

Referee report

1. Effect of volcanic aerosol on stratospheric NO₃ and N₂O₅ from 2002-2014 as measured by Odin-OSIRIS and Envisat-MIPAS

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General comments:

This work investigates the relationship between aerosol load enhancements after a volcanic eruption and the anomalies observed in the NO₂ vertical partial column over the 3-7 altitude range, as observed by OSIRIS and by MIPAS. The study reveals quite robust correlations between the aerosol optical depth and the NO₂ anomalies, especially when comparing between OSIRIS aerosol and NO₂ features, but the results are much less convincing while comparing with MIPAS NO₂ anomalies. NO₂ 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.

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.

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 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.

Specific comments:

Abstract

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

1. Introduction

- L. 8, p.1: The authors should define NO_x like they defined NO_y . Possibly at the same place to lighten the text, see technical comment on L. 4.
- L. 16, p.2: For the sake of clarity: “ Nox/NO_y decreased with increasing aerosol surface area”.

2. Satellite and model datasets

2.3 Photochemical modelling

- 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.
- L. 18, p.4: I don't understand what the authors mean with “but fixed to a specified Julian day”. Please clarify.
- 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.
- 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 $2 \cdot 10^{-2} \text{ km}^{-1}$, 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 dependence mentioning Thomason's work, but their choices are quite different from 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.

- 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.
- 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.

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?
- 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?
- 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.

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.

4. Results

4.1 NO₂, N₂O₅, 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 Southern 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.
- 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.
- 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 inconsistent 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 extent 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.
- 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?
- 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.

- 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.

4.2 OSIRIS NO₂ profiles

- L. 25, p.10: The authors should remove the sentence “At this latitude, decreases in NO₂ are observed between ~10-20 km”. This sentence is confusing, since in some cases (e.g. in 2002), an increase is observed in NO₂ instead of a decrease, and anyway, the next sentence expresses appropriately and in more detail what the authors mean.

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

Technical corrections:

- L.19, p.1: The authors could consider using “relationship” in the singular, or more precise, the word “correlation”?
- L.23, p1: “periods affected by volcanic aerosol”
- L. 4, p.2: reformat the parentheses after BrONO₂ (no subscript). Writing “NO_y species (where NO_y = NO_x +HNO₃+etc.+BrONO₂, and NO_x=etc.; e.g. Coffey 1996)” might be more fluent. See also specific comment on L. 9.
- 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.
- L. 6, p.5: “to keep the scattering efficiency constant”.
- L. 15, p.8: It seems there is a problem of cross reference for Table 2.
- L. 13, p.10: “time [blanco] series”