

Interactive comment on "Effect of volcanic aerosol on stratospheric NO_2 and N_2O_5 from 2002–2014 as measured by Odin-OSIRIS and Envisat-MIPAS" by Cristen Adams et al.

Cristen Adams et al.

cristenlfadams@gmail.com

Received and published: 10 April 2017

Response to Referee 3

We thank you for your comments, which have helped to improve our manuscript. Below we address the recommended changes point-by-point.

General comments:

Since these general comments have been repeated/clarified in the specific comments below and by other reviewers, we have labelled them A, B, C (as listed below), so that the responses could be more easily cross-referenced.

C₁

A. NO_2 anomalies in presence of polar stratospheric clouds, a case for which discussion is avoided in the paper, seems not to reflect the denitrification expected in this case

The reasons for the variations in AOD at the Southern Hemisphere high latitudes are unknown. In order to remove profiles potentially affected by PSCs from the analysis, layers of extinction profiles that were measured at temperatures < 195 K, the temperature for PSC formation, were removed from the analysis. The high monthly mean AODs at these latitudes were not removed with this new filter. Since the reasons for these fluctuations are unknown, we have decided to remove 60°S to 90°S from the analysis. We have adjusted the figures accordingly and have added the following text to the first paragraph of Sect. 3.1

"Latitudes south of 50°S were excluded from the analysis because there is no evidence of volcanic aerosol at these southern hemisphere high latitudes and OSIRIS AODs are dominated by seasonal variation."

Overall, the paper provides interesting observations, but I think that two aspects have not been considered appropriately and might explain the inconsistencies found between MIPAS and OSIRIS measurements.

B. First, the authors consider monthly zonal means, with 10° latitude bins. This is a very coarse grid to study volcanic plume such as the ones considered in this study. The effects the authors want to study are largely diluted in the bin averaging and the mean values obtained from the binning are likely to be biased depending on the coverage of the instrument. Obviously, the effect of the bias is expected to increase if the comparison concerns data from two different instrument (with different coverage). Another similar source of bias could be the use of climatological data for ozone, neglecting the effects of local chemistry

The 10° monthly latitude bins were selected for two reasons:

- 1. Bins of this size are often used in studies of long-term variations and trends. Therefore, the results of this work is more directly applicable when understanding, e.g., long-term trends in NO_2 timeseries.
- 2. This allowed a large amount of data to be averaged in most bins (typically ~100 measurements for both OSIRIS and MIPAS), which helped to decrease noise and smear out short-term natural variation. This is particularly useful when fitting contributions from QBO and estimating baseline NO₂, which varies seasonally and with latitude.

In order to test the effect of the bin sizes on our ability to assess processes around volcanic aerosol, we recalculated OSIRIS AODs in 5° latitude bins at a sampling of four times per month (approximately weekly). Based on the OSIRIS sampling, these are the smallest bins that would yield reasonable results. The observed AODs are shown in Figure 1 for latitude bins centered around $60^\circ N$, a latitude at which there were several episodes of enhanced volcanic aerosol. The maximum AODs for both bin sizes are very similar, suggesting that the smaller bin sizes do not give additional information on the volcanic events. Similar results were observed at other latitudes. The maximum observed AOD for all latitude/time bins was 0.0086 for the 10° monthly bins and was 0.0088 for the 5° , $\frac{1}{4}$ month latitude bins.

The 10° monthly bins are suitable for this analysis because the lifetime and transport of sulfate is slow compared with SO_2 and ash. Furthermore, individual OSIRIS profiles with very large extinctions (>2x10⁻³ km⁻¹), which could be associated with ash or very fresh plumes (~1-2 weeks) were removed from this analysis, which allowed for the direct focus on sulphate/NO₂ chemistry.

An example of the MIPAS sampling relative to OSIRIS AOD sampling is given in Figure 2 and Figure 3 for the 50°N latitude bin. This latitude was chosen to explore sampling because this latitude had valid MIPAS measurements for larger OSIRIS AODs (see Fig. 4a of the paper). Both OSIRIS AODs and MIPAS NO₂ measurements are sampled fairly consistently in latitude and time, with no obvious biases that would skew

СЗ

the relationship between MIPAS NO_2 anomalies and OSIRIS partial column AODs, nor that would lead to large biases in ozone between the MIPAS and OSIRIS sampling. Furthermore, comprehensive tests on MIPAS and OSIRIS sampling have been performed for ozone by Toohey et al. (J. Geophys. Res., 118 (11), pp847–11,862, doi:10.1002/jgrd.50874, 2013) and found that MIPAS and OSIRIS have sampling biases of < 2% and < 5% for most latitudes/altitudes.

We have added the following text to the paper to address these comments:

- Sect. 3.1, second paragraph "Bins were tested for smaller latitude and time ranges, but yielded similar ranges of AODs, suggesting that smaller bin sizes did not capture more detailed processes in the volcanic plume."
- Sect. 4.1, fifth paragraph "These discrepancies could not be attributed to differences in sampling between OSIRIS and MIPAS, since MIPAS and OSIRIS both sample throughout the monthly 10° latitude bins. MIPAS measurements are not clustered in parts of the bin where smaller OSIRIS AODs were observed."
- C. The second weakness of the paper, in my opinion, concerns the discussion of the comparison between OSIRIS and MIPAS data, where arguments based on the limited degrees of freedom provided by MIPAS are used to justify the disagreement between results obtained from both instruments. Before drawing definitive conclusions from this hypothesis, the authors should check their interpretation by degrading both datasets to similar resolutions. These issues should be addressed before publication of the manuscript.

In order to test the effect of the MIPAS resolution on the data, representative MIPAS NO $_2$ averaging kernels, A, were considered for the 50°N and 0° latitude bins and applied to the OSIRIS NO $_2$ profiles. The representative profiles had averaging kernel diagonal elements (AKD) closest to the mean averaging kernel diagonal elements (\overline{AKD}) within the vertical range of interest that the VCDs were calculated over. E.g., the representative MIPAS averaging kernels minimized

$$\sum_{j} \left(AKD(z_{j}) - \overline{AKD}(z_{j}) \right)^{2},$$

where z_j are altitude layers in the 5-km altitude range used to calculate the VCDs. The representative averaging kernels, in Figure 4, show reduced resolution over the altitude ranges used to calculate the partial columns in this paper, indicated by the dashed lines.

Since MIPAS NO_2 retrievals are logarithmic, smoothed OSIRIS NO_2 profiles, x_s , could be calculated using from original OSIRIS profiles, x, using

$$x_s = e^{A \cdot ln(x) + (1-A) \cdot ln(x_a)},$$

where x_a is the MIPAS a priori. It then follows that the OSIRIS percent difference profiles, PD, can be interpolated to the MIPAS grid smoothed using

$$PD_s = e^{A \cdot ln(PD+1)} - 1$$
.

Figure 5 and Figure 6 show the unsmoothed and smoothed OSIRIS profiles for 0° latitude and $50^\circ N$ latitude, respectively. The dashed lines indicate the altitudes over which the partial columns were calculated. The unsmoothed OSIRIS profiles are what is shown in Fig 6 and Fig 7 of the paper respectively, but are presented here as a line plot so that the values of the NO $_2$ percent differences can be compared more closely. For both latitude ranges, the magnitude of negative NO $_2$ percent differences is reduced after applying the MIPAS averaging kernels. For example, at $50^\circ N$, NO $_2$ percent differences of approximately -45% to -50% are smoothed to approximately -30% after the MIPAS averaging kernel is applied. Therefore these tests demonstrate that the resolution of MIPAS decreases the observed NO $_2$ anomalies under the presence of volcanic aerosol.

These tests could not be applied quantitatively to the entire analysis because the MI-PAS averaging kernels vary for each profile and are correlated with the amount of NO₂ present. Individual MIPAS averaging kernels could not be applied directly to the OSIRIS data because MIPAS averaging kernels are only available for the measure-

C5

ment times and locations of MIPAS, which are not coincident with OSIRIS measurement times and locations. This is why representative averaging kernels were used to assess this problem. The mean DOFS of the MIPAS VCDs tended to be lower during periods with enhanced OSIRIS partial column AODs, described in the text of the paper and shown in Figure 7. This decreased resolution would further smooth out the MIPAS measured NO_2 anomalies.

The following text has been added to Sect. 4.1, 5th paragraph:

"In order to test this, representative MIPAS averaging kernels were applied to the OSIRIS NO_2 percent difference profiles at 50°N and 0° latitudes. Representative averaging kernels were used because MIPAS NO_2 is retrieved in the logarithmic domain and the averaging kernel thus refers to the logarithm of the mixing ratio. By applying the averaging kernel directly to the percent difference profile, the MIPAS a priori profile does not need to be included in the calculations. The magnitude of the largest percent differences in the NO_2 percent difference profiles decreased from approximately -45% in the original OSIRIS profiles to approximately -30% in the smoothed OSIRIS profiles, demonstrating this damping effect. These tests did not account for variation of MIPAS DOFS with partial column AOD. The smaller DOFS observed for larger OSIRIS partial column AOD would lead to further damping of the MIPAS NO_2 percent differences."

Specific comments:

Abstract

L. 23: The authors mention percent difference of up to ~25%. They should mention with respect to what (to OSIRIS? To guiescent periods?)

We have clarified this by adding "relative to baseline levels" to the text.

1. Introduction

L. 8, p.1: The authors should define NOx like they defined NOy. Possibly at the same place to lighten the text, see technical comment on L. 4.

A definition of NOx has been added

L. 16, p.2: For the sake of clarity: "NOx/NOy decreased with increasing aerosol surface area".

This has been edited as recommended

2. Satellite and model datasets

2.3 Photochemical modelling

All three reviewers had comments regarding the inputs to the photochemical model. In order to address these comments, we have changed the input ozone climatology and have revised our model errorbars using a series of perturbation tests. These tests are described in Sect. 2.3 and summarized in Table 1, and included in Fig. 5 of the revised paper. The break-down of the major contributions to the uncertainty estimates is given in Figure 8. These tests are described in Sect. 2.3 and summarized in Table 1, and included in Fig. 5 of the revised paper. The break-down of the major contributions to the uncertainty estimates is given in Figure 8. Aerosol (black) are for the two error terms related to extinction to surface area conversions. Ozone (blue), temperature (red), albedo (cyan), and NOy (green) refer are for year-to-year variability in these inputs. Systematic errors in ozone, temperature, albedo, NOy, NO $_2$ cross-section, and NO-O $_3$ reaction rates are mostly 1-2

L. 16, p.4: Which type of climatological data are used for ozone and temperature? And why don't they use MIPAS ozone data which would be fully consistent with the used data for SO_2 , N_2O_5 , HNO_3 and NO_2 ? Perliski et al. (1989), cited in Randeniya et al. (op. cit., 1997), indicate that the ozone behaviour at high latitudes is expected to be dominated by local chemistry during summer, which is the time and region of interest for the present study, and where Eq. (2) is expected to have the greatest impact on ozone. Hence, using climatological data for the ozone field in this specific study focussing on the effect of volcanic eruptions might bias the results of the analysis.

C7

We initially used a standard O_3 climatology (from McPeters et al., JGR, 1999). However, we have since switched to an OSIRIS zonal, monthly-mean climatology for the revised version. (From previous experience we knew OSIRIS agreed very well with other ozone climatologies and so we simply used our model default inputs.) For temperature we use a climatology from Nagatani and Rosenfield (1993). OSIRIS does not measure temperature. We could have used MIPAS temperatures, and it has a small (2 K) bias in the lower stratosphere (Stiller et al., Atmos. Meas. Tech., 5, 289–320, 2012), but as demonstrated in the sensitivity study, systematic errors in temperature, make very little difference. The uncertainties associated with interannual variability in ozone were estimated and included in the errorbars in Fig. 5 of the revised paper. The contributions to these error estimates from ozone are shown in Figure 8, and are ~3-10% for most of the data points, except in the tropics (see Figure 8 of this reviewer response).

Nagatani, R. M., and J. E. Rosenfield (1993), Temperature, net heating and circulation, in The Atmospheric Effects of Stratospheric Aircraft: Report of the 1992 Models and Measurements Workshop, NASA Ref. Publ. 1292, edited by M. J. Prather and E. E. Remsberg, pp. A1–A47, NASA, Washington, D. C.

L. 18, p.4: I don't understand what the authors mean with "but fixed to a specified Julian day". Please clarify.

The model is integrated forward in time but instead of moving from JD to JD+1, where there would be differences in solar illumination, it again repeats JD. It continues to loop over this same day, up to 30 times, until the model converges. That is, concentrations at the end of the day match the beginning ("in a 24-hour steady-state").

The text has been modified to read "All remaining species are calculated to be in a 24-hour steady-state by integrating the model for as many as 30 days, but where the model remains fixed on the original, specified Julian day".

L. 20, p.4: Since Thomason's climatology covers the whole period 1979-1995, it would

be useful to explain in a few words which data are used in the present study (specific year and/or region?). If they only use the approximated expressions (5) of Thomason's work as discussed in the following of the section, the authors could already announce that here (e.g.: "(...) using the aerosol surface area climatology of Thomason et al (1997) as explained later, (...)"). Actually, I don't know why the authors want to consider heterogeneous chemistry on background stratospheric aerosols, since they aim at studying typically volcanic situations. Besides, they mention further that they match the extinction coefficient with OSIRIS values, which should normally reflect this volcanic feature. All these points should be clarified.

The sentence in question is an artifact of a much earlier draft of this paper that was not properly adjusted to reflect the methodology used herein. We did not use the Thomason et al. climatology at all, as the reviewer questions. Further, the use of the word "background" is also not correct in this context, also caught by the reviewer. The sentence now reads "Heterogeneous chemistry on stratospheric sulfate aerosols is included, but polar stratospheric clouds are not included."

L. 24, p.4 to L. 10, p.5: The estimation of the SA in function of the extinction seems particularly crude, with a succession of approximations with choices which are not always clear nor convincing. The authors use the fact that in the case of Kasatochi, the mean particle size (see also next comment) decreases and that in the case of Sarychev, it increases, to choose a dependence between SA and the extinction which reflect none of these cases. This is a questionable way to approach this investigation focussing on a selection of recent volcanic eruptions (including those two ones). It is also worth to mention that Thomason et al (1997, op. cit.) uses such linear expression to describe cases where the extinction is higher than 2. 10^{-2} km⁻¹, which is a really high value rarely encountered in the period considered here. The wavelength used to characterize the extinction in the equation in L. 26, p.4 is not mentioned, making it meaningless and preventing to compare this value with Thomason's coefficient (equal to 2000 to characterize the extinction at 1020 nm). They also "test" a non-linear SA

C9

dependence mentioning Thomason's work, but their choices are quite different form what Thomason proposes. The choice of p=0.7 is quite similar to Thomason's one in the case of the weakest extinction values, but the choice p=1.3 is much higher than the value of 1 used with the highest extinction values. The final scaling "to account for potential errors" gets rid for good of any reference to real cases, making the reader definitively lost in the accumulation of assumptions.

While it is true that in the case of Kasatochi and Sarychev one can attempt to model them in more detail based on the size distribution findings from the cases studies referenced (Sioris et al. and O'Neill et al.), this additional level of information is not available for most of the eruptions considered in this paper. But even this would be limited since the size distributions constantly evolved following the eruption (e.g., Kasatochi shifted the distribution towards smaller particles, and then later on, towards larger particles). Our goal was to use a consistent approach for all eruptions, and hence our "middle of the road" decision to use a linear $k{\rightarrow} SA$ relationship. While some of our choices for the sensitivity study are debatable, we opted to err on the conservative side (e.g., to consider a p=0.7 value which is more extreme than in Thomason et al.). This reflects the fact that not all values in the literature are directly applicable (as pointed out above) to our case, and that even these larger perturbations did not have a serious impact our modeled change-in-NO $_2$ – AOD relationships.

L. 5, p.5: Sioris et al. (2010) don't claim that the stratospheric particle size decreases in the case of Kasatochi. They consider statistical values (through median radius and Angström exponent) which show a decrease of the averaged particle size. This is not the same thing. A particle size decrease supposes the occurrence of some evaporation process, while what happens here is the addition of a significant amount of very thin particles. The authors should change their formulation to avoid the confusion.

We agree with the reviewer, and were simply sloppy with our wording here. We have adjusted this to read "For example, two months after the Kasatochi eruption, there was a shift in the ambient size distribution toward smaller particles (Sioris et al., 2010)

whereas Sarychev led to a shift towards larger particles (O'Neill et al., 2012)".

L. 1, p. 5: The reference by Hansen and Travis, 1974 is a very interesting general reference on light scattering, but I don't see anything in this reference able to clarify the choices and formulation of the equation in L. 26, p.4. Hence, I am not sure it is really useful here.

We assume you are referring to equation (3), on the bottom of page 4. The Hansen and Travis reference does not have this equation literally, although all the 'pieces' are there and it would have been left up to the reader to pull our equation out of this. In light of this, we explicitly derive our equation (3) in a short Appendix that was added to the manuscript and we reference it instead of Hansen and Travis.

3. Calculation of monthly averages, anomalies, and baseline levels

3.1 OSIRIS and MIPAS

L. 13-20, p. 5: The use of monthly zonal means over latitude intervals as large as 10° might really bias the data and limit the quality of the correlative study of quantities derived from two different experiments such as OSIRIS and MIPAS in the present case. If as few as 5 measurements are possible for one bin, it could be possible, for instance, that one instrument covers only regions with low aerosol background while the other one catches the plume of an eruption. Or that one instrument covers a large region of the bin and returns average values of a quantity while the other one only catches some very local spot with specific (low or high) volcanic load. Did the authors prevent this kind of situation in some way?

See response to General Comment B at the beginning of this reviewer response.

L. 1 p. 7: Concerning the NO₂ response to the QBO, do the authors mean the fitted response as illustrated by the cyan curve in Figure 1b?

Yes, we mean this and have replaced "the NO₂ response to the QBO" with "the fitted response to the QBO, as illustrated by the red dashed line in Fig. 1b" in the text to

C1:

clarify this.

L. 11-15, p. 7: same remark as for L. 13-20, p. 5: I suppose that the model covers the whole 10° latitude monthly bin, while corresponding measurements might cover a reduced region of the bin, introducing potentially a bias in the analysis.

The sampling of the OSIRIS measurements is fairly uniform across the bins, as demonstrated in the response to General Comment B, shown above in this document. Furthermore, the model data is calculated for a range of partial column AODs and then interpolated to the mean partial column AOD, as measured by OSIRIS. Therefore, the modelled NO_2 anomalies are for the mean partial column AODs as sampled by OSIRIS.

- 3.3 Conversions between partial column AODs, aerosol extinction, and extinction at various wavelengths
- L. 21-24, p. 7 (or L. 15-18, p. 5): I guess that profiles for which valid data don't cover the whole interval 3-7 km above the tropopause are rejected. This could be mentioned for the sake of completeness.

We have clarified this in Sect. 3.1 with the following: "If a profile does not have valid data over all five measurement layers, it is not included in the analysis."

4. Results

4.1 NO₂, N_2O_5 , and HNO₃ VCDs

L. 26, p.8 and Figure 3: Overall, there is indeed a very clear correlation between the AOD enhancement and the NO₂ anomalies found by OSIRIS in the Northern latitudes. On the contrary, in the Southern polar latitudes, most of the time, no significant NO₂ decrease is found by OSIRIS and even more, strong local enhancements are observed each year. In the case of the Sounthern polar latitudes, the high AOD is most probably due to PSC, for which one would expect a denitrification, thus a decrease in NO₂. MIPAS NO₂ seems rather to behave in the opposite way, although the very limited coverage of the "volcanic regions" defined by the cyan curves would impose to be

cautious in any conclusion.

See response to General Comment A, above.

L. 2, p.9: I don't understand the reason given by the authors why the AOD should be excluded from the correlation before of its large variability. Large volcanic eruptions are able to produce an even larger variability.

This text was not written very clearly. Data were not removed due to large variability in AODs, but because the large variability in AOD was not related to stratospheric aerosol. We have removed this statement from the text, as have removed data for 60°S to 80°S throughout the analysis, as described in the response to General Comment A, above.

L. 6-7, p.9: Again, the authors consider monthly zonal bins with 10° latitude intervals for their analysis. This might be the reason for the apparently inconsistant behaviour (MIPAS vs OSIRIS, Northern latitudes vs. Southern latitudes) shown in Figure 3 (see comment on L.26, p.8 and Figure 3). Although the use of monthly zonal means is widely used in the community, as mentioned above, such large intervals are not well suited to study atmospheric processes at the level of a volcanic plume for such kind of eruptions, because the spatial extend and the temporal duration of the volcanic perturbation is relatively limited with respect to the spread of the bins. Hence, biases are potentially important and make any conclusion uncertain, especially if different instruments with different coverage and data rates are compared. The authors should remake their analysis by considering much shorter time and latitude intervals to see if the inconsistencies persist.

See response to General Comment B, above.

L. 9-15, p.9: The validity of the explanation given by the authors could be easily checked by degrading the OSIRIS using the MIPAS' averaging kernels (and possibly vice-versa). This way would allow comparing like with like. Did the authors make such check?

C13

See response to General Comment C, above.

L. 29, p.9-L. 1, p. 10: The concept "somewhat linear" is strange. Overall, the interpretation of the shape of the curve is highly subjective, and the superposition of the modelled values on the plots biases our perception. As an example, the OSIRIS plots at 20° N show a behaviour which is neither linear at low AOD, nor saturated at the highest AOD values. The authors should remove these dubious interpretations.

We have deleted the following "For smaller AODs ($< \sim 3x10^3$), this relationship appears to be somewhat linear. For larger AODs, however, this relationship displays greater curvature with additional aerosol having an increasingly smaller impact, which is consistent with the saturation at higher aerosol levels (e.g., Fahey et al., 1993)."

L. 4-5, p.10: In the same way, the agreement in shape and in quantity between modelled and observed data is very relative, and in some case, quite bad. Hence, the authors should qualify their affirmation.

We have replaced this sentence "The modelled data agrees well quantitatively with the OSIRIS measurements across latitudes and AOD ranges and reproduces the shape of the curve approaching saturation." with the following text:

"The modelled data agree well with the OSIRIS measurements and are within the estimated model errors for most OSIRIS data points."

We have also changed the abstract and conclusion to reflect this.

4.2 OSIRIS NO2 profiles

L. 25, p.10: The authors should remove the sentence "At this latitude, decreases in NO_2 are observed between ~10-20 km". This sentence is confusing, since in some cases (e.g. in 2002), an increase is observed in NO_2 instead of a decrease, and anyway, the next sentence expresses appropriately and in more detail what the authors mean.

This has been deleted

5. Conclusions

L. 20-21, P. 11: I think the conclusion concerning the influence of the DOFS on the disagreement between MIPAS and OSIRIS is premature, and should be verified as proposed above, before it is claimed.

This has now been confirmed – see response to General Comment C.

L. 22, p. 11: This line, with the qualification of "somewhat linear", should definitely be removed. The characterization of this relationship using the correlation coefficient is more than sufficient. In the same way, the expression "perfect linearity" on the next line should also be removed. As long as observations are concerned, there cannot be any perfect linearity.

We have replaced "relationships... are somewhat linear" with "relationships... are observed".

We have removed the reference to "perfect linearity" and have replaced this with the following: "Heterogeneous chemistry becomes saturated toward larger aerosol concentrations (e.g., Fahey et al., 1993) and can vary throughout the timeseries with other factors, such as temperature and available sunlight (e.g., Coffey, 1996), all of which can affect the linearity of the correlation."

L. 27-28, p.11: The last sentence should be qualified according to the comment in L. 4-5, p.10.

We have change this to "The modelled data agree well with the OSIRIS measurements and are within the estimated model errors for most OSIRIS data points."

Technical corrections:

L.19, p.1: The authors could consider using "relationship" in the singular, or more precise, the word "correlation"?

This has been changed

C15

L.23, p1: "periods affected by volcanic aerosol"

This has been corrected

L. 4, p.2: reformat the parenthese after $BrONO_2$ (no subscript). Writing "NOy species (where $NOy = NOx + HNO_3 + etc. + BrONO_2$, and NOx = etc.; e.g. Coffey 1996)" might be more fluent. See also specific comment on L. 9.

This has been changed as recommended

L. 13, p.4: I suggest to write 10:00 LT to be consistent with the previous mention of time, and for the sake of clarity.

This has been changed

L. 6, p.5: "to keep the scattering efficiency constant".

This has been corrected

L. 15, p.8: It seems there is a problem of cross reference for Table 2.

This has been corrected

L. 13, p.10: "time [blanco] series"

"timeseries" has been replaced with "time series" throughout

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-242, 2016.

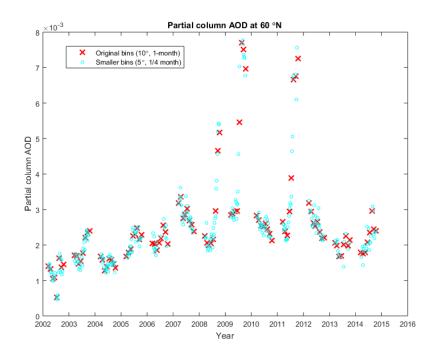


Fig. 1. OSIRIS partial column AOD at 60°N for the 10° monthly bins used in this paper and for smaller bins of 5° and $\frac{1}{4}$ of a month.

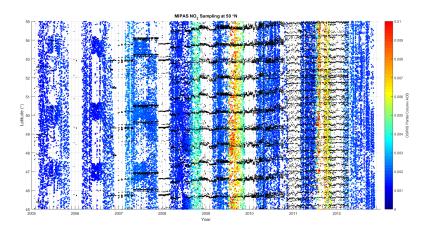


Fig. 2. OSIRIS partial column AOD (color-scale) and MIPAS NO2 measurement locations (black dots) versus time and latitude for data included in the $50^\circ N$ latitude bin.

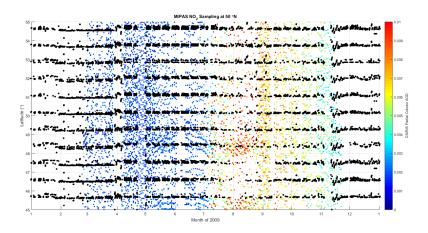


Fig. 3. As for Figure 2, for 2009 only.

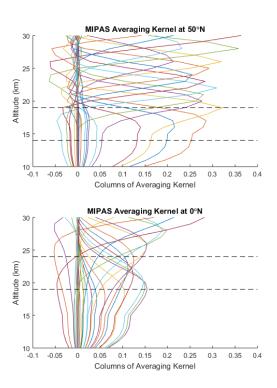


Fig. 4. Columns of representative MIPAS averaging kernels at (top) $50^{\circ}N$ and (bottom) 0° latitude.

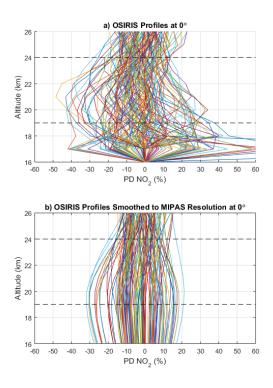


Fig. 5. Monthly mean NO2 percent difference profiles measured by OSIRIS (a) at original resolution and (b) smoothed using the OSIRIS averaging kernel at 0° latitude.

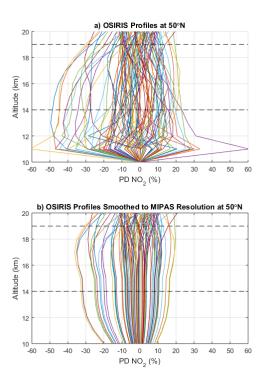


Fig. 6. As for Figure 5, for 50° N.

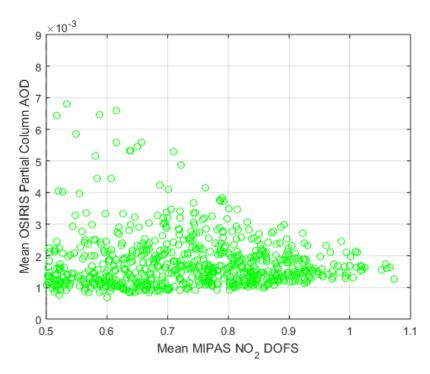


Fig. 7. For all latitude bins and dates included in the analysis, the mean MIPAS DOFS for NO2 versus the OSIRIS partial column AOD.

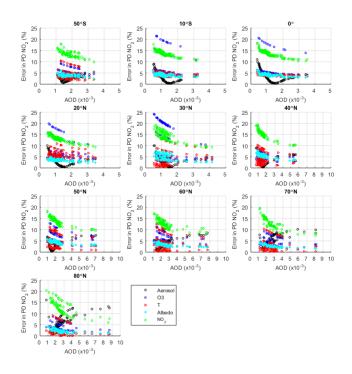


Fig. 8. Breakdown of parameters used to estimated total model uncertainty in Fig. 5 of the paper.