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Differences in ozone retrieval in MIPAS channels A and AB: a spectroscopic issue

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Response to reviewer 1:

We thank reviewer 1 for her/his helpful comments. Please find below our responses describing how the manuscript has been modified with respect to the comments. Blue passages denote the changes or updates in the revised manuscript.

Specific Comments

Comment: *"Pag.4, Section 3. The description of the errors in MIPAS spectroscopic databases should be moved before Sect.7.1, where differences between several spectroscopic databases are quantified, and hence the estimation of the errors on line intensities and line width can be more useful."*

Reply: Similar to the description of the MIPAS experiment and of the retrieval setup in Section 2, the error estimates for ozone lines in the MIPAS and HITRAN spectroscopic databases are a prerequisite for our investigations. Therefore we find it more appropriate to leave the description of these errors in Section 3 prior to the retrieval section. To address the reviewer's point, we will compare the spectroscopic errors with the VMR differences in Sections 4 and 7.1 by adding the sentences "This difference is larger than the relative error in line intensity given in Eqs. 1 and 2 for the strongest and medium scale ozone lines (at least for transitions with low to medium-sized rotational quanta JU and KU)." at page 4, line 26, and "Consequently, these differences are also larger than the relative errors in line intensity given in Eqs. 1 and 2." at page 7, line 15.

Comment: *"Pag. 5, line 17: what 'completely different forward models' means ?"*

Reply: This phrase is maybe a bit incomprehensible, because the forward model KOPRA we use at IMK/IAA has not been introduced before. For this reason we will replace the first sentence of the second paragraph of Section 2 by "To reinvestigate the channel AB-A bias in retrieved ozone, retrievals using the the processor of the Institut für Meteorologie und Klimaforschung and the Instituto de Astrofísica de Andalucía (IMK/IAA) were performed for 59 MIPAS orbits from January 3, April 1, July 2 and October 2-3, 2009. This processor uses the Karlsruhe Optimized and Precise Radiative Algorithm (KOPRA) (Stiller, 2000) for radiative transfer calculations and the Retrieval Control Program (RCP) of IMK/IAA for inverse modelling of spectra. ". Further we will change the critisised phrase into "a different radiative transfer model". Because by these modifications the acronym KOPRA is already explained in Section 2, we will change the subsequent sentence (page 5, lines 19/20) into "This agreement widely excludes the hypothesis that the bias is caused by deficiencies in the KOPRA forward model used at IMK."

Comment: *"Pag. 5, lines 9-14: I think that these sentences may be misleading in the paper. Indeed, Laeng et al., 2014 shows that from the comparison between MIPAS Ozone with ACE-FTS and MLS, MIPAS is larger than both of them. Since O3 retrieved from channel AB is larger than O3 retrieved from channel A, we can deduce that the use of only spectral intervals in band A may reduce the differences with respect to ACE-FTS and MLS. However, we have to consider that positive differences between MIPAS and ACE-FTS are probably not due, or at least not only due, to spectroscopic issues, since ACE-FTS performes measurements in the same spectral regions as MIPAS and for the O3 retrieval mainly spectral points in the region of MIPAS band AB are used (see http://www.ace.uwaterloo.ca/misc/ACE-SOC-0027-ACE- FTS_Spectroscopy-version_3.5_Jan222016_Rev1A.pdf). Furthermore, the* *tests reported in this paper do not indicate which of the two bands A and AB has smallest spectroscopic errors, but only that there are inconsistencies between the two bands. Finally, the change of used spectral intervals in order to reduce the bias with other correlative measurements, that do not represent the true, may not always be correct."*

Reply: We do not quite understand the referee's arguments in this comment. First of all, Laeng et al. (2014, Fig. 5) indeed show that the MIPAS ozone VMRs are larger than those of MLS at nearly all altitudes, but there is no general positive bias with respect to ACE-FTS. MIPAS ozone VMRs are up to 3% larger than those of ACE-FTS below 30 km, but up to 2% lower between 30 and 45 km. Between 45 and 55 km MIPAS ozone is even more than 10% lower than ACE-FTS ozone. Secondly, ACE-FTS does not perform measurements (ozone retrievals) in the same spectral region as MIPAS. ACE-FTS uses the spectral region 1027–1059 cm^{-1} (see document cited above), but MIPAS (data version O3_V5R_224) the region 687–791 cm^{-1}. Only above 50 km two channel AB microwindows at 1029–1031 and 1038–1039 cm^{-1} are added. Thus, differences between ACE-FTS and MIPAS can well have spectroscopic causes. We agree with the referee's statement that "the tests reported in this paper do not indicate which of the two bands A and AB has smallest spectroscopic errors". Just as he/she concludes, we only want to show "that there are inconsistencies between the two bands." Further, the referee might be right by stating that "the change of used spectral intervals ... may not always be correct". But there is justification for such a change, if a similar bias to several correlative instruments can be reduced in doing so. With this we can at least provide an explanation of the discrepancies encountered.

Comment: *"MIPAS spectroscopic database pf 3.2 sometime is mentioned in the paper (e.g. Pag.9, line 4) as MIPAS spectroscopy, other times (e.g. Caption of Fig.5) as Mipas pf 3.0. Please use consistent terminology. "*

Reply: We agree and will speak of MIPAS pf3.2 throughout the updated manuscript. We were a bit unprecise, because the ozone spectroscopy in MIPAS pf3.2 is the same as in MIPAS pf3.0.

Comment: *"Last sentence of the paper: 'as far as ozone is concerned we recommend to use version pf3.2 of the MIPAS spectroscopy and not the latest update pf4.45, because the ozone data set in this compilation is identical with HITRAN-2008.' A reference to the spectroscopic database pf4.45 should be added. The presence of 'inappropriate halfwidths' in HITRAN 2008 and following versions seems to involve only the 790- 850 cm-1 spectral region."*

Reply: We will add the reference "Flaud, J.-M., Perrin, A., and Ridolfi, M.: New release of the MIPAS spectroscopic database: hitran_mipas_pf_v4.45, Presentation at MIPAS QWG 38, ESA-ESRIN, 18-19 February 2015." for the spectroscopic database pf4.45. Concerning inappropriate halfwidths in HITRAN-2008: We showed one example of an obviously unphysical step in halfwidths at 797.05 cm^{-1} in HITRAN-2008 and subsequent editions. However, we can not draw general conclusions about the spectral ranges of inappropriate halfwidths in the HITRAN data bases. This issue has to be left to spectroscopists.

Technical Corrections

The requested technical corrections will be performed. Fig. 4 will be interchanged with Fig. 3 to obtain a consecutive discussion of the figures. In figures with several plots each plot will be identified with a letter. Finally, "diff / ppmv" will be replaced by "diff / km" in Fig. 1.

References

Laeng, A., et al.: Validation of MIPAS IMK/IAA V5R_O3_224 ozone profiles, Atmos. Meas. Tech., 7, 3971–3987, www.atmos-meas-tech.net/7/3971/2014/, 2014.

Response to reviewer 2:

Many thanks for reading our manuscript and your helpful comments. Please find below our responses describing how the manuscript has been modified with respect to your annotations. Blue passages denote changes or updates in the revised manuscript.

General Comments

For clarification: We do not use lines of the fundamental ν_3 band for channel A retrievals, but lines of the ν_2 band and higher transitions in the spectral range 687–791 cm^{-1} only.

Comment: *"The manuscript could possibly gain more widespread interest by i) including a comparison of pressure broadening parameters to including the MIPAS data base and ii) by quantifying the impact on total ozone columns. This would allow to better estimate the impact of this particular parameter on the existing bias between UV and IR comparison measurements (see Orphal et al., 2016, and references therein)."*

Reply: (i) We think that inclusion of the MIPAS pf3.2 database in the comparison of pressure broadening parameters in the main manuscript is not very conductive, because we do not want to find the reasons for the relatively small differences between ozone retrievals using the MIPAS pf3.2 and the HITRAN-2008 spectroscopy. To address the referee's suggestion, we will add the following paragraph at the end of Section 7.1: "The rather good agreement between the channel A as well as the channel AB retrievals using the MIPAS spectroscopy or the HITRAN 2004 edition and later ones indicates largely consistent spectroscopic parameters of identical ozone lines in these databases for the spectral range of the channel A and AB microwindows. Therefore a comparison between the line parameters of the MIPAS and HITRAN databases is not presented here, but as supplemental material only." and show the requested comparison as supplemental material. To emphasize the good intra-band agreement between most of the channel A and AB profiles (MIPAS vs HITRAN) we will add two graphs showing the absolute channel A and AB profiles to Figure 5. Further, we will slightly change the first paragraph of Section 7.2.2 into "The retrieval results also indicate mostly consistent spectral parameters in HITRAN-2008 and GEISA-2015 for the ozone lines used in MIPAS channel AB, but considerable spectroscopic differences in the region of the channel A microwindows. In the following, we will compare the HITRAN-2008 and GEISA-2015 ozone lines applied in channel A as well as in channel AB to identify the parameters responsible for these differences.

(ii) To consider the referee's second point, we will add a fourth graph to Figure 11 showing the relative differences between channel AB retrievals using the unchanged pressure broadening coefficients and the two sets of modified coefficients. Since these differences are around -4% and -5% over a large altitude range, they are a good estimate for the respective changes in ozone columns. We will shortly discuss this finding in Section 7.3 in comparison with the results of Schneider et al. (2008), who found a systematic difference of 4–5 % between IR measurements in the spectral range 991–1007 cm^{-1} and UV observations. Our results show that, beside the re-scaling of line strengths of the ν_3 and ν_1 lines by 4% as discussed in Smith et al. (2012), a change of air broadening coefficients can lead to a similar adjustment to the UV measurements.

Specific Comments

Comment: *"There are two possibly important omissions in the paper. As already pointed out above, the MIPAS database/spectroscopy deserves a short presentation so that similarities and differences with respect to the other data bases become clear. It would also be helpful to see a detailed comparison of line-broadening parameters between MIPAS and HITRAN and MIPAS and GEISA (such as in Figs. 8 and 9 for HITRAN and GEISA) to better understand differences in the data bases. This also because MIPAS is finally recommended to be preferred over the other data sets. The other issue is that line intensities (see line strengths of 2 = 1 0, (J + 1, J + 1, 1) (J, J, 0) transitions in Fig. 13, for example) are compared using reference* *temperatures at room, but at stratospheric temperatures the lower state energies (and to a lesser extent partition sums) also contribute. The quoted line strength uncertainty might thus be too optimistic. While partition sums cannot lead to an inter-band bias, lower state energies can. For the sake of completeness a discussion of the impact of possible differences in lower state energies or a comparison of low temperature intensities would be required. "*

Reply: A short presentation of the MIPAS spectroscopy is already given in the Introduction on page 2, lines 2–4. We will add some more information on the MIPAS spectroscopy here. As mentioned above, we will present and discuss a comparison between the spectral parameters in MIPAS pf3.2 and in HITRAN-2008 such as in Figs. 8 and 9 as supplemental material. As requested by the referee, we also performed a comparison of the lower state energies of the corresponding lines in MIPAS pf3.2, HITRAN-2008 and GEISA-2015 for the spectral region of our microwindows. Except of some very weak lines there is perfect agreement between the lower state energies. We will discuss the potential bias due to inconsistent lower state energies and the result of our inspection at the end of the first paragraph on page 9.

Comment: " *The manuscript preparation guidelines request that "works cited in a manuscript should be accepted for publication or published already" and the authors should therefore avoid utilizing personal communications. The communications used are not really required and seem to be problematic. For example, in Section 3 (Error estimates of ozone lines and band intensities), a pers. communication (J.M. Flaud) is given to motivate relative errors of the three fundamentals. Eq. (1) indicates that the relative error is the same for the 1, 2 and 3 bands. However, the comparison of experimental data with intensity calculations from the same author shows that the agreement in the 2 cold band is usually worse than in the other two fundamental bands (See section 5.2.2. of Wagner et al., 2002). This information therefore seems to be conflicting. Later it is stated that "These inappropriate halfwidths (M. Birk, pers. comm.) are the reason for the stronger ozone lines in the model spectrum using HITRAN-2008 data in Figure 12. This* *deficiency is still present in later versions up to HITRAN-2016." A priori, it is not clear which set of half widths should be correct and which not and why these half widths cause problems. Non-continuous behaviour is visible in both data sets (see Fig. 13 right). Wouldn't it be more informative and decisive to show the direct comparison between modelled and experimental spectra? "*

Reply: To address the referee's objection against personal communications we will change the phrase on page 2, lines 26-28 into "Since the uncertainties in line intensity of many lines of the ν_2 and ν_1/ν_3 fundamentals observed in channels A and AB, respectively, have been determined to be less than 2% (Wagner et al., 2002) ...". Further we will remove the phrase "(M. Birk, pers. comm.)" on page 10, line 20. However, the error estimates given in Section 3 are from an internal technical note by J.-M. Flaud and C. Piccolo for MIPAS data evaluation only and can not be cited in a more convenient way. The referee critizises that the relative error given in Eq. 1 "is the same for the 1, 2 and 3 bands". But exactly these error estimates were provided by Flaud and Piccolo. Wagner et al. (2002) with Flaud as co-author indeed state a lower accuracy for the ν_2 band at the end of Section 5.2.2, but obviously a weak degradation only ("The results are a little bit worse for the ν_2 band ..."). Coming to the issue of inappropriate halfwidths: As correctly noticed by the referee, non-continuous behaviour is visible in both data sets (Fig. 13, right), but the jump in HITRAN-2008 at 797.05 cm^{-1} is considerably larger than the jump in MIPAS pf3.2 (and HITRAN-2008) at 713 cm^{-1}. As suggested, we will add the results of broadband retrievals in the region 795–825 cm^{-1}, which clearly show that the halfwidths of MIPAS pf3.2 of the respective lines at 797.05, 805.02 and 812.99 cm^{-1} lead to much better agreement with the measurements than those of HITRAN-2008.

Comment: " *The study of Janssen et al. (2016) needs to be mentioned in the paper. It has evident methodological links and has already identified differences in pressure broadening parameters between GEISA (version of 2011) and HITRAN (version of 2012) being the main reason for ozone column retrieval differences in the 3 spectral region at 10 μm. It seems that the surprising effect (section 8:*

Additional observations) of systematic biases in the air broadened half width potentially leading to positive and negative feedbacks depending on the optical thickness of the atmosphere is discussed there as well. "

Reply: After having read the Janssen et al. (2016) paper, we think that its main link to our paper is the discussion in Section 3.2.2 (Sensitivity on pressure broadening coefficient). In this section these authors discuss the results of Table 4 and show the "striking feature" that for lines of the ν_3 band an increase in γ_air similar as an increase in line intensity leads to a negative change in the ozone column. This is consistent to the results in Section 8 of our manuscript. We will cite Janssen et al. (2016) and mention their similar findings after the last sentence of Section 8 (page 10) in our manuscript.

Comment: " *Fig. 6 requires correction. On the one hand some technical information on averaging kernel thresholds and orbit numbers are probably not very informative. On the other hand, the difference plot and the absolute values of the GEISA retrievals are not compatible in the altitude range $<$ 10 km. There is a clear offset (AB - B $>$ 0) between the two bands on the left panel, but the difference plot on the right shows AB = B. "*

Reply: The referee is right. The inconsistency below 10 km occured, because the cloud filter was switched on for calculation of the mean differences, but erroneously not applied for calculation of the mean absolute profiles. This error will be corrected. Further, we will remove technical information on averaging kernels, orbit numbers etc. in Fig. 6 and in similar figures.

Comment: " *Absolute deviations at the per cent level are difficult to perceive on the logarithmic scale. The left plot of Fig. 13 should show the relative deviation between intensities from HIT-08 and PF-3.0. "*

Reply: We rechecked the line intensities plotted in Fig. 13 (left) and realized that they are indeed identical. Therefore we changed the wording from "The line intensities are ... nearly equal ..." into ”The line intensities are ... identical ..." and decided to omit Fig. 13 (left) completely.

Technical corrections

Comment: *“p. 3, l. 27–29 : Phrase is incomplete/wrong”*

Reply: We can not find a clear omission or error in these sentences and ask the referee to give a more specific comment, please.

Comment: *“p. 4, l. 22 : The acronym IAA appears for the first time. Please explain.*

Reply: Since the acronym IAA will already be explained in Section 2 of the updated manuscript (cf. reply to referee 1), it does no longer need to be explained here.

All other technical corrections will be performed as suggested.

References

Schneider, M., Redondas, A., Hase, F., Guirado, C., Blumenstock, T., and Cuevas, E.: Comparison of ground-based Brewer and FTIR total column O_3 monitoring techniques, Atmos. Chem. Phys., 8, www.atmos-chem-phys.net/8/5535/2008/, 5535–5550, 2008.

List of relevant changes

4. Retrievals using the MIPAS spectroscopy:

To account for the request of reviewer 2, this section starts with a new paragraph decribing the MIPAS spectroscopy.

7.1 Comparison of different HITRAN versions:

To consider the suggestions of reviewer 2, this section ends with a new paragraph stating a rather good agreement between corresponding ozone lines in the MIPAS spectroscopy and in recent HITRAN databases (in the spectral range of the applied microwindows). For this reason the comparison of spectroscopic parameters of the ozone lines in MIPAS pf3.2 and in HITRAN-2008 is presented in a Supplemental only.

7.2.2 Comparison of spectral parameters:

As requested by reviewer 2 the lower state energies in MIPAS pf3.2 and HITRAN-2008 of the ozone lines under consideration were compared. This finding is mentioned in Section 7.2.2.

7.3 Channel AB retrievals using modified HITRAN-2008 lines:

To consider the suggestions of reviewer 2, a new paragraph has been added at the end of Section 7.3. Here we state that the bias in ozone column amounts measured in the mid-infrared and UV spectral regions can be reduced by change of the air-broadened halfwidths as well.

8. Additional investigations:

To consider the suggestions of reviewer 2, we added a comparison between simulated and measured spectra. The simulated spectra result from broadband retrievals in the spectral range 795–825 cm^{-1}. The comparison shows that the ozone lines at 797.05, 805.02 and 812.99 cm^{-1} of MIPAS pf3.2 agree much better with the measurements than the ozone lines of HITRAN-2008.

Marked-up manuscript version

Abstract. Discrepancies in ozone retrievals in MIPAS channels A (685–970 cm^{-1}) and AB (1020–1170 cm^{-1}) have been a long-standing problem in MIPAS data analysis, amounting to an inter-channel ~~ozone~~ bias (AB-A) of up to 8% between ozone volume mixing ratios in the altitude range 30–40 km. We discuss various candidate explanations, among them forward model and retrieval algorithm errors, inter-channel calibration inconsistencies, and spectroscopic data inconsistencies. We show that forward modelling errors as well as errors in the retrieval algorithm can be ruled out as an explanation because the bias can be reproduced with an entirely independent retrieval algorithm (GEOFIT) relying on a different forward radiative transfer model. Instrumental and calibration issues can also be refuted as explanation because ozone retrievals based on balloon-borne measurements with a different instrument (MIPAS-B) and an independent level-1 data processing scheme produce a rather similar inter-channel bias. Thus~~spectroscopic inconsistencies~~ , spectroscopic inconsistencies in the MIPAS database used for ozone retrieval are practically the only reason left. To further investigate this issue, we performed retrievals using ~~various~~ additional spectroscopic databases. Various versions of the HITRAN ~~spectroscopic database which, however, generally produce~~ database generally produced rather similar channel AB-A differences. Use of a different database, namely GEISA-2015, ~~leads~~ led to similar results in channel AB, but to even higher ozone ~~amounts~~ volume mixing ratios for channel A retrievals, i.e. to a reversal of the bias. We show that the differences in MIPAS channel A retrievals result from about 13% lower air-broadening coefficients of the strongest lines in the GEISA-2015 database. Since the errors in line intensity of the major lines used in MIPAS channels A and AB are reported to be considerably lower than the observed bias, we posit that a major part of the ~~inter-channel~~ channel AB-A differences can be attributed to inconsistent air-broadening coefficients as well. To corroborate this assumption we show some clearly inconsistent air-broadening coefficients in the HITRAN-2008 ~~data base. The inter-band~~ database. The inter-channel bias in retrieved ozone ~~profiles~~ amounts can be reduced by, e.g., increasing the air-broadening coefficients of the lines in MIPAS ~~band~~ channel AB in the HITRAN-2008 database by 6–8%.

1 Introduction

Ozone is one of the most important trace gases in the atmosphere. Stratospheric ozone to a large extent prevents solar ultraviolet (UV) radiation from reaching the Earth's surface. On the other hand tropospheric ozone is a harmful air pollutant. Therefore knowledge of its atmospheric concentration is of high interest. Remote sensing of ozone is performed in a wide spectral range covering the microwave, infrared and ultraviolet regions. For measurements in the infrared the strong ozone ν_3

band around 10 μm is of particular interest. To obtain accurate atmospheric ozone ~~VMRs~~ volume mixing ratios (VMRs) high quality line parameters are required. For this reason a large number of laboratory measurements has been performed. On the basis of three independent laboratory studies Flaud et al. (2003a) compiled a dedicated line list for evaluation of MIPAS ozone measurements, which ~~was~~ has been included in the HIgh-resolution TRANsmission database version 2004 (HITRAN-2004) (Rothman et al., 2005) and later ones. A review of various laboratory studies performed during the past decades to determine ozone line intensities in the 9–11 μm region has been given by Smith et al. (2012). According to these authors the goal of 1% absolute accuracy in line intensities, as demanded by Flaud and Bacis (1998), is ~~still not~~ not yet attained.

Initiated by several more recent laboratory intercomparisons of ozone absorption coefficients in the mid-infrared and UV spectral regions (Picquet-Varrault et al., 2005; Gratien et al., 2010; Guinet et al., 2010) a debate was reopened on whether the ozone line intensities in the 10 μm region, which ~~have been scaled down since HITRAN-2004~~in the MIPAS spectroscopy and in HITRAN version 2004 and later ones are lower by $\sim$4% than in HITRAN-1996, have to be increased by 3–5% again. This is also supported by an intercomparison of ~~groundbased FTIR and Brewer measurements of total column ozone~~ total ozone columns by Schneider et al. (2008), who found a bias of 4–5% between groundbased FTIR observations at 991–1007 cm^{-1} and Brewer measurements. A review of laboratory and field studies related to this topic was given by Orphal et al. (2016). However, rescaling of the ozone band intensities would only have an effect on the channel AB-A bias observed in MIPAS data, if the ν_2 band used in channel A would not be scaled by the same amount as the bands in the 10 μm region applied in channel AB.

MIPAS measurements are performed in several channels covering the mid-infrared spectral region. Especially suited for MIPAS ozone retrieval are the strong fundamental ~~ν_1, ν_2 and ν_3 bands centered~~ ν_3 band centered at 1042 cm^{-1}, but also the weaker fundamental ν_1 and ν_2 bands at 1103 ~~, 701 and 1042~~ and 701 cm^{-1}~~, respectively~~. While the ν_1 and ν_3 bands are mainly situated in MIPAS channel AB (1020–1170 cm^{-1}), the ν_2 band is ~~covered by~~ located in channel A (685–970 cm^{-1}). The fact that ozone retrievals using MIPAS channel AB (1020–1170 cm^{-1}) microwindows (MWs) are biased high by up to 8% compared to retrievals based on MIPAS channel A (685–970 cm^{-1}) MWs has already been reported by Glatthor et al. (2006) for measurements in the so-called high-resolution mode (2002–2004), who concluded that the major part of the differences results from spectroscopic inconsistencies. ~~Retrievals~~ These retrievals were performed using the ozone line list of the MIPAS database version pf3.2 provided by Flaud et al. (2003b). The bias between MIPAS channel A and AB retrievals is of particular importance, because ozone data retrieved at IMK from a combination of channel A and AB microwindows (versions V5R_O3_220 and V5R_O3_221) have been found to be biased high in the altitude region around 40 km (Laeng et al., 2014). Since the uncertainties in ~~band intensity of the ν_2 and ν_1/~~line intensity of many strong lines - especially in the ν_3 ~~fundamentals observed in channels A and AB, respectively, are~~ band - have been declared to be less than 2% ~~by spectroscopic experts (J.-M- Flaud, M. Birk, pers. comm.),~~ (Wagner et al., 2002), the problem has been reassessed, and various potential reasons for the deviations have been examined.

In Sections 2-3 we shortly describe the MIPAS experiment and the retrieval setup, followed by a presentation of the ozone profiles resulting from retrievals using the MIPAS pf3.2 spectroscopy in Section 4. In Sections 5-6 we show investigations, which widely exclude forward modelling, instrumental or calibration issues ~~. Thereafter~~ as reason for the observed bias. In Section 7.1 we apply the ozone line data of various versions of the HITRAN database~~(Section 7.1)~~. In Section 7.2 we present a comparison between retrievals using ozone lines of a completely different database, namely GEISA-2015 (Jacquinet-Husson et al., 2016), and retrievals ~~basing~~ based on the HITRAN-2008 data. ~~Further~~ Then we demonstrate the possibility to reduce the channel AB-A differences by changing the air-broadening coefficients (Section 7.3). In Section 8 we show internal inconsistencies of air-broadening coefficients in the HITRAN-2008 database, followed by a summary and conclusions in Section 9.

2 Instrument description and retrieval setup

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), ~~operated onboard~~ which was operated on board the ENVIronmental SATellite (ENVISAT) between 2002 and 2012, has e.g. been described in Fischer et al. (2008). Therefore we ~~here~~ give only a short description of the instrument. MIPAS was a limb-viewing Fourier transform infrared emission spectrometer covering the spectral region between 685 and 2410 cm^{-1} (4.1–14.6 μm). From June 2002 to April 2004 MIPAS was operated in its original high resolution (HR) mode and since January 2005 in reduced resolution (RR) mode. We present retrievals based on data of the RR nominal measurement mode, which ~~consists~~ consisted of rearward limb-scans covering the altitude region between 7 and 72 km within 27 altitude steps. The level-1B radiance spectra used for retrieval are data version 5.02/5.06 and 7.11 (re-processed data) provided by the European Space Agency (ESA) (Nett et al., 2002). The ~~respective~~ IMK notation for ~~the spectra~~ these spectra versions is V5R and V7R.

To reinvestigate the channel AB-A bias in retrieved ozone, retrievals using the processor of the Institut für Meteorologie und Klimaforschung and the Instituto de Astrofísica de Andalucía (IMK/IAA~~processor~~) were performed for 59 MIPAS orbits from January 3, April 1, July 2 and October 2-3, 2009. This processor uses the Karlsruhe Optimized and Precise Radiative Algorithm (KOPRA) (Stiller, 2000) for radiative transfer calculations and the Retrieval Control Program (RCP) of IMK/IAA for inverse modelling of spectra. Two microwindow setups were used, one in the spectral range of MIPAS channel A and the other in the range of channel AB (Tables **??**, **??**). Both of the setups consist of the large number of 30 MWs to obtain a high vertical resolution. The microwindows used in channel A span the wavenumber region 687–791 cm^{-1} covering the fundamental ν_2 band and those of channel AB the region 1028–1164 cm^{-1} covering the fundamental ν_1 and ν_3 bands, respectively. The strong lines of the ν_1 and ν_3 bands are suited for ozone retrieval in the middle atmosphere, but especially the ν_3 lines become saturated for limb scans through the ozone concentration maximum at $\sim$ 28 km and through the lower stratosphere, where the ν_2 lines are a suitable alternative. Consequently, the first four microwindows covering the central part of the ν_3 band are mostly omitted at these altitudes in the MW selection for channel AB retrievals because of saturation (Table ??). In addition to the lines of the fundamental bands, the spectral regions used for retrieval contain a large number of ozone lines from higher transitions.

Except of the use of dedicated microwindows restricted to MIPAS channels A or AB the setup is the same as for IMK retrieval version O3_V5R_220, consisting in a joint-fit of ozone, microwindow-dependent continuum profiles and a microwindow-dependent, but height constant spectral offset. Temperature-, pressure- and H_2O-profiles required for forward modelling were taken from preceding retrieval steps, and the profiles of the remaining interfering species were taken from the climatology provided by Remedios et al. (2007). For all but one retrieval tests presented here spectra of version V5R of the reduced spectral resolution period were taken. Since the subsequent set of MIPAS spectra (version V7R) presumably was produced using an improved calibration ~~schema~~scheme, one additional channel AB-A intercomparison was performed on the basis of this data set. While the spectroscopic data for ozone were changed in ~~different~~ several tests, the same line lists were always used for all other gases. More information on trace gas retrieval from MIPAS data as performed at IMK can be found in various papers, e.g. in von Clarmann et al. (2003) or in Höpfner et al. (2004).

3 Error estimates of ozone line and band intensities

As specified by Flaud and Piccolo (J.-M. Flaud, pers.comm.) the relative error SX in line intensity of ozone lines of the fundamental ν_1, ν_2 and ν_3 bands in the MIPAS database can be parameterized as follows:

$$\mathrm{SX} = 0.02 \times (1 + \mathrm{JU}/70 + \mathrm{KU}/25), \quad (1)$$

where JU is the upper state rotational J quantum number and KU the upper state rotational K quantum number. For the other transitions ending at the ground state the error is

$$\mathrm{SX} = 0.03 \times (1 + \mathrm{JU}/60 + \mathrm{KU}/20). \quad (2)$$

For higher transitions the relative error gradually increases up to

$$\mathrm{SX} = 0.10 \times (1 + \mathrm{JU}/35 + \mathrm{KU}/11) \quad (3)$$

for the transitions ending at strongly excited lower states.

The error entries estimates for ozone line intensities in the HITRAN-2008 database are of similar magnitude (Rothman et al., 2009). In the region of the channel A microwindows they are 1–2% for the strongest lines, 2–5% for lines of medium strength and 5–10% for weak lines. For the lines in channel AB no error estimates are given for line intensities. The HITRAN-2008 error estimates for the air-broadened halfwidths vary between 2–5% for the strongest lines and 10–20% for weak lines.

As mentioned above, Due to the high accuracy in line intensities as outlined in the Introduction, a potential systematic offset between the ozone bands in MIPAS channels A and AB is can be assumed to be lower than 2%. This is also justified by the fact that one of the spectroscopic data sets , which that were used as basis for the ozone line lists in the MIPAS database as well as in all recent HITRAN compilations (version 2004 and later ones), consists in simultaneous laboratory measurements of the ν_1, ν_2 and ν_3 band regions (Wagner et al., 2002).

4 Retrievals using the MIPAS spectroscopy

As already mentioned in the Introduction, a dedicated spectroscopic database for analysis of MIPAS data was established by Flaud et al. (2003b). Starting point for this database was the HITRAN-1996 edition. The new line lists were validated by comparison between atmospheric simulations and ATMOS (Atmospheric Trace Molecule Spectroscopy) as well as MIPAS measurements. The focus of these investigations was a spectroscopic update of the main target species of MIPAS, namely H_2O, CO_2, O_3, CH_4, NO_2 and HNO_3. The ozone spectroscopy was updated on the basis of three sets of highly consistent experimental data (Flaud et al., 2003a). More specific, a new compilation of the fundamental ν_1, ν_2 and ν_3 bands was created and complemented by all higher transitions contained in the HITRAN-1996 database. Since the line intensities in the fundamental ν_1 and ν_3 bands of the new data set were about 4% lower than the corresponding intensities in HITRAN-1996, the line intensities of all bands adopted from HITRAN-1996 were divided by 1.04.

Figure **??**a shows average ozone profiles resulting from retrievals in MIPAS channels A and AB using the ozone linelist of the MIPAS database version 3.2(Flaud et al., 2003b), , which has also been applied in an earlier investigation (Glatthor et al., 2006). This ozone line list is also applied for operational ozone retrieval at IMK/IAA. Averaging was performed over all geolocations of the 59 evaluated orbits . (cf. Section 2). Similar as in the the previous investigation, use of the MIPAS spectroscopy leads to systematically higher ozone values using MIPAS channel AB microwindows as compared to channel A MWs. The absolute differences are largest in the height region 28–45 km and amount to 0.4 ppmv at 36 km altitude (top rightFig. **??**b), which corresponds to a relative difference of 6% (second panel, left). Fig. **??**c). This difference is larger than the relative error in line intensity given in Eqs. 1 and 2 for the strongest and medium scale ozone lines (at least for transitions with low to medium-sized rotational quanta JU and KU). The large scatter of the relative differences below 20 km is caused by the much lower ozone VMRs and by averaging of increasingly less values due to cloud filtering. For different latitude bands or seasons (Fig. **??**d) or seasons (Fig. **??**e) the relative differences in the altitude range 30 to 45 km vary between 5 and 7.5%(second panel, right, and third panel, left).

The vertical resolution in terms of full width at half maximum (FWHM) in channel AB is up to 1 km worse than in channel A between 10 and 35 km altitude, nearly the same between 35 and 40 km and up to 0.5 km better above 40 km (Figure **??**, bottomFigs. **??**f, **??**g). This shows the somewhat better appropriateness of channel A microwindows for ozone retrieval in the lower stratosphere and of channel AB microwindows in the upper stratosphere and mesosphere. The effect of the differences in height resolution on the retrieved VMRs was tested by application of the averaging kernels from channel A retrievals to channel AB profiles and vice versa. This led to negligible changes of the profiles only (not shown).

The corresponding ozone profile of the most actual IMK ozone data version using V5R-spectra (V5R_O3_224.1) is practically identical to the channel A result presented here. The reason is a nearly identical set of microwindows used for the standard retrieval. The almost complete restriction to channel A microwindows (except of two channel AB MWs used above 50 km) resulted from a validation study (Laeng et al., 2014). This study had shown that the earlier ozone data version V5R_O3_220.1 was biased high in the altitude range 35–45 km due to a higher fraction of channel AB microwindows, applied at 36 km and above.

5 Exclusion of forward modelling issues

Similar ozone retrievals with microwindows situated in MIPAS channels A or AB were also performed with the Bologna Geo-fit Multitarget Retrieval Model (GMTR), which uses a ~~completely~~ different forward model (Carlotti et al., 2006). The differences between channel A and AB retrievals are very similar to those resulting from the IMK/IAA retrievals (Figure **??**). This agreement widely excludes the hypothesis that the bias is caused by deficiencies in the ~~Karlsruhe Optimized and Precise Radiative transfer Algorithm (KOPRA)~~ KOPRA forward model used at IMK~~(Stiller, 2000)~~. Nevertheless, a number of possible forward modelling issues has been investigated and is discussed below.

Channel-dependent accuracy parameters: To check ~~,~~ if the higher ozone amounts retrieved in MIPAS channel AB are caused by disproportionately high rejection of weak lines by KOPRA in modelling of the absorption coefficients in this spectral region, channel AB retrievals were ~~performend~~ performed with strongly increased accuracy in calculation of the absorption coefficients. ~~These~~ This leads to better consideration of weak lines. However, these retrievals resulted in nearly identical ozone profiles (not shown).

Channel-dependent continua: Since the continuum profiles fitted in the first four microwindows of the dedicated channel AB occupation matrix often exhibit unphysical negative excursions in the altitude region 35 to 40 km, additional channel AB retrievals were performed without these microwindows. The effect on the retrieved ozone profiles was negligible (not shown).

Non-local thermodynamic equilibrium (NLTE) effects: Another possible reason for the bias in ozone VMRs could be different strengths of NLTE effects in MIPAS channels A and AB. Like for standard MIPAS ozone retrievals from nominal mode measurements, time consuming modelling of NLTE effects was not taken into account for the channel A and AB retrievals. This is generally justified, because these effects are mostly small in the stratosphere, and spectral regions subject to NLTE effects are avoided in the microwindow selection. Nevertheless, channel A and AB ozone retrievals including modelling of NLTE effects had been performed by Glatthor et al. (2006), which had shown that neglect of NLTE modelling is not the dominant reason for the channel AB-A bias. Such calculations were not repeated here. Instead, since NLTE conditions generally persist during daytime, averaging was performed separately for day- and nighttime profiles of the data set (Figure **??**). It is evident that the channel AB-A differences are nearly the same for the whole data set as well as for the day- and ~~nightime~~ nighttime measurements. According to this estimation, NLTE effects have only little influence on the observed differences.

Non-Voigt line shape: As commonly done in radiative transfer calculations for spaceborne mid-IR measurements, line modelling with ~~the IMK/IAA processor~~ KOPRA is performed assuming a Voigt line shape. This assumption is confirmed by Tran et al. (2010), who showed that for the entire 10μm ozone band non-Voigt line shape effects~~(~~, represented by a speed dependent Voigt model~~) .~~, lead to errors in retrieved atmospheric ozone of less than 1%. These investigations were based on calculated as well as on measured spectra obtained by limb-viewing solar occultation and emission ~~mesurements.~~ measurements. Therefore we conclude that the channel AB-A bias for the most part can not be explained by neglect of non-Voigt effects.

6 Exclusion of instrumental and calibration issues

To exclude instrumental or calibration issues, ozone retrievals using channel A and AB microwindows were also performed for measurements of the MIPAS-balloon instrument (Friedl-Vallon et al., 2004), i.e. for a completely independent experiment with different level-1 processing and calibration procedures (Figure **??**). The balloon spectra were obtained on 31 March 2011 over Esrange, Sweden (67.9°N, 21.1°E) and on 14 June 2005 over Teresina, Brazil (5.1°S, 42.9°W). These retrievals resulted in ~~differences~~ channel AB–A differences of 0.5 ppmv in the altitude range of 30–40 km, which are similar to those found in the ~~the~~ spaceborne MIPAS observations. This agreement largely excludes inconsistencies in calibration of channel A and AB spectra, different detector alignment or instrumental line shape issues in the spaceborne MIPAS data. Nevertheless, instrumental and calibration issues have been investigated~~and are summarized~~ , and the most important ones are shortly discussed below.

Spectral calibration issues: To check deficiencies in spectral calibration, ozone retrievals were also performed with MIPAS spectra version V7R, which had been generated with an improved calibration scheme. However, this test resulted in even somewhat larger channel AB-A differences than retrievals with MIPAS V5R spectra (cf. Figure **??**).

Line of sight issues: The vertical field-of-view of the MIPAS experiment is assumed to be the same for each channel. However, due to detector misalignment ~~,~~ the effective line of sight might vary between the different MIPAS channels. Although a detector misalignment is widely excluded by the similarity of the ~~MIPAS-B~~ MIPAS-balloon results, this problem has been investigated. It turned out that the field-of-view of channel AB ~~had~~ would have to be shifted upward by more than 500 m to remove the channel AB-A differences in the height region 33-40 km (not shown). This shift is much larger than the instrumental requirement for inter-channel co-alignment, which is 1.3 mdeg or 68 m, and therefore rather unrealistic. Thus, this investigation confirms that inter-channel misalignment is not the cause of the channel AB-A bias.

7 Retrievals using additional spectroscopic databases

Since deficiencies in forward modelling, instrumental and calibration issues ~~as well as all other mechanisms investigated~~ can largely be ruled out as reason for the bias between channels A and AB, inconsistencies in spectroscopic data practically are the only explanation left. For this reason we performed additional ozone retrievals using different HITRAN versions and the GEISA-2015 database to check if there is any line list, which produces more consistent ozone profiles for channel A and AB retrievals.

7.1 Comparison of different HITRAN-versions

Figure **??** shows ~~MIPAS~~ average ozone profiles and channel AB-A differences for retrievals using ozone linelists of versions 1996, 2004, 2008 and 2016 of the HITRAN spectroscopic database (Gordon et al., 2017, and references therein) as well as of the MIPAS spectroscopy. Retrievals performed with ozone line data of HITRAN versions 2000 and 2012 are not shown, because ~~the former line list is~~ these linelists are practically identical to HITRAN-1996 and ~~the latter~~ to HITRAN-2008, respectively. HITRAN versions 2004, 2008 and 2016 ~~also~~ lead to nearly the same channel A (Fig. **??**a) and channel AB profiles (Fig. **??**b) as the MIPAS spectroscopy. Therefore these HITRAN versions also result in almost the same channel AB-A ~~differences in the altitude range 15–50 km.~~ differences (Figs. **??**c,d). For each of these databases the maximum difference is 0.5 ppmv (7%) at the altitude of 36 km, which is even somewhat larger than the bias resulting from the MIPAS spectroscopy. Consequently, these differences are also larger than the relative errors in line intensity given in Eqs. 1 and 2. Only the HITRAN-1996 spectroscopy leads to a ~~significantly~~ somewhat different result, namely positive channel AB-A differences of up to 0.5 ppmv above 32 km, but negative differences of up to -0.4 ppmv below this altitude. Similar results ~~for~~ in case of use of the HITRAN-1996 spectroscopy have already been shown by Glatthor et al. (2006). Since the correction scheme of detector nonlinearities is assumed to be improved ~~for~~ in generation of MIPAS V7R spectra, an additional retrieval test ~~has been performed using these spectra~~ was performed using this dataset and HITRAN 2008 line data. However, these spectra lead to even somewhat larger differences (yellow curve) than the V5R spectra.

The rather good agreement between the different channel A as well as channel AB retrievals indicates largely consistent spectroscopic parameters of corresponding ozone lines in the MIPAS spectroscopy and the HITRAN-2004 database as well as in later HITRAN versions for the spectral range of the channel A and AB microwindows. Therefore a comparison between the line parameters of the MIPAS and HITRAN databases is not presented here, but provided as supplemental material only.

7.2 HITRAN-2008 versus GEISA-2015

Since the channel AB-A bias ~~did not disappear~~ could not be removed by use of any of the HITRAN line lists, we performed an additional retrieval test with a different spectroscopic database, namely the Gestion et Etude des Informations Spectroscopiques Atmosphériques - version 2015 (GEISA-2015) compilation (Jacquinet-Husson et al., 2016) and compared the results with those ~~basing~~ based on HITRAN-2008. Although the line parameters of the three fundamental ozone bands in GEISA-2015 are principally obtained from the same sources as those in HITRAN-2008 (Jacquinet-Husson et al., 2008, and references therein), we found considerable differences.

7.2.1 Retrieval results

Figure **??** shows average ozone profiles resulting from retrieval in MIPAS channels A and AB using the ozone line lists of HITRAN-2008 (cf. Figure **??**) and of GEISA-2015. The GEISA-2015 database leads to nearly the same profile as HITRAN-2008 for retrievals using the channel AB microwindows, but to even higher ozone VMRs for retrievals using the MWs in channel A, i.e. to a reversal of the bias obtained with the HITRAN line data. The differences between channel AB and A profiles ~~caused by~~ obtained with the GEISA-2015 spectroscopy are negative in the height region 20–43 km, amounting up to -0.55 ppmv or -7.5% at 28 km altitude. Moreover, the differences between the channel A retrievals, which definitely have spectroscopic reasons, are even larger and amount up to 0.8 ppmv or about 10%. With regard to the validation study by Laeng et al. (2014) the channel A profiles obtained with the GEISA-2015 spectroscopy ~~ar~~ are most probably biased high.

7.2.2 Comparison of spectral parameters

The retrieval results indicate mostly consistent spectral parameters in HITRAN-2008 and GEISA-2015 for the ozone lines used in MIPAS channel AB, but considerable spectroscopic differences in the region of the channel A microwindows. In the ~~follwing, we~~ following, we will compare the HITRAN-2008 and GEISA-2015 ozone lines applied in channel A as well as in channel AB to identify the parameters responsible for ~~the~~ these differences.

First of all we compared the number of ozone lines of the two databases in the spectral region covered by the ~~channnel~~ channel A microwindows. In this wavenumber region the GEISA-2015 database contains 3631 lines, all of them having a corresponding line in the HITRAN-2008 edition. The latter contains 734 additional lines, which however are rather weak. Their intensities are between 2.013×10^{-26} and 5.858×10^{-24} cm^{-1}/(molecules cm^{-2}), while a considerable number of the lines contained in both databases have intensities between 1×10^{-22} and 1.66×10^{-21} cm^{-1}/(molecules cm^{-2}). ~~Unsurprisingly, a~~ A test retrieval without these additional lines (not shown) resulted in nearly the same ozone profiles as the retrieval using the complete HITRAN-2008 ozone linelist. In the spectral range of the channel AB microwindows the GEISA-2015 and HITRAN-2008 databases contain 3737 and 3804 lines, respectively. The number of corresponding lines is 3724, i.e. the HITRAN-2008 edition contains 80 lines, which are not in the GEISA-2015 compilation. Again, these lines have intensities below 9.21×10^{-25} cm^{-1}/(molecules cm^{-2}) only, while a lot of the lines, which are available in both databases, have much higher intensities between 1×10^{-22} and 3.9×10^{-20} cm^{-1}/(molecules cm^{-2}). In summary, the strong lines which make sizable contributions to the spectra are included in both data sets, and the missing weak lines are ruled out as cause of the discrepancy under investigation.

The spectral parameters which have the largest potential to cause the disagreement in channel A are line positions, line intensities or air-broadened halfwidths. Further parameters required for line modelling are lower state energies, the coefficients of temperature dependence of the air-broadened halfwidths and air-broadened pressure shifts. A comparison ~~of line positions showed exact coincidence~~ shows that the line positions and lower state energies of all GEISA-2015 and HITRAN-2008 lines inside the ~~MWs in channel A~~ channel A microwindows agree exactly. Inside the channel AB MWs these parameters differ slightly between the two databases, but for a small number of very weak lines only. ~~The same applies to the strong lines in channel AB .~~ Compared to air-broadening, the relative contribution of self-broadening is of the order of 10^{-5} only and thus negligible. The pressure shift of the ozone lines in GEISA-2015 and most of the HITRAN-2008 lines used in channels A and AB is zero. About one third of the HITRAN-2008 lines have weak shifts of only -0.0008 and of -0.0007 cm^{-1} in channel A and AB, respectively. Thus, differences in pressure shift are also negligible. The remaining parameters are line intensities and air-broadened halfwidths.

~~In~~ Figure **??** ~~(top) the~~ shows the relative differences between the intensities of the ozone lines of the GEISA-2015 and the HITRAN-2008 database used for channel A and AB retrievals~~of the GEISA-2015 database are plotted against the corresponding values of the HITRAN-2008 edition. The bottom panel shows the respective relative differences.~~. It is evident that the intensities of the strongest lines used in channel A are practically identical. There are slight deviations of 5% for a part of the weaker lines only. This indicates that the differences in channel ~~A-retrievals~~ A retrievals do not result from inconsistent line strengths. The respective ~~scatter~~ difference plot of the lines used in MIPAS channel AB also shows nearly identical intensities of the strongest lines. Again there are differences of +5% for weaker lines and larger differences of up to ~~-30~~ ±25% for very weak lines. However, the good agreement of the channel AB profiles (Figure **??**) shows, that a potential influence of the different line strengths of the weak lines is low.

~~In a similar manner~~ Next we correlated the air-broadened halfwidths $\gamma_{air,0}$ of the ozone lines used for channel A and AB retrievals of the two databases, colour-coded by the respective line strengths (Figure **??**). In both cases the air-broadened halfwidths of the HITRAN-2008 ozone lines are larger than those of the GEISA-2015 compilation, but there are clear systematic differences between the channel A and channel AB correlations for the strongest lines. In channel AB the air-broadened halfwidths of the strongest lines are largely identical. However in channel A the $\gamma_{air,0}$-values of the strongest lines are significantly lower (∼13%) in the GEISA-2015 database. Differences between line broadening parameters in the GEISA and HITRAN databases have already been reported by Jacquinet-Husson et al. (2008, 2011).

The air-broadened halfwidths $\gamma_{air,0}$ given in the line databases are reference values for ~~p0~~ p_0 = 1013.25 hPa and ~~T0~~ T_0 = 296 K. The γ_{air}-values for actual temperature ~~T and pressure p~~ T and pressure p are calculated as follows:

$$\gamma_{air}(p,T) = \gamma_{air,0} \times p/p_0(T_0/T)^n ,. \tag{4}$$

~~where the~~ The coefficients n of temperature dependence are also given in the spectroscopic databases. Figure **??** shows a scatter plot of the coefficients n of GEISA-2015 versus HITRAN-2008, again colour-coded by line strength. In channel A the coefficients ~~n~~ n of the strongest lines are nearly equal ~~in~~ for both databases. Thus there is no compensation of the large differences in the air-broadened halfwidths of channel A via the coefficients ~~n~~n. In channel AB there are somewhat larger differences of some of the coefficients n belonging to the strongest lines, namely the coefficients at the ordinate value of 0.76. However, since the channel AB profiles are nearly identical, these deviations obviously have no large effect. For example, ~~adjustment~~ shifting of the HITRAN-2008 values of 0.71 of several strong lines (red pluses) to the main diagonal changes γ_{air} by 1.3% only for a stratospheric temperature of 230 K.

The correlation analysis gives strong indication that the differences of the ozone VMRs resulting from retrieval in channel A using HITRAN-2008 ~~and~~ or GEISA-2015 data are not caused by differences in line strengths but rather by the differences between the air-broadened halfwidths of the strongest lines. To check this assumption we replaced the $\gamma_{air,0}$-values and the coefficients ~~n~~ n of temperature dependence in the GEISA-2015 database by the respective HITRAN-2008 values and performed additional retrievals using these line parameters. Figure **??** shows the ~~original and the new retrieval results for channels A and AB~~channel A and AB retrieval results using the original and the modified linelist. After modification of the GEISA-2015 data the average ozone profile retrieved in channel A is nearly identical to the profile resulting from the HITRAN-2008 spectroscopy. Furthermore, the small differences between the retrievals in channel AB are even more reduced. These results confirm This result confirms that the bias in channel A-retrievals A retrievals between use of the HITRAN-2008 and of the GEISA-2015 database is caused by the differences in air-broadened halfwidths. Furthermore, the small differences between the retrievals in channel AB are even more reduced.

7.3 Channel AB retrievals using modified HITRAN-2008 lines

Since the relative differences between the intensities of the ozone bands in MIPAS channels A and AB are declared assumed to be very small (1-2%), we posit that a considerable part of the line database-internal channel AB-A biases might be caused by inconsistent $\gamma_{air,0}$-values as well. To check , We performed two additional tests to check if the differences can be reduced by modification of the air-broadened halfwidths, we performed two additional tests. In the first test we increased the halfwidths of the HITRAN-2008 ozone lines in the spectral region of MIPAS channel AB by 0.005 cm^{-1}/atm@296K (5–7%). Caused by this change the deviations in the height range 32–45 km are considerably reduced to 0.15 ppmv or less as compared to the retrieval using the unmodified HITRAN-2008 database, namely to 0.15 ppmv or less (Figure **??**). On the other hand there are now larger negative deviations of up to -0.2 ppmv in the height range 18–32 km. In the second test we increased the halfwidths of the strongest lines on the main diagonal in Figure **??** (right) by 0.008 cm^{-1}/atm@296K to obtain a similar distribution like the channel A correlation in Figure **??** (left). This led to even lower differences between 18 and 32 km, but to somewhat larger negative differences between 18 and 32 km.

As a side aspect, Figure **??**d shows the differences between the channel AB retrievals with increased air-broadened halfwidths and the channel AB retrievals using the unmodified HITRAN-2008 spectroscopy. Since these differences are relatively constant around -4 and -5% over the altitude region 10–40 km, they are good estimates of the respective changes in ozone column amounts. Thus, this result shows that, alternatively to the proposed re-scaling of line intensities in the 10μm region (cf. Section 1), the bias between ozone column amounts measured in the mid-infrared and UV spectral regions as e.g. shown by Schneider et al. (2008) could probably also be reduced by change of the air-broadened halfwidths.

8 Additional investigations

To substantiate the ~~assumption~~ possibility of inconsistent $\gamma_{air,0}$-values, Figure **??** shows modelled ozone spectra using the HITRAN-2008 and the MIPAS spectroscopy for a tangent altitude of 30 km. In the difference plot (Fig. **??**c) a number of ~~conspicious lines~~ conspicuous lines belonging to the fundamental ν_2 band can be identified between 797 and 830 cm^{-1} ~~. These lines belong to the fundamental ν_2 ozone band. Figure **??** shows that the~~ (note that these lines are not contained in our set of channel A microwindows). The line intensities of these transitions are ~~nearly equal in both spectroscopies~~ identical in both spectroscopic line lists and thus not responsible for the deviations between the model spectra. However, there are substantial differences between the air-broadened halfwidths (Figure **??**). The halfwidths of the lines of the MIPAS database continuously decrease between 700 and 850 cm^{-1}. The HITRAN-2008 values are slightly larger, but decrease in a comparable manner up to $\sim$790 cm^{-1}. However, there is a strong increase ~~between 790 and~~ of more than 0.01 cm^{-1}/atm@296K at $\sim$797 cm^{-1}, leading to significantly larger values up to 850 cm^{-1}. ~~These inappropriate halfwidths (M. Birk, pers. comm.) are~~ This jump in $\gamma_{air,0}$ is the reason for the stronger ozone lines in the model spectrum using HITRAN-2008 data in Figure **??**. This ~~deficiency~~ artefact is still present in later versions up to HITRAN-2016. In addition to the spikes at and above 797.05 cm^{-1} the difference plot (Fig. **??**c) also exhibits somewhat smaller peaks for the ν_2 transitions at lower wavenumbers, e.g. at 789.11, 781.18, 773.29 and 765.43 cm^{-1}. This indicates potential additional spectroscopic inconsistencies below 797 cm^{-1} in the HITRAN-2008 database.

To further demonstrate the inconsistencies of the spectroscopic parameters of the lines above 790 cm^{-1} in the HITRAN-2008 data base identified above, broad-band ozone retrievals were performed in the wavenumber range 795 to 825 cm^{-1} for MIPAS orbits 39680–39693. In addition to ozone, peroxyacetyl nitrate (PAN), CCl_4, $ClONO_2$ and HCFC-22 were jointly fitted. The spectral signatures of additional gases were modelled by use of pre-fitted or climatological profiles. Figure **??** shows the residuals between mean modelled and measured spectra using the HITRAN-2008 and the MIPAS pf3.2 ozone spectroscopy for the latitude region 30°S–30°N and tangent heights covering the altitude region of ∼14–44 km. For the major part of the spectral region the residuals resulting from both spectroscopies are very similar, leading to a nearly complete coverage of the HITRAN-2008-residual. The large "blue" residuals are mainly caused by inadequately modelled CO_2 and H_2O lines. However, for tangent heights of 14, 20 and 30 km use of the HITRAN-2008 spectroscopy leads to large residuals (red) at the positions of the lines with the higher $\gamma_{air,0}$-values (797.05, 805.02 and 812.99 cm^{-1}). The spectroscopic parameters of these lines are not consistent to those of all the other ozone lines in the broad spectral range used for retrieval, and thus these lines can not be fitted properly. At 44 km these residuals have disappeared, showing that the influence of different air-broadened halfwidths becomes negligible at this altitude.

At first glance it seems a bit strange that increased air-broadened halfwidths lead to modelled ozone lines which exhibit stronger peak radiances (Figure **??**, right). To investigate this behaviour more in detail, model calculations with KOPRA were performed for an isolated ozone line. For a homogeneous, optical thin atmosphere larger $\gamma_{air,0}$-values lead to lower radiances in the line center and to higher radiances at the line wings both for the monochromatic line and after folding with the apodized internal line shape (AILS) (not shown). However, for a limb observation through the Earth's atmosphere the result becomes different. For a tangent altitude of 30 km a larger air-broadened halfwidth also leads to a larger halfwidth and lower peak radiance of the modelled monochromatic line (Figure **??**, leftFig. **??**a), but there is a significantly larger growth in radiance at the wings as compared to the decline in the line center . (Fig. **??**c). In this case, convolution with the AILS as performed in MIPAS retrievals indeed leads to a higher peak radiance (Figure **??**, rightFigs. **??**b,d).

9 Summary and conclusions

We have reassessed the bias in the altitude range 30–45 km between ozone retrievals using microwindows in MIPAS channels A (685–970 cm^{-1}) and AB (1020–1170 cm^{-1}). We found that the bias, originally detected in retrievals using V3O-spectra of the MIPAS high-resolution measurement period and the so-called MIPAS spectroscopy, also occurs for retrievals using later versions of level-1B spectra (V5R, V7R) of the MIPAS reduced-resolution mode. The effect amounts up to 8% at the altitude of 36 km. Forward modelling issues as reason for the problem could ~~widely~~ be excluded by the fact that similar differences also resulted from retrievals using the processor of the University of Bologna. Spectral calibration or line-of-sight issues could ~~also~~ largely be excluded, because retrieval results of a different experiment (MIPAS-balloon spectra) also resulted in similar differences. Nevertheless, a number of forward-modelling, instrumental and calibration issues were examined, but did not lead to an explanation. The most plausible explanation left was inconsistencies in spectroscopic data.

Therefore additional retrievals were performed using several editions of the HITRAN database, which however led to rather similar, even somewhat larger channel AB-A differences in retrieved ozone as those resulting from application of the MIPAS line list. One exception is the HITRAN-1996 edition, which causes positive differences above, but negative differences below 32 km. Since the channel AB-A bias did not disappear by application of any of the HITRAN databases, we performed another retrieval test with a different spectroscopic database, namely the GEISA-2015 compilation. For measurements in MIPAS channel AB, the resulting ozone profiles are rather similar to those ~~basing~~ based on the HITRAN-2008 spectroscopy. However, for channel A retrievals the stratospheric ozone VMRs ~~basing~~ based on GEISA-2015 are up to 0.8 ppmv or about 10% larger than those ~~basing~~ based on HITRAN-2008 and even larger than the VMRs retrieved in channel AB. According to the results of the validation study by Laeng et al. (2014) these VMRs are obviously too high. We showed that the differences in channel A retrievals are not caused by inconsistent line intensities, but by ∼13% lower air-broadened halfwidths of the strongest ozone lines in the channel A microwindows in the GEISA-2015 database.

Since the relative differences between band intensities in the spectral ranges used in MIPAS channels A and AB are assumed to be considerably lower than the observed bias of 6–8%, we suggest that a major part of the channel AB-A differences might be caused by inconsistencies in air-broadened halfwidths in the individual line databases as well. To substantiate this assumption we identified several ozone lines in the HITRAN-2008 database, which exhibit too large air-broadened halfwidths. The bias between channel A and AB can partly be reduced by increasing the air-broadened halfwidths of the lines in channel AB. In summary, the air-broadened halfwidths of ozone lines in the spectral regions of MIPAS channel A as well as of channel AB (i.e. the regions of the ν_2, ν_1 and ν_3 fundamental bands) should be reassessed both for the GEISA and for the HITRAN databases. This is especially necessary for the GEISA-ν_2 lines in MIPAS channel A.

According to ~~our investigations~~ the investigations with our microwindow datasets, for the time being the best choice of an ozone line data compilation for evaluation of MIPAS measurements is the MIPAS spectroscopy (Flaud et al., 2003b), because on the one hand the channel AB-A differences are somewhat smaller than those resulting from the HITRAN databases and on the other hand it does not contain the inconsistency in some air-broadened halfwidths identified in HITRAN-2008, which was also transferred to later HITRAN versions. However, as far as ozone is concerned we recommend to use version pf3.2 of the MIPAS spectroscopy and not the latest update pf4.45 ~~,~~ (Flaud, et al., 2015, http://atmos.difa.unibo.it/spectdb/), because the ozone data set in this compilation is identical with HITRAN-2008.

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