**Atmospheric
Measurement
Techniques**
Discussions

Interactive comment on "Tropospheric column amount of ozone retrieved from SCIAMACHY limb-nadir-matching observations" *by* F. Ebojie et al.

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Response to anonymous reviewer #3 (C3015)

We thank the anonymous reviewer #3 for his/her valuable comments, most of which we agree with. We believe that this review has helped us to improve our paper.

Reviewer #3 (Comments to Author):

The manuscript describes a technique to estimate tropospheric ozone columns using limb and nadir measurements from SCIAMACHY and shows comparisons of SCIAMACHY-derived tropospheric ozone columns with those from ozonesondes and other satellite retrievals. The topic is relevant to AMT. The paper could be published in AMT, but only after revision.

Reply: We thank the reviewer for his/her encouraging comments.

Major points: 1.The estimation of errors is admirable and in some ways goes beyond what has been done in previous studies; however, key assumptions are questionable. Several of the error terms given in Eq. 6 will not produce Gaussian errors (for example cross section, aerosol, and tangent height errors). Therefore, using a root-sum-square approach to get an estimate of the total error is not appropriate and will lead to an underestimate of errors.

Reply: Thank you for this point. Errors in aerosol have both systematic and random contributions to the retrieved stratospheric ozone data product but as recommended we have separated the systematic (err1) and random (err2) parameters error contributions to the retrieved stratospheric ozone columns as summarized below: err1=abs(err in cross-section) + abs(err in aerosol) + abs(err in tangent height) err2=sqrt[(err in albedo)ˆ2 + (err in pressure)ˆ2 +(err in temperature)ˆ2 +(effect of tropopause height)ˆ2]

err_in_strat.col=sqrt(err1ˆ2 + err2ˆ2) err_in_trop.col=sqrt(err_in_strat.colˆ2 + err_in_tot.colˆ2)

The above expression are now implemented in section 4 of the revised manuscript. We also mentioned in section 4 that the assumption of Gaussian distribution for all errors may lead to an underestimation while an assumption that all errors simply add up overestimates the actual total error.

We also would like to mention that establishing an error budget of such a satellite data product will never be possible in a strict mathematical sense, because full statistical

characterization of the individual error sources is not possible. However, we are convinced that an error budget like the one presented in this manuscript is quite valuable, if all assumptions made are transparent and clearly stated.

2. Furthermore, some of the details in the error analysis section need clarification. There is quite a lot of detail given for the calculation of stratospheric errors, while similar detail is not given for the tropospheric column errors and error due to tropopause height specification. Two tables are given for stratospheric error. Instead of providing these errors as a function of height, they should be given for the stratospheric column. A similar table should be provided for tropospheric column errors. More description of the error due to tropopause height should be given. This error appears to be too small as compared with what has been found by others (see Morris et al., http://scholar.valpo.edu/phys_astro_fac_presentations/5/ who found that particular definitions of tropospause heights can produce large differences in tropospheric column amounts found by residual methods).

Reply: We followed the reviewer's suggestion and provide tables for errors in the stratospheric ozone columns due to uncertainties in atmospheric parameter, registration and spectroscopy (please see Table 2). Table 1 summarizes now the estimated total uncertainty in the stratospheric, total and tropospheric ozone columns.

A detailed analysis of the retrieval of total ozone column (TOC), an error analysis, and validation of the TOC from SCIAMACHY have been published e.g. (Coldewey et al. 2005, Bracher et al. 2005, Weber et al. 2005, Weber et al. 2013). Coldewey et al. (2005) described in detail the different error sources in TOC WFDOAS retrievals. This is mentioned in sections 3.2 and 4.

Here, we investigated the influence of a shift in the tropopause height on the retrieved tropospheric ozone, while an investigation of the influence of the methods used to calculate the tropopause is beyond the scope of this study.

3. The latter point raises serious concerns regarding the comparisons between the

different methods. A long description is given as to how the tropopause height is computed for the SCIAMACHY retrievals and radiosondes (they are somewhat different in details), but no description is given for the OMI/MLS or TES retrievals.

Reply: Thank you for this point. In section 2.2 of the revised manuscript, we have added the discussion on tropospheric ozone column retrievals from TES and OMI/MLS and where appropriate citations were inserted. We cite Ziemke et al. (2006), which gives a detailed description of the tropospheric ozone columns retrievals from OMI/MLS. For tropospheric ozone column retrieval from TES, we cite Bowman et al. (2006).

4. Differences in tropopause definitions can lead to significant differences in computed tropospheric columns. The dependence of tropospheric columns on tropopause height can lead to confusion when comparing different data sets or analyzing the data, particularly in terms of seasonal variability as well as its interpretation (for example, how much of the tropospheric column variation is due to changes in the tropopause height as compared with chemical or transport mechanisms? The latter may be measured with the satellite instruments while the former is not, but is only due to how the specified tropopause changes). To avoid this issue, several studies have instead compared mixing ratios (see for example the Zhang et al., 2010 reference below) or integrated amounts up to a fixed pressure (Schoeberl et al., 2007).

Reply: Yes we agree that the differences in tropopause definitions can lead to significant differences in computed tropospheric ozone. We have included the following sentences in section 5.1 of the updated manuscript: "The difference in the tropospheric ozone columns might be due to different tropopause heights employed in the retrievals from the different instruments. Differences might also come from retrieval algorithms in terms of a-priori value, cloud treatment and air mass factors employed by the different satellite instruments. Comparison of tropospheric ozone columns from satellites with the values from ozonesonde, which are sparse, are prone to noise mostly in dynamically active subtropics where rapid fluctuation in tropopause heights occur. Such comparisons exhibit a lot of scatter and the regression line deviates from the line of

unity."

As suggested in point 6 below, the comparison plots between ozonesondes and the satellite instruments have been changed to reflect the tropospheric ozone anomaly from different instruments, which are better to compare (please see figures 6 - 11). The tropospheric ozone data product from SCIAMACHY is derived in columns. We do not have tropospheric ozone profile information, therefore we cannot implement mixing ratios or integrate the ozone amount up to a fixed pressure. For our analysis we used OMI/MLS and TES tropospheric ozone columns.

5. The comparisons carried out in the paper do not make any attempt to account for differences due to a priori information. This can be accomplished in a straight-forward manner with the averaging kernel approach that is used in TES validation and other papers. The Zhang et al. (2010) reference below shows that when comparing tropospheric ozone data sets with different a priori information and sensitivities, these factors can lead to artificially large differences if not accounted for. The comparisons in the present work leave the reader with questions as to why there are differences and do not provide insight into the differences. Proper account of a priori differences should be done and may decrease actual differences between the data sets, leading to greater confidence in the SCIAMACHY results. As it stands, the comparisons with the other data sets are not particularly useful, particularly within the context of previous results, except to show that basic well-known features are captured by SCIAMACHY (most of the time). However, there are some notable differences between the different data sets as noted in the other reviews, leading to concerns regarding the SCIAMACHY data set.

Reply: Thank you again for this point. The comparison plots have been modified (please see figures 6 - 11) as suggested in point 6 below. Differences between the instruments are analyzed in more detail. We also discussed in section 5.2 of the revised manuscript that the differences in the comparison between different data products could be due to differences in the retrieval algorithm, cloud treatment, etc. The residual method employed by SCIAMACHY doesn't use any a-priori knowledge about

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the tropospheric ozone.

Averaging kernel approach is useful when comparing datasets with different vertical resolution. In our study we have used tropospheric ozone data products from all three instruments which have similar vertical resolution providing only one piece of information in the troposphere. The only case where this approach is needed are comparisons with ozonesonde results. In this case the suitable averaging kernels are represented by a box-car function, i.e. an integration of the sonde profiles throughout the entire troposphere is done.

The impact of the a-priori profile on the stratospheric ozone profiles retrieved from limb-scatter observations is rather limited, because the a priori is updated with the output of the last iteration in the iterative retrieval scheme, unlike in the original optimal estimation scheme by Rodgers.

Longitudinal line plots that show the variability and differences between the three satellite instruments for different seasons are included in the manuscript (please see Fig. 14). These plots clearly show that differences between the tropospheric ozone data products from the different satellite instruments depend on season and longitude and vary, but the only prominent feature is that OMI/MLS seem to systematically underestimate the tropospheric ozone data.

6. In figures that focus on seasonal variations, it would be useful to remove biases before comparing the different data sets.

Reply: Thank you for pointing this out, we followed the reviewer's suggestion and created Figures 6 – 11, which has the biases in the different data products removed. These figures have now been used to replace figures 6 – 13 in the previous manuscript.

7. There is no discussion of the treatment of cloudy data in the TES and OMI/MLS data sets. The SCIAMACHY data are filtered to remove cloudy data, so there may also be a clear-sky to all-sky bias. Some discussion on this point must be added.

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Please see and reference Zhang, L., Jacob, D. J., Liu, X., Logan, J. A., Chance, K., Eldering, A., and Bojkov, B. R.: Intercomparison methods for satellite measurements of atmospheric composition: application to tropospheric ozone from TES and OMI, Atmos. Chem. Phys., 10, 4725-4739, doi:10.5194/acp-10-4725-2010, 2010.

Reply: Thank you for this point, cloudy data are rejected in both SCIAMACHY and OMI/MLS tropospheric ozone retrievals (Ziemke et al., 2006, Ziemke et al., 2011) in a similar way. In tropospheric ozone retrieval from TES, cloud top pressures and cloud effective optical depth are used to screen the data as discussed by Kulawik et al., (2006b) and Eldering et al., (2008). Furthermore, as mentioned in section 5.1 of the revised manuscript, the limited sampling by TES might result in a bias, which is similar to "cloud free" to "cloudy-sky" bias.

We followed the reviewer's suggestion and cited Zhang et al, (2010). We have included in section 2.2 of the revised manuscript, some discussion on the retrievals of tropospheric ozone column from TES and OMI/MLS and also cited some references where appropriate.

8. The paper contains many grammatical errors and typos; I did not point out all of them (stopped midway through the paper). As this is a major distraction, copy-editing is recommended. Parts of the paper (for example, the introduction) would benefit from reorganization to improve the flow. There are also a number of subjective and even incorrect statements. These must be cleaned up before publication.

Reply: Thank you for pointing this out, we tried to correct all the typos and grammatical errors that were found.

Technical points: 9. Sometimes SCIAMACHY is referred to as in the present as in the abstract "which flies as part of: : :" and other times as in the past as in the first sentence of the introduction. As it is unlikely that the satellite will be revived, it would make more sense to consistently refer to it as in the past.

Reply: This is noted and we have used the past tense.

Introduction 10. 1st par.: "Retrieval of...yields important trace constituents" -> "Retrievals from: : : yield important information about trace constituents"

Reply: We have modified the statement as suggested.

11. 4th par. This paragraph consists of one very long sentence. Suggest breaking it up.

Reply: Thank you for this point, we have divided the paragraph as suggested.

12. 5th par. "Enhanced sensitivity" with respect to what? What does "good global coverage" mean exactly (this is a subjective statement). What is meant exactly by "current generation of sensors"? Does it mean SCIAMACHY and others? If so, say exactly which sensors.

Reply: We have modified the statement as suggested. The phrase "Enhanced sensitivity" has been replaced. The statement "good global coverage" has been reworded

13. 6th par. How do nadir-viewing instruments have a lower probability for cloud interference? Their large fields of view actually have a high probability of cloud interference, but there are established techniques for accounting for the cloud effects. To say these instruments have good horizontal spatial resolution is subjective and some may not agree. Older references (SBUV) exist for this statement and should be added here (they appear later after a discussion of the retrieval of tropospheric ozone by spectral fitting). Again, this paragraph consists of one very long and somewhat unwieldy sentence, so suggest breaking it down.

Reply: Thank you for this point. The nadir foot print of SCIAMACHY is much smaller than the limb foot print. The limb foot print is about 500 km by 240 km. Because of the smaller air volumes sampled in nadir geometry cloud interference in the case of broken cloudiness will be lower than for limb measurements. This is what we intended to state. In case of completely overcast conditions there is of course no difference in

cloud interference between nadir and limb measurements. The proposed references have been included e.g., Heath et al., (1975), Kramarova et al., (2013).

14. 7th par. Some may not agree that tropospheric ozone has a relatively short lifetime. Please give a range of its lifetime instead. Information about sources and sinks of trace gases is not directly acquired by satellites. Models are needed to help interpret the satellite data and derive such information.

Reply: Yes, thank you for this point. We have provided the average tropospheric ozone lifetime, which is about 22 days (Stevenson et al. 2006) depending on altitude, latitude, and season. We have paraphrased the statement in the introduction page of the revised manuscript as follows:

Global retrievals of tropospheric ozone from satellite instruments enable its global amounts and distributions to be studied, as it has in spite of its reactivity a relatively long effective lifetime ($\sim$ 22 days Stevenson et al, (2006) depending on altitude, latitude, and season) and consequent variability. Important information on tropospheric ozone sources, sinks, transport, and seasonal behavior can also be acquired from satellite instruments through chemistry (climate/transport) models.

15. 8th par. A number of studies have been lumped into a category of "different variants of this method" described in the preceding sentence as "using a combination of two different instruments". Not all of the methods use two different instruments and not all would qualify as variants. Please provide more distinction.

Reply: We are thankful for pointing this out. We have rephrased the statements

16. 9th par. 1st sent. Optimal estimation uses everything described in the first part of this sentence and therefore should not be mentioned as a separate technique. Other techniques, such as neural networks, have also been used and should be referenced: see e.g., Sellitto P., B. R. Bojkov, X. Liu, K. Chance and F. Del Frate, Tropospheric ozone column retrieval from the Ozone Monitoring Instrument by means of neural net-

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works algorithms, Atmos. Meas. Tech., 4, 2375-2388, doi:10.5194/amt-4-2375-2011, 2011.

Reply: Thank you for this point. we have removed optimal estimation from the sentence: We have also included other tropospheric ozone retrieval technique and cited the reference suggested by the reviewer.

17. 10th par. The first 3 sentences would make more sense if they were placed before the discussion of the spectral fitting. The technique is essentially the same but with a more limited number of wavelengths so that the information content is not as great as that from instruments with more continuous spectral coverage. The 3rd sentence is long and convoluted. The last few sentences belong in a separate paragraph along with the following paragraph. Here the following work should be referenced: Natraj V., X. Liu, S.S. Kulawik, K. Chance, R. Chatfield, D.P. Edwards, A. Eldering, G. Francis, T. Kurosu, K. Pickering, R. Spurr, H. Worden, Multispectral sensitivity studies for the retrieval of tropospheric and lowermost tropospheric ozone from simulated clear sky GEO-CAPE measurements, Atmos. Environ., 45, 7151-7165. doi:10.1016/j.atmosenv.2011.09.014, 2011.

Reply: We are appreciative to the reviewer for this point. We have moved the first three sentences and separated the third sentence. The last few sentences have been moved to join the following paragraph as requested. We have also included the suggested reference.

18. p. 7816, par. 2: I do not believe the first sentence is true, particularly WRT total ozone. Surface reflectivity data bases are typically used for the retrieval of total and tropospheric ozone from UV observations and because they use wavelengths that are not terribly sensitive to the surface. It could be argued that these climatologies do not need to be highly accurate (and here highly accurate needs to be defined).

Reply: Thank you for this point. We have paraphrased the statement. In the WFDOAS algorithm an effective scene UV albedo (2890 nm) is derived and used in the total

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column retrieval as described in Coldwey-Egbers et al. 2005. The total column error due to albedo is indeed, quite small.

19. The next sentence starts a discussion on thermal infrared techniques and belongs in a new paragraph. Here, references for retrievals from the TES instrument should be included as that instrument was designed specifically to measure tropospheric ozone by resolving absorption lines including pressure broadening effects.

Reply: Thank you for this point, we have moved the sentence to a new paragraph and provided a reference for TES (Beer et al., 2006) as suggested.

20. The 4th sentence starts a discussion on the use of combined UV/thermal approaches. This could go in a new paragraph. Here, also the above reference to Natraj et al., 2011 should be mentioned as well as Worden, J., X. Liu, K. Bowman, K. Chance, R. Beer, A. Eldering, M. Gunson, H. Worden, Improved tropospheric ozone profile retrievals using OMI and TES radiances, Geophys. Res. Lett., 34(1), L01809, 10.1029/2006GL027806, 2007.

Reply: Thanks for this point. We have moved the sentence to a new paragraph and added the reference that was suggested.

21. In the last sentence, it does not appear that the Bovensmann et al. reference applies to the implementation of the method with GOME-2 and IASI. Here, the following work should also be referenced: Fu, D., J.R. Worden, X. Liu, S. S. Kulawik, K. W. Bowman, and V. Natraj, Characterization of ozone profiles derived from Aura TES and OMI Radiances, Atmos. Chem. Phys., 13, 3445-3462, doi:10.5194/acp-13-3445-2013, 2013.

Reply: We have removed Bovensmann et al. and separated the sentences. The suggested reference is also included in the revised manuscript.

22. p. 7816, last par., 2nd sentence: This statement is subjective and it can be argued that it is not true given that many other studies have already characterized stratospheric

inhomogeneity and provided global monitoring of tropospheric ozone.

Reply: Thank you for pointing this out. We have rephrased the statement.

23. p. 7817, 1st two paragraphs should be joined. 2nd paragraph fragments should be separated by semicolons or periods. Sect. 2.1 First par. Envisat will not continue to orbit at the stated altitude. Towards the end of the mission, the orbit was already degrading.

Reply: We thank the reviewer for pointing out this information. The first two paragraphs in pages 7817 have been joined as recommended. The second paragraph has been separated and the first paragraph of section 2.1 has been modified accordingly.

24. p. 7818: Discussion of limb and nadir observation modes would be more clear if contained in separate paragraphs along with another separate paragraph about how they can be collocated. The last sentence of sect. 2.1 belongs at the first part of this section with the discussion of the satellite status.

Reply: Thank you for mentioning this, we have separated the discussion on SCIA-MACHY limb mode from those on the nadir observation. We have also moved the last sentence of sect. 2. 1 as requested.

25. Sect. 2.2: All instrument abbreviations should be spelled out at their first use. It should be noted that TES discontinued limb mode observations early in the mission.

Reply: We thank the reviewer for this information, we have noted and implemented the suggestion provided.

26. p. 7819, bottom: It should be stated that OMI/TOMS is a retrieval of total ozone.

Reply: The statement has been modified.

27. Sect. 2.2: The vertical range and approximate vertical resolution of MLS should be given. A more complete web link to the OMI/MLS tropospheric ozone should be given. From this link, only Tropospheric ozone from the "cloud-slicing" technique is

mentioned. If it is OMI/MLS from Ziemke et al. 2006, then the reference needs to be made here.

Reply: This is a good point. We have added the following information to section 2.2 of the revised manuscript: The spatial resolution of MLS is 5 km cross-track x 500 km along-track x 3 km vertical depending on the parameter under observation as discussed by Waters et al. 2006. The OMI/MLS tropospheric ozone data product used was described by Ziemke et al. 2006.

28. p. 7820, line 15: carried aloft by

Reply: The statement has been corrected.

29. p. 7821, bottom: period is missing for the last sentence.

Reply: Period has been included as pointed out.

30. p. 7823, line 10: The plurality of the subject and verb do not agree and this sentence is awkward.

Reply: The sentence in line 10 of page 7823 has been modified.

31. Sect. 2.3, 1st sent.: The SCIAMACHY LNM retrievals

Reply: The statement containing "The SCIAMACHY LNM retrievals" has been modified as suggested .

32. p. 7824, lines 3-7, this sentence is confusing and is not correct as stated

Reply: The sentence has been reworded.

33. p. 7826: Discussion of how clouds are handled in the total O3 algorithm is confusing. First, there is a discussion of SACURA, but then it says that clouds are treated as Lambertian without further details. This must be clarified. The discussion on SCODA for the limb mode would more clearly go in a separate paragraph with the 1st sentence of the following paragraph.

Reply: Thanks to the reviewer for mentioning this, we have clarified the information and discussed the following in section 3.3 of the revised manuscript: "Cloud-top-height and cloud fraction from SACURA are used to determine an effective bottom of atmosphere (BOA) height. In case of a clear-sky scene this becomes the surface height. For a fully cloudy scene the cloud-top-height is the BOA. The effective albedo of BOA is determined using the Lambertian Equivalent Reflectivity (LER). This is described in Coldewey-Egbers et al. (2005) ." The discussion on SCODA for limb mode has been moved to a separate paragraph as suggested.

34. p. 7627, lines 3-4: This is not clear. Exactly what kind of sensitivity analysis was conducted and how does the cloud fraction threshold reduce the tropospheric ozone. Is the cloud-shielded ozone not accounted for in the "total" ozone retrieval with the use of a priori information (if so, then it is not really a total ozone retrieval) or is this due to bias in the a priori information?

Reply: Thank you for pointing this out, we have clarified the statement by adding the following information to section 3.3 of the revised manuscript: "The cloud-shielded ozone was accounted for in the total ozone retrieval through the use of an ozone climatology. In cases where pollution increases tropospheric ozone to above the climatological ozone, the retrieved tropospheric ozone may underestimate the true tropospheric ozone below the cloud."

35. p. 7827, line 19: impact of albedo errors; line 20: compared with

Reply: The statements in both lines 19 and 20 have been modified as recommended.

36. p. 7828, line 9: This is unclear, is the error in the monthly mean a bias? The paragraph should be broken at line 9 - the start of discussion of errors in the tropospheric column belongs in a separate paragraph.

Reply: Thank you for this, we have changed the word to reflect variability in section 4 of the revised manuscript. The paragraph is broken as requested. The beginning of

the discussion of errors in tropospheric column has been moved to a new paragraph.

37. p.7829, line 13 and 7830 lines 2 and 23 and 26: plurality of subject and verb do not agree

Reply: We have corrected the sentence.

38. p. 7830, line 23: typo

Reply:The typo has been corrected.

39. p. 7832, Java, not JAVA, line 19: This is not a sentence.

Reply: The word JAVA has been replaced by Java.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 7811, 2013.

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