

Reviewer comments are in black and our replies are in blue.

Reviewer #1:

5 General comments

Well written and well organized.

We thank Chris Boone for this positive assessment.

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Just a few relatively minor comments, the most significant being the question of why CF₄ is not included in the analysis. Are the molecules indicated in Tables 3 and 4 all the interferers considered in the analysis? On page 13, line 8, reference is made to H₂O, O₃, and 14 other species in relation to interferers, but only 14 species

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appear in the tables (H₂O, O₃, plus 12 other interferers). Conspicuously missing is CF₄. The microwindow 1282.5-1283.55 is presumably included because it contains the strong Q-branch of the molecule, and the CF₄ signal should extend quite high in altitude (i.e., above the 30 km lower altitude limit of the microwindow). Excluding it from the analysis would be problematic.

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Actually CF₄ is included as interfering gas in the analysis. We forgot to include its error contribution in Tables 3 and 4. This information has been added in the updated manuscript. The second interferer missing in Tables 3 and 4, which however also is included in the retrievals, is CO₂. Its error contribution has also been added.

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Also absent is HDO. I gather differences in isotopologues are generally ignored (both N₂O and CH₄ have lines from subsidiary isotopologues in the given wavenumber region, which will have slightly different VMR profiles than the main isotopologues), but there are HDO lines in some of the low altitude microwindows (i.e., microwindows that extend down to 6 km), and atmospheric fractionation for

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HDO relative to the main isotopologue is around a factor of 2 different from the H/D reference factor assumed when scaling HITRAN intensities to isotopic abundance. If using the same VMR profile as main isotopologue H₂O, the HDO signal will be significantly overestimated. Can that really be ignored in the analysis?

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As the reviewer notes correctly, retrieval of isotopologues was not performed in MIPAS CH₄/N₂O analysis. Due to his concerns we checked the influence of a joint-fit of HDO on the retrieved CH₄- and N₂O-profiles. The outcome is a clear reduction of the RMS in spectral fits for the altitude region 9–15 km, but only a small increase of the CH₄-VMRs by up to 6 ppbv (0.35%) and of the N₂O-VMRs of up to 2.5 ppbv (0.8%) for profiles averaged over one orbit. For individual scans

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the differences mostly range from -10 to +20 ppbv for CH₄ and from 1 to 5 ppbv for N₂O. Thus, fortunately, the error caused by neglect of joint-fitting of HDO is rather small. The reason is that there are quite a lot of other microwindows in this altitude region, which do not contain prominent HDO lines. We included the sentences

45 “Although some of the microwindows used for CH₄ and N₂O retrieval (cf. Sect. 3.4) contain prominent lines of the water vapour isotopologue HDO, it is not jointly retrieved. Test calculations showed that a joint-fit of HDO mostly led to changes of the order of 1% or less in the lower parts of the CH₄ and N₂O profiles only.”

50 at the end of Sect. 3.1 of the revised version.

There are some other molecules that could contribute weakly to the signal in this region (e.g., C₂F₆), but perhaps those contributions are adequately accounted for by the “continuum” fitting parameters.

55 Additional modelling of C₂F₆ radiances leads to about 3 ppbv lower orbit-averaged CH₄-VMRs and up to 1 ppbv lower orbit-averaged N₂O-VMRs in the altitude region 9-14 km. This effect is even smaller than the error caused by neglect of a joint-fit of HDO and compensates for the latter deficit to a considerable degree. Deviating from our announcement, we did not include a comment on C₂F₆ in the
60 revised version because of its minor influence.

In this wavenumber region, the spectroscopy for HNO₃ has been improved in recent years, but there remain missing hot bands in the HITRAN database, which could impact the analysis results.

65 We use the HNO₃ line data of the hitran_mipas_pf_v4.45 database (Flaud et al., 2015), which in the spectral region around 7.6 μm has been improved for missing bands by A. Perrin. Flaud et al. (2015) have shown a very good consistency between HNO₃ retrievals around 11 μm (MIPAS standard retrievals) and 7.6 μm, when this improved HNO₃ spectroscopy is used. Thus we do not suspect large problems with
70 modelling of HNO₃ in our CH₄- and N₂O-retrievals. With regard to this topic we added the sentences

“For O₃ and HNO₃, versions pf3.32 and pf4.45 of the MIPAS spectroscopy were used, respectively. In the latter database, the HNO₃ band in the 7.6 μm region is
75 thoroughly adjusted to the band in the 11 μm region, which is essential in order to use the HNO₃ prefit.”

in Sect. 3.5.

80 Page 19: line 16: “We suspect that this bias might be due to the spectroscopic data used.”

Likely a major (maybe the biggest) factor, but not necessarily the only one. It is perhaps worth noting strong correlations between the CH₄ and N₂O differences in Figure 8. Between latitudes 40 S to 40 N and altitudes 10 to 30 km, there is a
85 common pattern: an increase relative to V5 between 10 and 15-20 km, a decrease relative to V5 near 20 km, and an increase relative to V5 between 25 and 30 km. I gather the two increases worsen the level of agreement with correlative data. The fact that N₂O and CH₄ share this pattern suggests a common source for the differ-

ences, something mechanical in the retrieval (changes in tangent heights, changes
90 in pressure and temperature profiles, etc.). This in turn suggests there could be con-
tributions to the biases other than just spectroscopy.

Maybe we did not express clearly what we intended to say. As stated in Section
5.3, there has already been a high bias compared to measurements near 3000 cm^{-1}
in the V5 versions of CH_4 - and N_2O -retrievals. This bias generally has further
95 increased below 25 km for V8 CH_4 and N_2O . We changed the sentence on P19,
L16 into

“We suspect that the high bias of both gases to a large part might be due to the
spectroscopic data used, which suffer from large uncertainties. The high bias of
100 N_2O presumably has additional yet unidentified causes, since it clearly exceeds the
reported spectroscopic uncertainties.”,

and added the sentence

105 “Further, we attribute the observed oscillations in the V8-V5 differences, which
are partly correlated between CH_4 and N_2O , to the better vertical resolution in V8
profiles.”,

110 which is different from our announcements, because we came to the conclusion
that the major reason of the oscillations is the better vertical resolution.

Further, the whole paragraph beginning on P19, L10 as well as Sect. 5.3 have been
completely modified with respect to the change (discussion of V8-V5 differences
separately for FR and RR data) announced in the reply to reviewer #3.

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Minor issues:

Page 2, line 32: a formatting issue (truncated sentence that is continued on the next
page).

This formatting issue has been corrected.

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Page 16, line 21: ”even some more” even more

Has been changed accordingly.

Reviewer #2:

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The paper is well written and well suited for AMT. I’d suggest publication after a
few minor corrections, listed below.

We thank the reviewer for this positive evaluation.

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Page 1, lines 4-5: "... with respect to the atmospheric state variables jointly retrieved with the target gases CH₄ and N₂O ..." is awkwardly phrased. Does this mean that CH₄ and N₂O are now retrieved simultaneously and weren't previously?

No, MIPAS CH₄ and N₂O have always been retrieved simultaneously at IMK/IAA.

135 For better understanding the passage

"... with respect to the atmospheric state variables jointly retrieved with the target gases CH₄ and N₂O ..."

140 has been changed into

"... with respect to the atmospheric state variables that are jointly retrieved along with the target gases CH₄ and N₂O ..."

145 Page 2, line 18: "Submillimeter Wave Radiometer" should be "Sub-Millimetre Radiometer" (note the spelling of sub-millimetre)

The phrase has been changed accordingly.

Page 3, line 30: Please specify what the high bias is with respect to

150 The passage

"Both MIPAS IMK/IAA CH₄ and N₂O profiles always had a high bias in the troposphere and the lower stratosphere (Laeng et al., 2015; Plieninger et al., 2016)."

155 has been replaced by

"Both MIPAS IMK/IAA CH₄ and N₂O profiles always had a high bias in the troposphere and the lower stratosphere with respect to ground-based in-situ measurements and remote measurements in other spectral regions (Laeng et al., 2015; Plieninger et al., 2016). For MIPAS V5 data this bias was of the order 0.1–0.2 ppmv for CH₄ and of ~20 ppbv for N₂O."

Page 5, lines 14-16: I assume you mean H₂O and HNO₃ are interfering species that are no longer joint-fitted. If so, please clarify in the text. Also, please explain what is meant by "hardly any influence"

165 The sentence

"Contrary to the previous data version, H₂O and HNO₃ were no longer joint-fitted, but their previously retrieved V8 profiles were used instead."

170 has been replaced by

"Contrary to the previous data version, the interfering species H₂O and HNO₃ were no longer fitted jointly with the target species, but their previously retrieved V8

175 profiles were used instead.”.

Section 3.1.2: Please briefly explain the physical justification for how the radiance offset could be altitude-dependent?

180 As outlined in Kleinert et al. (2018) there is an offset variation with altitude, which “is related to straylight rather than an instrumental offset.” The passage

185 “While the radiance offset correction fitted in previous data versions was allowed to be microwindow dependent, but forced to be altitude-constant, it is now allowed to be altitude-dependent as well. By this means the instrumental characteristic can be reproduced better.”

has been changed into

190 “The radiance offset correction fitted in previous data versions was allowed to be microwindow dependent, but forced to be altitude-constant. Now it is allowed to be altitude-dependent as well, because Kleinert et al. (2018) have shown that the offset increases with decreasing altitude, which they attribute to straylight from Earth or clouds.”

195 Page 8, line 11: If CO₂ is an interfering species, why isn't its VMR listed as an uncertainty in Table 3?

CO₂ has been forgotten in Tables 3 and 4 and has been added.

Page 17, line 4: for consistency, “methane” should probably be “CH₄”

200 Has been changed.

Page 17, line 8: it seems a bit odd to be using vague phrases like “had been as high as” and “could be reduced to” and then give very specific values. Why not just give the rates for both V5 and V8 (as done in the next sentence), or say the version that had a non-convergence rate of 8.4%?

205 The sentence has been changed into “For FR measurements, the portion of non-converged retrievals were reduced from 8.4% to 0.3%.”

210 Figure 6: In my opinion, there's more data here than necessary, which makes it difficult to see the region of interest. Maybe consider only showing the northern latitudes to highlight the oscillations better.

To highlight the oscillations better we restricted the lower row of Fig. 6 to the latitudes 75N–90N–45N and to the altitudes 5-50 km.

215 **Reviewer #3:**

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. I have the following comments.

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Thank you for this favourable judgement.

General Comments:

1. The description of horizontal gradients in Sect. 3.2 should be clarified. Where
225 does the information 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
230 3D a priori temperature fields is described in detail in Kiefer et al. (2021). The
information on horizontal temperature variability 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
235 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 replaced the sentence on P6, L8ff

240 “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).”

245 by

“Thus, in the V8 retrieval of temperature and line-of-sight a 3D temperature a
priori field was scaled to the retrieved profile values, and in addition a horizontal
temperature gradient was determined for the upper range of the retrieval altitudes
250 (see Kiefer et al. 2021). This information is used in the CH₄ and N₂O retrievals.”,

which is a bit different from our announcement, but makes the topic clearer. Further
we added the sentence

255 “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 ± 400 km around the tangent points. No additional
assumptions are made.”

260 after the sentence (P6, L14ff)

“Instead, the horizontal gradients ...”

265 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?

The sentence on P10, L21ff

270 “For each single CH₄ and N₂O profile, individual noise estimates are supplied. For the other error sources, mean error estimates are provided for each typical atmospheric condition (northern/southern, polar/midlatitude/tropics, winter/spring/summer/autumn, day/night), which can be assigned to the profile under assessment.”

275 has been changed into

280 “Error profiles representing the effect of measurement noise, provided by the retrievals, are reported separately for each single CH₄ and N₂O profile in the data base. In addition, the data base includes estimates of the total random, total systematic, and total error. These are evaluated for typical atmospheric conditions (northern/southern, polar/midlatitude/tropics, winter/spring/summer/autumn, day/night), and to each CH₄ and N₂O profile the representative error estimates are assigned. In cases of multiplicative error components, the representative error estimates are adjusted to the actual profile.”

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which, again, is a bit different from our announcement, but makes things even clearer.

290 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.

295 A consistency check 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 ±50°. 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
300 above, the agreement between FR and RR data becomes fairly well both for CH₄ and N₂O.

305 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 replaced Fig. 8 by an update containing

V8-V5 differences for the FR- and the RR-period and discussed 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 did not change much for the larger RR data sets. 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 those differences between the RR- and FR-differences, we also added the following sentences wrt the V8-V5 offset at the end of Sect. 5.3.:

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“Since there is a generally good consistency between V5 data of the FR and RR period both for CH₄ and N₂O, the deviations between the left-hand and the right-hand plots in Figure 8, especially in the lower atmosphere, indicate an offset between the V8 data of the two periods. The deviations are largest at high latitudes around 10 km and in the tropics around 15 km, with CH₄ differences between RR and FR data of 10-15% and of 5%, respectively. For N₂O, the respective RR-FR differences are 7% and 2%. However, at higher altitudes the consistency between V8 RR and FR data is good.”

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However, deviating from our announcement, we did not repeat this statement in the conclusions.

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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 Supplement. 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.

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We preferred 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.

335 **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?

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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. This means, 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
350 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 $788.2\text{--}796.25\text{ cm}^{-1}$ and $832.3\text{--}834.4\text{ cm}^{-1}$ is below 4. With increasing cloud contamination this ratio decreases further to values close to 1 (Spang et al., 2004). We changed the passage

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“Like for previous CH_4 - and N_2O -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 less than 1.8
360 between the analysis windows $1246.3\text{--}1249.1\text{ cm}^{-1}$ and $1232.3\text{--}1234.4\text{ cm}^{-1}$.”

into

365 “Like for previous CH_4 - and N_2O - 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 792 and 833 cm^{-1} was required to accept spectra as cloud-free. In addition to this, for the CH_4 and N_2O retrieval spectra were discarded when the ratio of the mean spectral radiances
370 between the analysis windows $1246.3\text{--}1249.1\text{ cm}^{-1}$ and $1232.3\text{--}1234.4\text{ cm}^{-1}$ 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?

375 As shown by Kleinert et al. (2018) there is a systematic positive offset in MIPAS spectra, which in channel A is about $2.5\text{ nW cm}^{-2}\text{ sr}^{-1}\text{ cm}$ at high altitudes and increases to $8\text{ nW cm}^{-2}\text{ sr}^{-1}\text{ cm}$ at 33 km . Kleinert et al. (2018) attribute this increase to straylight from Earth or clouds. This becomes 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?

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

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We changed the passage

395 ”Since in limb sounding the line of sight extends over long horizontal distances
in the atmosphere, horizontal 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

400 ”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 Jacobians provided by the radiative transfer forward
model KOPRA, operated in a 2D mode for the specified atmosphere. From these
405 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).”

410 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 total high bias (taking the bias of the
V5 data into account) of tropospheric and lower stratospheric CH₄ and N₂O of up
415 to 0.3 ppmv and of up to 45 ppbv, respectively. 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 larger
than the spectroscopic uncertainty in N₂O. Addressing this point we added the
sentence

420 “In summary the CH₄ bias at all altitudes is well below the total error presented in
Sect. 4.1.”

at the end of the third paragraph in Sect. 5.3 and the sentences

425 “At 10 km in the tropics this apparently might cause an increase of the bias against
ACE-FTS from 30 to 45 ppbv. This bias is larger than the total N₂O error presented
in Sect. 4.1 for the lowermost altitudes.”

430 at the end of the fifth paragraph in Sect. 5.3. However, we abstained from repeating
these statements in the conclusions.

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 spec-
435 troscopic 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 #1, the altitude dependent differences in the delta validation indeed should have additional reasons. To take this into account, we included the changes and additions outlined in our answers to reviewer #1.

Technical Corrections:

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1. p. 4, Table 1: Please provide in the caption some information on what is meant with the column "Retrieval 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 exchanged log/lin into lin/log and add the sentence "lin/log indicates retrieval of VMR or log(VMR)."

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

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Yes, it is the ratio between the mean radiance of the two windows. We changed "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 ..."

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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.

We added "(cf. Sect. 3.2)" here.

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

We changed 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."

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into

"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."

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5. p. 16, l. 12: "the 1.6 ppmv contourline" ? "the CH₄ 1.6 ppmv contourline"

We changed the phrase accordingly.