whole set of mean error estimates for each of the different atmospheric conditions.

20 Specific Comments:

1. p. 4, l. 2–11: As described in this paragraph, the oscillation detector only changes the initial guess between one iteration and the next. Does this imply that there is not enough information in the measurements themselves to remove the oscillations and you just apply an additional constraint on smoothness of the profile? Does this have an effect on e.g. vertical resolution?

There seems to be a misunderstanding: here we do not discuss oscillations of profiles in the altitude domain but oscillations of each single value in the course of the iteration. I.e., we encountered cases where a value jumps back and forth from iteration to iteration without reaching the minimum of the cost function which presumably lies between these extreme values. This has no effect on vertical resolution.

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2. p. 4, l. 13: What is meant with "cloud-threshold of 4.0"? Does this refer to a maximum cloud optical depth?

A cloud-threshold of 4.0 means that a measurement is rejected, if the ratio of the mean radiances in the wavenumber regions 788.2-796.25 cm⁻¹ and 832.3-834.4 cm⁻¹ is below 4. With increasing cloud

5 contamination this ratio decreases further to values close to 1 (Spang et al., 2004). We will change the passage

"Like for previous CH4- and N2O-retrievals, a more restrictive cloud filter than for temperature and tangent altitude retrieval (cloud-threshold of 4.0 using analysis windows around 792 and 833 cm⁻¹) was used for the V8 data version by additionally discarding spectra with a mean spectral radiance contrast of

10 less than 1.8 between the analysis windows 1246.3–1249.1 cm⁻¹ and 1232.3–1234.4 cm⁻¹." into

"Like for previous CH4- and N2O- retrievals, a more restrictive cloud filter than for temperature and tangent altitude retrieval was used for the V8 data version. As for the temperature and tangent altitude retrieval by Kiefer et al. (2021) a minimum spectral radiance ratio of 4 between analysis windows around

15 792 and 833 cm⁻¹ was required to accept spectra as cloud-free. In addition to this, for the CH4 and N2O retrieval spectra were discarded when the ratio of the mean spectral radiances between the analysis windows 1246.3–1249.1 cm⁻¹ and 1232.3–1234.4 cm⁻¹ was less than 1.8."

3. p. 5, l. 30–31: Please explain what is meant with "instrumental characteristic". Do you refer here to e.g.
altitude dependent spatial stray light which is not corrected?

As shown by Kleinert et al. (2018) there is a systematic positive offset in MIPAS spectra, which in channel A is about 2.5 nW cm⁻² sr⁻¹ cm at high altitudes and increases to 8 nW cm⁻² sr⁻¹ cm at 33 km. Kleinert et al. (2018) attribute this increase to straylight from Earth or clouds. This will become clearer with the changes outlined in the answers to reviewer #2.

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4. p. 14, section 4.3: Please specify which information is used to determine the horizontal averaging kernels. Are these derived solely from the measurements or are e.g. model data used?

The horizontal averaging kernels are calculated from 2D Jacobians provided by the radiative transfer forward model KOPRA, operated in a 2D mode for the specified atmosphere. From these 2D Jacobians the 2D averaging kernels are calculated as proposed by von Clarmann (2009a, their Eq. 5). The way to

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calculate these 2D averaging kernels thus follows exactly the rationale behind the well-established profile averaging kernels, except that 2D Jacobians are used.

We will change the passage

"Since in limb sounding the line of sight extends over long horizontal distances in the atmosphere, hori-

5 zontal averaging kernels are an issue. Here we rely on the concept by von Clarmann et al. (2009a). Some more information on the technical application of this concept can be found in Kiefer et al. (2022)." into

"Since in limb sounding the line of sight extends over long horizontal distances in the atmosphere, horizontal averaging kernels are an issue. The horizontal averaging kernels are calculated from 2D

10 Jacobians provided by the radiative transfer forward model KOPRA, operated in a 2D mode for the specified atmosphere. From these 2D Jacobians the 2D averaging kernels are calculated as proposed by von Clarmann (2009a, their Eq. 5). The way to calculate these 2D averaging kernels thus follows exactly the rationale behind the well-established profile averaging kernels, except that 2D Jacobians are used. Some more information on the technical application of this concept used here can be found in Kiefer et al. (2022)."

15 al. (2022)."

5. p. 17/18, section 5.3: Maybe the delta validation results should be related to the results from the error analysis. Are the observed changes within the expected systematic uncertainties of the products? According to the delta validation there is a high bias of tropospheric and lower stratospheric CH4 and N2O

- of up to 0.3 ppmv and of up to 45 ppbv, respectively, in total (taking the bias of the V5 data into account). We think that rather this total bias should be compared to the error analysis. The high bias in CH4 is well within the spectroscopic uncertainties given in HITRAN2016. The high bias in N2O is within, or of the order of, the spectroscopic N2O uncertainty. We will add one or two sentences addressing this point at the end of Sect. 5.3 and specify the wording in the conclusions on P19, L14f, respectively.
- 6. p. 19, l. 16: "We suspect that this bias might be due to the spectroscopic data used, which suffers from large uncertainties." Why does an uncertainty in the spectroscopic data result in altitude dependent biases? Is this e.g. related to dependencies on pressure or temperature? Please explain.

We did not express ourselves clearly here. We suspect that the high bias generally found in V8 and V5 MIPAS CH4 and N2O data to a considerable degree is due to the spectroscopic data. As noted by reviewer

30 #1, the altitude dependent differences in the delta validation indeed should have additional reasons. To

take this into account, we will include the changes and additions outlined in our answers to reviewer #1.

Technical Corrections:

1. p. 4, Table 1: Please provide in the caption some information on what is meant with the column "Re-

5 trieval log/lin". This is explained later in the text, but at this stage it is not clear if log/lin refers to the altitude axis or the retrieved mixing ratios.

We will add the sentence "log/lin indicates retrieval of log(VMR) or of VMR."

2. p. 4, l. 14: Please define "mean spectral radiance contrast". Is this the ratio between the mean radianceof the two windows?

Yes, it is the ratio between the mean radiance of the two windows. We will change "with a mean spectral radiance contrast of less than 1.8 between the analysis windows ... " into "with a ratio of less than 1.8 between the mean spectral radiance of the analysis windows ...

15 3. p. 5, l. 12–13: "In the V8 retrievals, for the first time horizontal mixing ratio gradients of both target gases were additionally retrieved along with the other unknowns." I suggest to add a cross reference to section 3.2 here.

Ok, we will add "(cf. Sect. 3.2)" here.

20 4. p. 11, Table 3: Please define "target-ESD".

We will change the sentence

"The minimum and maximum ingoing noise errors given for temperature, tangent altitude and vmr are calculated from the target-ESD of the respective retrieval. into

25 The minimum and maximum ingoing noise errors for temperature, tangent altitude and vmr of interfering species were available from the preceding retrievals of these quantities.

5. p. 16, l. 12: "the 1.6 ppmv contourline" ? "the CH4 1.6 ppmv contourline" Ok, we will change the phrase accordingly.