Replies to Interactive Comments

AMT2013-336

"Daytime stratospheric NO2 retrievals"

Authors' response to Anonymous Referee #1

(1) An important point is the large bias (30-50%) between OMI (KNMI DOMINO v2.0) and SCIAMACHY nadir (KNMI-BIRA TM4NO2A v2.3) retrievals, especially if both data sets are photochemically converted. It is puzzling me because Dirksen et al. (2011) and Hendrick et al. (2012) found a good agreement between OMI, SCIAMACHY nadir and ground-based observations, suggesting the absence of such a large bias between OMI and SCIAMACHY. Can this feature be attributed to the change in the OMI version (DOMINO v0.8 in Dirksen et al. (2011) and DOMINO v2.0 in the present study) ?

We have not performed any direct comparison between different OMI DOMINO versions: there have been changes associated with every version, of course, but none should have affected the slant column significantly. We are inclined to believe that it is the accuracy of the ground-based comparison that is most limited by uncertainty.

Also, you mention the fact that the offset in the new OMI NO2 stratospheric columns is due to wavelength calibration, liquid water, and O4 contributions issues. I think it would be interesting to illustrate this statement by adding some plots in the manuscript or at least give more details on the sensitivity tests performed.

More definite results will follow from an independent group study and published in a separate manuscript. Suffice it to mention that sensitivities to the spectral fit algorithm are currently reported as large as 20-40% (Krotkov, 2014).

(2) You used only TM4 model results obtained after assimilation of the OMI NO2 total columns. Since you don't perform any assimilation in the case of the SD-WACCM model, it would be interesting for the reader to add comparisons with TM4 model results obtained without assimilation, in order to determine/quantify the impact of the latter on the agreement with satellite data and to see the real performances of both models.

TM4 has a poor stratospheric chemistry module, and it is no longer supported – so we do not think it interesting to analyze its performance further. We would defer a

thorough model study to a future paper that features newer and better supported models.

(3) Since aerosols and especially sulphate aerosols can significantly affect the NO2 abundance in the stratosphere, it would be interesting to know how aerosols are initialized/implemented in both models. So, please add a few sentences on this in the description of the models (pages 907-908).

Revised as suggested. The stratospheric aerosol in WACCM is initialized using the SAGE climatology. Additional information regarding heterogeneous processes in WACCM is included in the auxiliary material of [Kinnison, 2007].

(4) The SD-WACCM model is used for comparison with satellite data but also for their photochemical conversion to the HIRDLS LST. Did you consider the risk of circular argument by doing this ?

Would that be the risk of reasoning like: "Since the WACCM photocorrection is correct, then the WACCM (photocorrection) model is correct?" Apparently, the model may be faulty at places but the photocorrection still be correct – that is, correct to first order.

(5) Discrepancies (too low and too large NO2 peaks, especially in the extratropics) are found between models and limb sensors. Is it expected and is it related to the chemistry, transport, or both ? Maybe it would be interesting to have feedback from modellers on these issues.

Good point. The discrepancies between model and observations are mostly a springtime extratropical lower stratospheric phenomenon (see Figure 7b), but we do not know at the moment what the cause may be. One could be suspicious of the representation of heterogeneous processes associated with N2O5, HNO3 and ClONO2 exchanges. Transport could be playing a role too. Obviously, one needs more observational evidence in order to make a statement. Indeed, we are circulating these results and trying to get feedback from modelers.

(6) Page 906: Since you show comparison results up to 60N and 60S, is the photochemical correction also accurate in case of denoxification?

We do not expect any large denoxification outside of polar latitudes, thus only minor photocorrection errors in that respect. A polar study would need a more careful

approach: denoxification, planetary waves, very high zenith solar angles… they could all contribute to photocorrection errors at high latitudes.

(7) Page 910, lines 11-12: SCIAMACHY limb is larger than MIPAS by up to 30% around 30hPa in the tropical stratosphere. Any explanation(s) for this feature ?

Not at this moment – this is probably a question for the respective development teams.

(8) It would be useful to have an overview about the structure of the paper at the end of the Introduction.

Agreed. The following paragraph has been introduced:

"This paper is structured as follows. Section 2 provides a description to each of the datasets that intervene in the intercomparison: the satellite records in Section 2.1, the model-based photochemical correction in Section 2.2, and the model simulations in Section 2.3. Section 3 contains the main intercomparison results, with the gradual inclusion of limb profiles (Section 3.1), model profiles (Section 3.2) and nadir columns (Section 3.3) into the observed collection, followed by some discussion. Finally, Section 4 brings a summary and conclusions. "

(9) In Rodgers theory, A is used for the averaging kernels and K for the weighting functions. So, using K for expressing the averaging kernels as you did on page 903 can be confusing. I suggest to replace K by A.

Agreed.

(10) Table 3: it would be interesting to have the differences also expressed in %.

Agreed.

(11) Other technical corrections…

All agreed.

1/'Dirksen 2011' should be replaced by 'Dirksen et al. (2011)'. The same remark applies for almost all references, so please check the entire manuscript.
2/Page 915, line 8: 'Boersma, 2008' should be replaced by 'Boersma et al. (2008)'.
3/Page 920: (Cook, 2009) is missing in the reference list.

Cook, P. A., Roscoe, H. K., "Variability and trends in stratospheric NO2 in Antarctic summer and implications for stratospheric NOy", ACP, 9, 3601-3612, doi:10.5194/acp-9-3601-2009.

4/Page 923, line 6: the publication years should appear between brackets.
5/Figs. 5, 6, and 7a are still difficult to read for me, especially due to the small fonts for the axis labels.

The graphical files have very high resolution: please, zoom in to see details.

6/Legend of fig. 13, page 947: 'ff' in 'differences' is not correctly written.

This appears to be related to the journal's own LaTEX compilation. We let them know.

Authors' response to Anonymous Referee #2

1) p901 l9: According to Boersma et al., 2007 a third order polynomial is used.

   Please note that this polynomial is updated to order five in Boersma et al., 2011.

2) p904 l11: Please provide a typical value of alpha?

   Agreed. For noiseless observations (i.e. small S), alpha is driven by the difference between observed and model vertical columns over clean areas. If the difference between the OMI and (free-running) TM4 stratospheric columns is 50% then alpha will be 0.5.

3) p904: In Dirksen et al., 2011 it is described that the assimilated stratospheric column is based on the TM4 model field before the observation update. I assume the same approach is used here. Please add a sentence mentioning this. This is relevant information for the argumentation under Point 1 on page 916.

   Agreed. The assimilated stratospheric column is a model forecast of the assimilated model state . A new reference to Dirksen et al, 2011 is inserted in this paragraph (pp 904, line 6).

4) p903 l20: Explain the terms S And V in Eq. 8. Mentioning the term Kalman filter may be in order.

   A mention to the term Kalman filter is inserted (pp 903, line 16). The terms S (observation noise covariance) and V (a priori state covariance) are described in the text. For more details, the reader is referred to Dirksen et al, 2011.

5) p905 l10/11: Is it correct to classify ClONO2 photolysis as slow when its lifetime is in the order of hours?

   Good point. The photolysis of ClONO2 is fast. But the net NOx production that results is low, because ClONO2 "rapidly" reaches an equilibrium with ClO. There is no such equilibrium for N2O5, since its cycling partner (NO3) is virtually destroyed during daytime, and its net conversion into of NOx much more efficient.

6) p902 l4/5 21/22: the fixed temperature for the NO2 x-section was already mentioned on p901.

   Thank you. We believe that this is redundancy with a purpose.

7) p906 l16: Do you use a look-up table or is the photochemical correction recalculated each time it is applied?

   The photochemical correction is stored in a look-up-table.

8) p906 l25: Do you mean to say that the photochemical correction in these areas has large uncertainties due to twilight conditions? If so, please rephrase accordingly.

   Not entirely. The sentence is modified into "correspond to latitude sectors that suffer daytime to nighttime conversions at large solar zenith angles, and should be treated with caution". (pp 906, line 26)

9) p907 l24: Briefly mention how WACCM did perform compared to the other models in the study by Eyring et al., 2010.

   Inserted "As far as stratospheric dynamics, transport and chemistry are concerned, the WACCM model performed overall better than average." (pp 907, line 24)

10) p908 l1-17: Please mention the time steps used for updates in transport and chemistry for TM4, same for the WACCM model in the previous section.

    Agreed. Time step is 30 minutes for both models.

11) p911 l11-12: This sentence is hard to read/understand, please rephrase.

The sentence has been deleted (pp 911, line 11)

12) p911 l21-26: Please mention/discuss the fact that the plots in Fig 7a show that for 30-60S the peak of the TM4 NO2 profile occurs at too low altitude. This could also be mentioned in the summary of the paper.

Thank you. We briefly mention that the TM4 peak NO2 densities occur at too low altitude, but do not delve into details since this model has very poor stratospheric chemistry and is no longer supported. We would postpone a thorough model inquiry to a future paper that features newer and better supported models.

13) p912 l4-7: The concept of equatorial production zone of NOy merits to be discussed in the introduction.

We modify the introduction to mention that NOy is "produced (as NO) by oxidation of N2O to reach a maximum in the tropical mid-stratosphere" (pp 904, line 25).

14) p912 l25/26: this seems a repetition of the first part of the sentence.

Deleted "into the winter hemisphere" (pp 912, line 26).

15) p913 l1: please provide a reference.

The sentence has been deleted (pp 913, line 1). It is a guess inferred from the steady-state solution for the NO:NO2 ratio as in e.g. [Jaegle et al., GRL, 1994], but it is not either relevant nor clarifying.

16) p913 l1-6: This sentence is too long and difficult to read, please rephrase.

The sentence is modified to "Asymmetries in the averaged NH/SH distributions seen in Fig.9a, such as the larger winter abundances in the Southern Hemisphere, should be attributed to first order to the slight asymmetry in the HIRDLS LST-latitude curve (Fig.3). The dependence of the HIRDLS local solar time on latitude renders observations over the southern hemisphere more exposed to nighttime conditions during the winter season." (pp 913, line 1)

Also, changed "affects these latitudes" into "plays a role" (pp 913, line 12)

17) p914 l23: Water vapor has some absorption features in the DOAS fit window, I'm not sure about liquid water.

Liquid water appears to have an absorption signature too. Please see:

Boersma, F., Geffen J., Maasakkers, B, Eskes, H., Williams, J., Veefkind, P, Algorithm improvements for Dutch OMI NO2 retrievals, 18th OMI Science Team Meeting, De Bilt, Netherlands , 11-13 March, 2014.

http://www.knmi.nl/omi/documents/presentations/2014/ostm18/wednesday/Boersma_NO2_algoritm_update_OMISTM18_2014.pptx

18) p915 l7: Please elaborate what you mean by retrieval configurations.

In the Summary and Conclusions, the retrieval configuration refers to "wavelength calibration, absorption cross-section for NO2, spectral fit window width, number of interfering species, spectral resolution, solar reference spectra, ring spectra, etc…"

19) p915 l20: This should be version 1.0.

Corrected.

20) p917 l4: What do you mean with "assimilation top"?

It is the approximate level beyond which the Kalman filter will start rejecting observations for having too large a tropospheric component.

21) On p916 l11 you state "biases in nadir stratospheric column" and in the summary of Point #1 (p917 l5) you say "biases in slant column densities" I assume it should be slant column on both occasions. I find this subsection a bit hard to read.

To my understanding the message you want to convey is that "biases in the observed slant columns lead to biases in the stratospheric NO2 columns without affecting the tropospheric columns, because only the observations over remote/unpolluted areas are used to assimilate the stratospheric NO2 model field".

Thank you. The sentence in pp 917, line 5 is modified as suggested. A certain amount of complexity is required in this section to quantify the tropospheric error made when stratospheric and tropospheric components are separated via assimilation.

Furthermore I am a bit puzzled by the shapes of the averaging kernel shown in Fig 10. Fig 1 of Eskes et al., 2003 shows a much stronger effect of clouds, with the cloudy kernel being almost twice as large as the clear sky kernel at 600 hPa.

That would be correct for a particular scene (low albedo) with a particular cloud top height (e.g. 800 hPa as in Fig 1 of Eskes et al., 2003). Our figure 10 is representative of a global average of cloud conditions on March 21st 2005, i.e. all cloud altitudes and all surface albedos included.

22) p918 l4/5: This statement appears a bit circular as a tropospheric signal over a presumed clean area would be classified as stratospheric, hereby automatically reducing the remaining tropospheric column.

That is correct. The sentence is modified to “A tropospheric source may on average be safely discarded as a cause of stratospheric bias. Exceptions to this rule may occur over areas with significant differences between model and observed tropospheric columns. In this case, a characteristic error signature would arise that is easy to identify.” (pp 918, line 4)

23) p939 (Fig 7a): What is the vertical binsize for the partial columns? I would expect that integration/summation of the partial columns yields the stratospheric column.

That is correct. The vertical binsize for the partial columns is derived from a uniformly spaced log-pressure grid, defined as p(i) = 1000x10^(-i/24) for i=0,120 in pp 909, line 11.

Spelling-related:

All agreed.

- p896 l17: ".. limb observations." add ", respectively"
- p896 l20: long time series -> long-term time series
- p897 l14: long-term studies -> long-term trend studies
- p898 l24: "limb retrieval" add "of NO2"
- p899 l13: "Fourier" add "transform"
- p899 l20: micron -> “mu”-m (applies to p900 as well)
- p900 l10 in -> by
- p901 l26: earth -> Earth
- p903 l6: assimilated gas -> assimilated trace gas
- p906 l16: Equation number is missing.
- p907 l13: in -> by
- p908 l5: degrees -> degrees, respectively
- p909 l6: Equation number is missing.

- p913 l5: renders southern -> renders observations over southern
- p913 l11: long-term time trends -> long-term trends
- p913 l23: a very tight -> close
- p913 l26: we define -> we can define
- p913 l26: tight -> close
- p914 l8: high -> too large
- p914 l10: or 35% -> (35%)
- p915 l8: New paragraph
- p916 l10: remove "should"
- p916 l17: gas -> trace gas
- p916 l19: clear -> clear sky
- p916 l22: of -> the
- p918 l4: to a 5% -> to 5%
- p920 l7 13: SCIA -> SCIAMACHY
- p932 (Table 3): Please add the latitude ranges for SH/Eq/NH in the caption.
- p944 (Fig 10): in clear -> for clear sky. In the middle panel please add a trace indicating unity kernel values.

Authors' response to Nickolay A. Krotkov

(1) The paper should acknowledge those who first presented the OMI strat NO2 discrepancies with both NASA GMI model and concurrent satellite measurements.

The authors offer their excuses for the omission. The following citation is included in the introduction:

"Just as previous work has acknowledged the need to investigate differences in satellite stratospheric NO2 columns (Krotkov, 2012), this work sets out to quantify the extent to which current observation systems are consistent with one another."

(Krotkov, 2012) → Krotkov, N. A., Bucsela, E. J., Celarier, E. A., Lamsal, L. N., and Swartz, W. H.: Improved OMI NO2 Standard Product: Algorithm, evaluation, and results, EOS Aura Science Team Meeting, Pasadena, California, 1-3 October, 2012.

(2) The OMI stratospheric NO2 positive bias has been traced to the current OMI operational NO2 fitting algorithm, which is shared between DOMINO (V2) and standard (SPv2.1) NO2 products. The new NASA NO2 fitting algorithm essentially removes the bias in OMI strat NO2.

A 20-40% decrement in stratospheric columns as reported in (Krotkov, 2014) is certainly consistent with the differences that we observe relative to the limb reference. However, claiming that the new NASA NO2 fitting algorithm removes the bias in OMI stratospheric NO2 should be the subject of a dedicated but separate study. In any case, the following paragraph is moved in Section 3.3 from [pp 914, line 20] to [pp 919, line 28]:

"The apparent offset in OMI stratospheric columns is currently under investigation and has been preliminarily traced to spectral DOAS sensitivities to wavelength calibration, liquid water and O2-O2 contributions (Jos van Geffen, personal communication, 2013)."

And the following sentence is included:

"Independent adjustments to the OMI spectral fitting algorithm report decrements as large as 20-40% in vertical column densities (Krotkov, 2014), which are consistent with the differences that we observe to the limb reference."

(Krotkov, 2014) → Krotkov, N. A., Joiner J., Bhartia P.K., Lamsal L. N., Marchenko S., Celarier, E.A, and Swartz, W. H., Li, C., Key improvements in OMI NO2 and SO2 products, 18th OMI Science Team Meeting, De Bilt, Netherlands , 11-13 March, 2014.

http://www.knmi.nl/omi/documents/presentations/2014/ostm18/wednesday/Krotkov_KeyImprovements_OMI_NO2_SO2_OMISTM18_2014.pdf

(3) Using new NASA fitting OMI NO2 SCDs are reduced by 20%-40%. This is turn causes reduced tropospheric NO2 VCDs over polluted regions. Therefore, the following statement in the abstract is not consistent with our preliminary results: "It is highlighted that biases in nadir stratospheric columns are not expected to affect tropospheric retrievals significantly "

Thank you. We argue that “as long as biases in nadir stratospheric columns are additive, they are not expected to affect the tropospheric estimates significantly”. The additive quality of the stratospheric/SCD errors is key to their not affecting the tropospheric component. Indeed, preliminary studies conducted at KNMI indicate that spectral fit biases are additive (not multiplicative).

In order to emphasize this point, the manuscript is modified:
On [pp 896, line 17]
On [pp 916, line 11]
On [pp 917, line 9]
By inserting “Therefore, as long as biases in nadir stratospheric columns are additive, they are not expected to affect tropospheric estimates significantly, because they are advected over polluted regions by the chemical transport model.”