

Reply to the comments of reviewer #1

Norbert Glatthor et al.

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

General comments

Well written and well organized.

5 We thank Chris Boone for this positive assessment.

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
10 14 species 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.

Actually CF₄ is included as interfering gas in the analysis. We forgot to include its error contribution in
15 Tables 3 and 4. This information will be 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 will also be added.

Also absent is HDO. I gather differences in isotopologues are generally ignored (both N₂O and CH₄ have
20 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 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,
25 the HDO signal will be significantly overestimated. Can that really be ignored in the analysis?

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 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 plan to include a comment on neglect of HDO retrieval in the revised version.

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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.

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 lower than the error caused by neglect of a joint-fit of HDO and luckily compensates for the latter deficit to a considerable degree. Here we also plan to include a comment in the revised version.

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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.

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 modelling of HNO₃ in our CH₄- and N₂O-retrievals.

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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 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

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

5 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 CH₄- and N₂O-retrievals. This bias generally has further increased below 25 km for V8 CH₄ and N₂O. We will change the sentence on P19, L16 into

“We suspect that this bias to a large part might be due to the spectroscopic data used, which suffers from
10 large uncertainties.”

and add the sentences

“However, the oscillations in the V8-V5 differences presented in the delta validation in Section 5.3, which are partly correlated between CH₄ and N₂O, point to additional problems in the retrieval setup.
15 These may result from the V8 pressure and temperature profiles used in the retrievals as well as from the higher vertical resolution of the V8 CH₄ and N₂O profiles.”

Further, the whole paragraph beginning on P19, L10 as well as Sect. 5.3 will be slightly modified wrt the change (discussion of V8-V5 differences separately for FR and RR data) announced in the reply to
20 reviewer #3.

Minor issues:

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

This formatting issue will be corrected.

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

Will be changed accordingly.