July 1, 2014

Characterisation of a stratospheric sulphate plume from the Nabro volcano using a combination of passive satellite measurements in limb and nadir geometry
by M. Penning de Vries et al.

Authors' reply to the interactive comment by Simon Carn

We thank Simon Carn for his extensive review of our paper. We regret that he finds some of the analysis "hard to follow" and we hope our replies to his comments and the changes to the manuscript will improve the readability and clarity. Simon Carn has pointed out two important issues that we would like to resolve here and clarify in the revised version of our manuscript: (1) the ambiguity regarding aerosol composition and (2) the different OMI UVAI data sets and their respective viewing angle dependence.
For our replies, we use the following colour-coding:
Green: Referee comment.
Black: Authors' reply.
Red: Modified text in manuscript.

(1) Aerosol composition

Referee comment: My main criticism of the paper is that the precise composition of the aerosol in the volcanic plume remains ambiguous – the authors appear unable to distinguish between sulphate and ice using the available data. Of course, I would expect both species to be present but the challenge is identifying the dominant component. The fact that the aerosols were at high altitude and strongly scattering (high SSA) seems robust, but either ice or sulphate (or a combination) could produce a negative UVAI. This is clearly apparent in Fig. 1, where the area of meteorological cloud north of the volcanic plume exhibits strongly negative UVAI. The authors acknowledge this ambiguity and even avoid mentioning sulphate explicitly in the conclusions. Given this uncertainty, referring to a 'stratospheric sulphate plume' in the title and 'sulphate aerosols' in the abstract seems a little disingenuous; 'stratospheric aerosol plume' would be more appropriate. Overall I would like the authors to be more candid regarding what can be robustly concluded from their data.

Authors' reply: Using the UVAI, we can only distinguish between absorbing aerosol types (i.e., ash) on the one hand, and non-absorbing types (sulphate or ice) on the other. In principle, the limb retrieval provides information on aerosol size — with which small sulphate droplets could be distinguished from larger ice particles — but since the Ångström exponent and asymmetry parameter are both a priori constraints to the retrieval, this information cannot be regarded as independent. Therefore, we revert to measurements by other sensors to determine the nature of the aerosols in the volcanic plume.

Although, as noted in minor comment 8 below, no information on aerosol type from the ground-based lidar measurements at Sede Boker is given in Sawamura et al. (2012), we have access to measurements of particle size distributions from the co-located AERONET station. These show that as the densest part of the volcanic plume passed over the station, more than 75% of measured optical thickness could be attributed to fine particles; the optical thickness caused by coarse-mode particles can quite confidently be attributed to mineral dust (based on data from preceding days). During the overpass of the plume a distinct peak is seen to grow in the size distribution at about 0.1 micrometers, which clearly indicates a dominance of sulphate particles. CALIOP lidar data, available from June 15 onwards, also point to a plume dominated by sulphate aerosols: Clarisse and co-workers (2014) used low depolarization ratios, indicative of sulphate aerosols, to locate the volcanic plume in CALIOP level 1 data. Fairlie et al. (2013) say, about the same measurements: "These features are characterized by low depolarization (not shown), characteristic of sulfate aerosol". And in Vernier et al. (2013) it is noted that "Given the low VDR [volume depolarization ratio], compared with pure volcanic ash (0.36), it is very likely that the plume is a mixture of ash and recently formed sulfuric acid droplets." In (Theys et al., 2013) it is mentioned that "there is no evidence that this plume was ash laden", which is confirmed by our UVAI data. Apart from these findings, supporting the presence of sulphate aerosols in the volcanic plume, MODIS IR data, which clearly show the presence of an ice cloud on June 13, do not show any indication of ice in the Nabro

plume on June 14. This was already mentioned in Sect. 4.1 of the original manuscript (starting on line 24, p. 7749).

It thus seems very likely that the plume mainly contained sulphate aerosols — although we cannot completely exclude the presence of ice. We have added a statement on the nature of the detected aerosols to pages 7749-7750, as follows, but opt to keep the title as it is. The section was altered in compliance with referee comment 5 by an anonymous referee, and now reads:

Most of the particle extinction observed in the MODIS images from 13 June (Fig. 1G and H) is probably due to the presence of ice particles, visible in MODIS IR cloud data (Fromm et al., 2013) and in SEVIRI data (Vernier et al., 2013). On the following day, the plume can no longer be seen in MODIS true-colour images (Fig. 2G and H) and neither is a cloud detected by the IR retrieval (not shown here, but see: http://ladsweb.nascom.nasa.gov/browse_images/granules.html). This does not exclude the possibility that ice was present in the volcanic plume after June 13, but the low depolarization ratio detected by CALIOP is evidence that it consisted mainly of sulphate aerosols (Vernier et al., 2013; Fairlie et al., 2013, Clarisse et al., 2014). In addition, the overpass of the volcanic plume over the joint MPLNET/AERONET (MicroPulse Lidar NETwork (Welton et al., 2001) and AErosol RObotic NETwork (Holben et al., 1998)) site Sede Boker corresponded to a large increase in the number of fine particles (of which the size distribution peaked at approximately 0.1 μm, see: http://aeronet.gsfc.nasa.gov), also clearly indicating that the plume was made up of sulphate aerosols; the optical depth assigned to coarse-mode particles is very similar to that measured on the days before the eruption and can confidently be attributed to mineral dust. Based on the evidence presented above, we conclude that ice contributed substantially to the negative UVAI signal detected on June 13, but that the UVAI signal on June 14 was caused by sulphate aerosols.

To emphasize our conviction that the plume we observed from June 14 onwards mainly consisted of sulphate aerosols, we inserted "sulphate" into the sentence in the conclusions (Sect. 5.3) on p.7758, line 25.

(2) OMI UVAI data sets

Referee comment: Regarding the negative UVAI, this is a fairly common feature of fresh volcanic plumes that are not dominated by ash. In the latter case aerosol absorption dominates and a positive UVAI results. Exploring the origin of negative UVAI further is certainly worthwhile. However, it is important that the authors explicitly state which OMI UVAI dataset they are using, since there are (confusingly) several different UVAI data products available. Also, for very high SO_2 loadings the SO_2 absorption at longer UV wavelengths can also affect the UVAI. I have looked at OMI UVAI data from both the operational OMI OMAERUV product (which uses wavelengths of 354 and 388 nm) and the UVAI from the operational OMI SO_2 and O_3 products (which uses wavelengths of 331 and 360 nm to be consistent with the old TOMS UVAI), and the appearance of the Nabro volcanic plume is not the same in both datasets. Unlike the OMAERUV UVAI (which I presume the authors used in the paper), the 'TOMS-like' UVAI does not show the positive values in the western part of the aerosol plume on June 14. Hence I suggest that the authors conduct more radiative transfer calculations to assess the wavelength dependence of the UVAI effects perhaps this would provide more information on the nature of the aerosol.

Authors' reply: We are a little surprised by the statement that "negative UVAI [constitute] a fairly common feature of fresh volcanic plumes", as we are aware of only one appropriate literature reference (i.e., Rose et al., 2003). We did, however, mention other cases of volcanic plumes with UVAI< 0 in Sect. 5.1: Soufrière Hills and Nyamuragira in 2006, Okmok and Kasatochi in 2008, and Sarychev Peak in 2009; but these are our own unpublished findings. In the TOMS archive (http://so2.gsfc.nasa.gov/toms_1979_2005.html) we found other sightings of UVAI< 0, possibly caused by sulphate aerosols during eruptions of Pinatubo (1991), Marchena (1991) and Nyamuragira (1994, 1996, 1998, 2001). We added this information to Sect. 5.1 in the revised manuscript.

The referee is right in pointing out that there are different UVAI products for OMI, but the OMAERUV UVAI (which we used) is calculated from 340 and 380 nm reflectances; OMAERO, KNMI's product, uses 354 and 388 nm reflectances, whereas OMTO3 uses the reflectances at 331 and 360 nm and is altogether determined in a slightly different way (e.g., the reflectance at the shorter wavelength is used as a reference, whereas in the "classical" UVAI approach, employed in all other algorithms, the reference is the reflectance at the longer wavelength; also, a "modified Lambert-equivalent reflector" model is used in the OMTO3 UVAI calculation — more details can be found in Penning de Vries et al., ACP 2011). Although OMTO3 UVAI is more sensitive to aerosols, we chose OMAERUV UVAI for this study for the sake of comparability with UVAI from SCIAMACHY and GOME-2, which are both only determined at (approximately) 340 and 380 nm. In addition, we, like the

referee, noticed that for very high SO_2 loadings (like those detected in the Nabro plumes on June 13 and 14) the OMTO3 UVAI depends on SO_2 amount: for loadings larger than about 50 DU, the UVAI is highly correlated with the SO_2 column ($r^2 = 0.9$). Radiative transfer calculations show that the effect of SO_2 absorption on OMTO3 UVAI is on the order of 0.5-2 units for an SO_2 column of 100 DU, depending on the Sun's position and viewing geometry. The SO_2 absorption cross section at 331 nm is about one order of magnitude larger than at other wavelengths used for UVAI calculation, so that apart from the OMTO3 UVAI, no UVAI product is affected (even at 388 nm the SO_2 cross section is too small to cause significant artifacts in UVAI).
A comparison of the three OMI UVAI products, shown in the figure below, confirms that, as Simon Carn mentioned, the viewing angle dependence is only apparent in the OMAERUV (340/380 nm) and OMAERO (354/388 nm) products. In addition, the OMTO3 UVAI appears to be affected by SO_2 absorption (most apparent in the lower right panel). Although these are interesting findings, they do not fall within the scope of the study presented here. We are currently investigating the viewing angle dependence of UVAI in more detail and will present the results in a forthcoming publication.

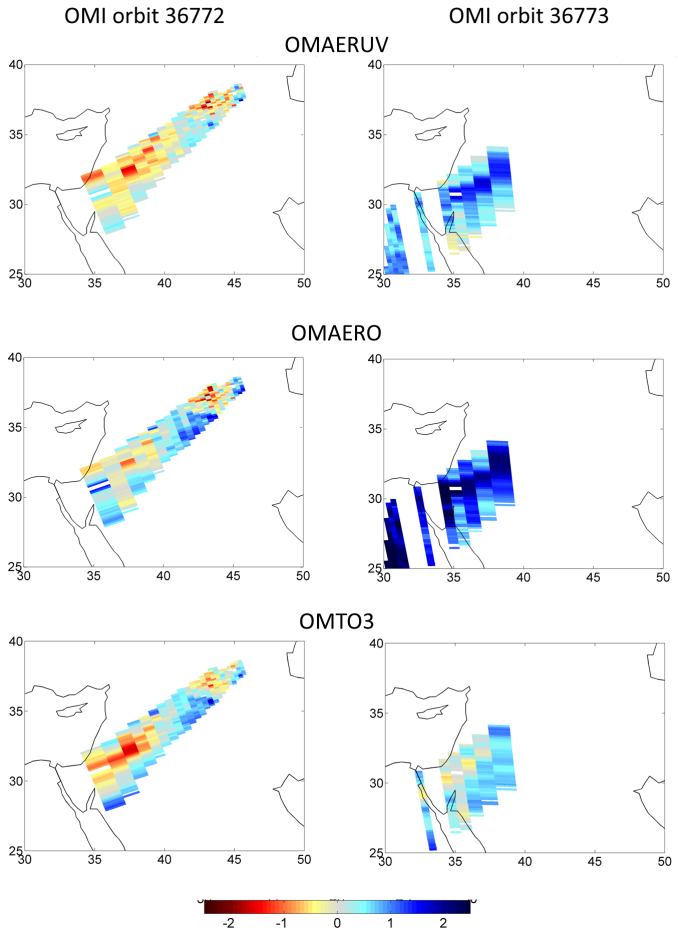

Figure 1: *UVAI from the three different OMI products for the two orbits sampling the volcanic plume on June 14, 2011. From top to bottom: OMAERUV (340/380 nm), OMAERO (354/388 nm), and OMTO3(331/360 nm).*

In the revised manuscript, we expanded Section 2.2 by describing UVAI in more detail (see the reply to comment 12). We now also more clearly state which OMI UVAI product we use for our analysis (comment 13). In addition, we changed the paragraph on UVAI viewing angle dependence in Sect. 5.1 (page 7757, lines 9-19) to include a note about the differences in angle dependences between the OMI UVAI products:

Apart from the limb data, the most clear evidence that sulphate aerosols were detected by the UVAI comes

from OMI, where the strong viewing angle dependence of UVAI indicates the presence of a high-altitude ($>$ 11 km) aerosol plume with very high single-scattering albedo ($>$ 0.97). We note that due to the slightly different definitions of UVAI, the viewing angle dependence is not the same for the three available OMI UVAI products (OMAERUV, OMAERO, and OMTO3). The strong viewing angle dependence complicates the interpretation of UVAI, but could be turned into an advantage with regard to the determination of aerosol properties. For elevated aerosol plumes large enough to cover the entire OMI swath, or plumes captured twice by OMI from opposing viewing angles (as in the case of the Nabro plume on June 14) the viewing angle dependence can be exploited to determine the aerosol absorption and plume altitude. This approach can be applied to volcanic plumes, as shown here, but potentially also to extensive smoke or elevated pollution plumes. We are currently investigating the viewing angle dependence of UVAI in more detail using RTM calculations and OMI UVAI data.

Referee comment: 1. 7740, L5-7: 'Formation of sulphate aerosols in the stratosphere takes about a month...' this statement could be misleading since, whereas the average SO_2 lifetime in the stratosphere may be $\approx$ 1 month, aerosol formation starts immediately.

Authors' reply: The referee is right; the statement was changed to:

It is generally assumed that the formation of sulphate aerosols in the stratosphere is a relatively slow process, but in plumes from explosive eruptions significant amounts of aerosols have been seen to form within a few hours.

Referee comment: 2. 7740, L24-25: the very first lines here suggest that sulphate aerosols are a primary product of volcanic eruptions, whereas it is mostly SO_2 that is emitted and subsequently converted to sulphate. This should be clarified. Furthermore, the long stratospheric lifetime of SO_2 is also due to slow gas-phase oxidation rates.

Authors' reply: We clarified this by changing the sentence to:

Volcanic eruptions are an important source of sulphate aerosols to the stratosphere due to the emission of large quantities of the precursor SO_2 .

And we changed lines 25-26 to:

In the stratosphere, low gas-phase oxidation rates of SO_2 cause a relatively slow build-up of sulphate aerosols and low deposition rates cause aerosol lifetimes on the order of months to years (e.g. Bluth et al., 1997, and references therein).

Referee comment: 3. 7741, L3: I think at least one reference should be given for the direct radiative effect of sulphate aerosol.

Authors' reply: All literature references addressing the different effects of sulphate aerosols were swept together in the list in line 6, because different aspects are mentioned in each paper and citing them separately would make the section quite unreadable. For completeness we added a reference to Robock et al. (2000) to the list and modified the section (starting on page 7740, line 26) as follows:

The presence of sulphate aerosols in the stratosphere has wide-ranging effects: their purely scattering (non-absorbing) nature in the UV-visible wavelength range causes radiative cooling at the Earth's surface as the aerosol layer reflects more solar radiation into space. Local warming of the stratosphere due to absorption of upwelling long-wave surface radiation by sulphate droplets has also been shown to occur. In addition, the increased total aerosol (reactive) surface area leads to enhanced ozone destruction (see, e.g. Hofmann and Solomon, 1989; Hansen et al., 1992; McCormick et al., 1995; Bluth et al., 1997; Robock et al., 2000; SPARC, 2006 and references therein for all above-mentioned effects).

Referee comment: 4. 7741, L12: whether the Nabro eruption was exceptional in terms of SO_2 release depends on context (e.g., it was much smaller than Pinatubo or El Chichon), so please clarify.

Authors' reply: Since the eruption of Nabro was the largest since Pinatubo in terms of SO_2 and sulphate emissions (as mentioned by the referee in comment 16), we would like to continue calling it "exceptional".

Referee comment: 5. 7741, L16: the altitude of the tropopause at Nabro should be specified.

Authors' reply: Done by adding (dynamical tropopause at 16.8 km, according to ECMWF) to line 16.

Referee comment: 6. 7741, L23: SO_2 removal mechanisms depend on altitude, and dry/wet deposition may be very significant in the 'atmosphere' as a whole (since most SO_2 is emitted at low altitudes).

Authors' reply: We thank the referee for this remark; to avoid excluding any processes, but without going into too much details that are unnecessary to our study, we changed line 23 to:

One of the main removal mechanisms of SO_2 in the atmosphere is the oxidation to H_2SO_4 and the subsequent formation of (or uptake in) aerosol particles.

Referee comment: 7. 7741, 28 and 7742, 2: the vast majority of volcanic eruption plumes are dominated by water vapor, both entrained atmospheric water and volcanogenic water, and I find it highly unlikely that Nabro was exceptional in this regard. I would not use the Smithsonian reports as evidence for a water-rich plume as there can be no actual measurements to confirm this. It may have been motivated by the 'bright' (i.e., high albedo) appearance of the Nabro plume in visible satellite imagery (e.g., MODIS), but many fresh volcanic plumes have a similar appearance.

Authors' reply: In response to the second comment by referee 2, the section was completely revised, making this comment redundant. The section now reads:

This assumption is based on data from eruption plumes at altitudes above 25 km (Pinatubo and El Chichón) and SO_2 life times are much shorter for volcanic plumes at lower altitudes or with higher water vapour content: life times on the order of 1-2 weeks were found for Kasatochi (Krotkov et al., 2010), Sarychev (Haywood et al., 2010), and Cerro Hudson (Constantine et al., 2000). For the earliest emitted "high-altitude" plumes (at 15-18 km) from Nabro, an SO_2 life time of 5 days was found (Theys et al., 2013) and, consequently, an amount of sulphate aerosols large enough to be visible from space could be observed within at most 36 hours of the eruption. This is in agreement with a study by (Sawamura et al., 2012), who report the presence of an aerosol plume in the lower stratosphere at 17 km altitude using data from a ground-based lidar station in Sede Boker, Israel, approximately 30 hours downwind of the first explosion of Nabro.

Referee comment: 8. 7742, 5: note that the Sawamura et al. (2012) study is ambiguous about the nature of the aerosol detected at Sede Boker they speculate that 'ash and sulfate' may be present but I would not say that they are 'in agreement' with the results presented here. I don't think they even used depolarization information to distinguish solid vs. liquid phase aerosol.

Authors' reply: This is true, but the altitude of the aerosol layer is in agreement with our findings and, although Sawamura and co-workers did not try to characterize the aerosols, data from other investigations point to sulphate aerosols as a major component of the volcanic plume. As this issue was addressed in detail above, we prefer to leave the statement as it is.

Referee comment: 9. 7742, 10: the full names of the satellite sensors should be given here, rather than later on.

Authors' reply: Done.

Referee comment: 10. 7743, 6: 'horizontally narrow' does this refer to the horizontal extent of the plume, and/or is vertical thickness also important?

Authors' reply: The phrase 'horizontally narrow' refers only to the horizontal extent of the plume. Due to the onion-peeling approach, the retrieved limb profile is not affected by vertical thickness as long as the overlying optical depth does not become too high. We changed the phrase to:

plumes with a limited horizontal extent.

Referee comment: 11. 7744, 13: OMI pixels are only 13x24 km in size close to nadir; they increase in size gradually towards the swath edge.

Authors' reply: This was implied by the addition of 'away from the swath edges', but to make the point more clear, we changed the phrase to:

at the centre of the swath, the pixels measure 13 km x 24 km.

Referee comment: 12. 7745, 12: the actual definition of UVAI (i.e., an equation) is not given here; given that UVAI is one of the main foci of the paper, I think it should be explicitly stated.

Authors' reply: This is a good point. We added a more extensive description of UVAI to Section 2.2:

To investigate if ash was present in the volcanic plume we use the UVAI (also known as 'residue'). The definition of the UVAI is based on the principle that aerosol optical properties differ from those of molecules, so that the presence of an aerosol layer leads to a change in contrast between the reflectance at a wavelength λ (e.g. 340 nm) and at a reference wavelength λ_0 (e.g. 380 nm) in comparison to an aerosol-free atmosphere. The calculation of UVAI requires radiative transfer modelling of reflectances at λ and λ_0 for aerosol-free scenes bounded by a Lambertian surface with an albedo value chosen such that measured and modelled reflectances at λ_0 are equal. The UVAI is obtained by calculating the ratio between the measured (R^{meas}) and modelled (R^{model}) reflectances at λ (Torres et al., 1998):

$$UVAI = -100 \cdot log_{10} \left(\frac{R^{meas}}{R^{model}} \right)_{\lambda}$$

Positive values of UVAI are commonly referred to as AAI (Absorbing Aerosol Index), which is a well-known and much-used qualitative indicator of aerosols that absorb radiation in the UV range, such as mineral dust, biomass burning smoke, and volcanic ash (Herman et al., 1997; Torres et al., 1998; de Graaf et al., 2005). The AAI depends on plume altitude and is most sensitive to elevated layers of absorbing aerosols, in particular if these are located over bright surfaces such as ice or clouds (see, e.g. Hsu et al., 1999; Fromm et al., 2010). The SCI (SCattering Index), which consists of UVAI<0, was only recently introduced as an indicator for aerosols that do not or only weakly absorb UV radiation (Penning de Vries et al., 2009), such as sulphate aerosols and other secondary aerosols formed from anthropogenic or biogenic emissions of trace gases. UVAI generally depends on aerosol amount, size distribution, absorption, and altitude. The AAI is more sensitive to aerosol amount and plume altitude than the SCI because absorption by aerosols causes larger changes in spectral contrast in the UV range than aerosol scattering (Torres et al., 1998; de Graaf et al., 2005; Penning de Vries et al., 2009). A big advantage of the UVAI is that it can be meaningfully interpreted in the presence of clouds; however, clouds influence UVAI and must be taken into account for quantitative analysis of UVAI (Penning de Vries and Wagner, 2011).

Referee comment: 13. 7745, 15: the authors should clarify exactly which operational OMI aerosol product was used OMAERUV? Also, the OMI UVAI in OMAERUV does not use exactly the same wavelengths as SCIAMACHY (354 and 388 nm are used).

Authors' reply: We used the OMAERUV product, which is calculated from 340 and 380 nm reflectances. OMAERO, KNMI's product, uses 354 and 388 nm reflectances, whereas OMTO3 uses the reflectances at 331 and 360 nm. This is now clarified in the manuscript by changing line 14 to:

For consistency, OMI "OMAERUV" UVAI data obtained from the same wavelengths were used (downloaded from: http://mirador.gsfc.nasa.gov/).

Referee comment: 14. 7746, 13: some parameters require definition here. Is 'g' the asymmetry parameter?

Authors' reply: Yes, it is. This is clarified by changing the part in brackets to:

(single-scattering albedo 1.0, asymmetry parameter g=0.6, corresponding to a size distribution with a mean diameter of 80 nm and width=1.6 (Deshler et al., 2006))

Referee comment: 15. 7747, 4: this sentence needs rewriting it starts 'It can be very well measured from space...' but then continues '...both from the ground and from space'.

Authors' reply: We corrected the typo by removing the first 'from space'.

Referee comment: 16. 7748, 6: again, the SO_2 emission was large but not 'exceptional'. It was likely the largest volcanic SO_2 emission since Pinatubo, if the entire Nabro eruption ($\tilde{1}$ month) is considered.

Authors' reply: As it was a remarkably large eruption in terms of SO_2 load, we would like to stick to the term exceptional, as mentioned in reply to comment 4.

Referee comment: 17. 7750: in the discussion of the variable OMI UVAI signal here, it is critical to explicitly state which definition of the UVAI the authors are using (see major comment above) as the change in sign of the UVAI is not observed in all versions.

Authors' reply: We now mention explicitly that we use OMI OMAERUV UVAI in Section 2.2. See the

discussion following the second main comment above and the answer to minor comment 13.

Referee comment: 18. 7750, 15: in addition to the clouds clearly visible in the MODIS image, I think the presence of thin 'subvisible' clouds (e.g., thin cirrus) collocated with the volcanic plume cannot be ruled out. This could also include small ice particles in the volcanic plume itself.

Authors' reply: True — subvisible cirrus cannot be ruled out, and neither can small ice particles. However, as the amount is so small that the MODIS IR retrieval does not pick them up, we do not expect those to affect the results significantly. See also our replies to issue (1) above and to comment 23 below.

Referee comment: 19. 7751, 3-5: were the optical properties of the sulphate aerosols obtained from the literature? If so, a reference should be provided. Otherwise, some further justification for the choice of these parameters (and also the AOT values used in RT calculations) is needed. This also applies to P7746, L12-15.

Authors' reply: The optical properties were taken from Deshler et al., 2006 and represent volcanic aerosols. The AOT values used in the RT simulation of the volcanic plume are loosely based on the AOT measured on June 14 by the AERONET station at Sede Boker. This information was added by changing the section (starting on line 3) to:

We performed RTM simulations using the aerosol parameters representative of volcanic sulphate particles: g = 0.6, single-scattering albedo of 1.0 (from (Deshler et al., 2006), also used in the limb retrieval) and with an Ångström exponent of 1.5. The maximum AOT detected by AERONET is approximately 0.4 (after subtraction of a constant background), therefore we modelled the UVAI from the sulphate plume by assigning an AOT (at 380 nm) of 0.4 to those pixels with SO_2 column densities exceeding 30 DU, an AOT of 0.2 for pixels with SO_2 column densities between 15 and 30 DU, and an AOT of 0.1 to those with SO_2 columns smaller than 15 DU.

Referee comment: 20. 7751, 13-14: the authors should also note that the region of meteorological clouds to the north of the volcanic plume also shows a change in UVAI with viewing angle (Fig. 3). Presumably the UVAI remains negative or close to zero in this case due to the lower cloud altitude?

Authors' reply: We thank the referee for pointing this out: we hadn't noticed the feature yet. We now mention it in the manuscript by inserting the following sentence after line 16:

Note that the UVAI caused by clouds directly north of the volcanic plume also changes with viewing angle (compare panels A and B). The variation in UVAI is not as pronounced as that observed for the volcanic plume because the meteorological clouds are at much lower altitude, but also because the viewing angle dependence of clouds is smaller than that of aerosols (compare Figs. S1 and S2 in the supplement).

Referee comment: 21. 7756, 15: but if small ash particles had acted as ice nuclei and become coated by ice then they would likely not produce a positive UVAI either.

Authors' reply: This is a true and interesting point, however it does not change the fact that we found no direct evidence of ash. We only want to point out that the bulk of detected aerosols did not consist of ash; the possible presence of a small amount of ash, whether or not coated in ice, is not of importance here. We therefore prefer to leave the section as it is.

Referee comment: 22. 7756, 20: I think it is highly improbable that the positive UVAI outside the SO_2 plume is due to volcanic ash – there will be significant amounts of desert dust in the region.

Authors' reply: Yes, we think so too (as stated in lines 20-22). Encouraged by this referee comment, we changed the word 'uncertain' on line 22 to 'unlikely'.

Referee comment: 23. 7757, 9: I dont think the fact that the ice cloud disappeared from MODIS visible imagery can be used as proof that there was no longer any ice present in the volcanic plume. Small subvisible ice particles could still be present.

Authors' reply: We can never exclude the presence of subvisible particles, but the fact that the MODIS IR retrieval did not detect any (ice) cloud particles is a strong indication that no significant amount of ice was present (line 25, page 7749). We now repeat this argument in the discussion section:

Yet, a negative UVAI signal corresponding to the SO_2 plume can be discerned in SCIAMACHY and OMI

UVAI data on 14 June, albeit much weaker, as shown in Fig. 2 (panels E and F), whereas there is no indication of ice particles in MODIS visible or IR data (as mentioned in Sect. 4.1).

Referee comment: 24. 7757, 17: exploiting the viewing angle dependence would seem to be highly dependent on the assumed phase function (and hence also on particle size and scattering regime).

Authors' reply: That would appear so, but initial calculations on our part show that the effect of varying g is similar to the effect of varying single-scattering albedo, i.e., raising or lowering the UVAI, but not affecting the shape of the viewing angle dependence. This is shown exemplarily for one case (AOT = 1, SSA = 1 , aerosol layer at 18-19 km altitude, g varied between 0.5 and 0.7) in the figure below. We added this information to the supplementary material. The viewing angle dependence of UVAI is an interesting topic that we are currently investigating in more detail.

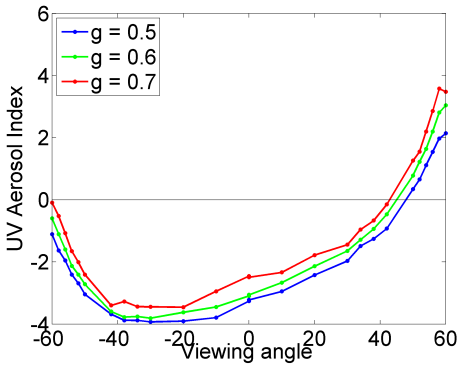

Figure 2: *Modelled line-of-sight dependence of UVAI for various aerosol scenarios. RTM calculations were performed with McArtim; solar zenith angle was set to 20 degrees and surface albedo 0.1. Aerosols have AOT = 1.0, SSA = 1.0, and Ångström coefficient 1.5 (AOT given at 380 nm); the aerosol layer was located at 18-19 km.*

Referee comment: 25. 7758, 9: the Sawamura et al. (2012) paper should be referenced here I presume this is where the AOT value of 0.17 comes from?

Authors' reply: Yes it is; the reference was inserted.

Referee comment: 26. 7758, 10: the Sede Boker lidar data were collected on June 14, so it would seem more consistent to compare the lidar AOT with the SCIAMACHY limb AOT measured on the same day (i.e., the profile in Fig. 5)?

Authors' reply: The argument sounds logical, but is not necessarily true: the part of the plume sampled on June 14 by SCIAMACHY is only the tail end, whereas the AOT obtained by lidar was probably measured where the plume was thickest. The reason we chose the profiles from June 16 for the comparison is that those show the most well-defined plumes. But we agree with the referee and replaced the AOT by values measured on June 14 and 15, which are not very different from those measured on June 16. The section now reads:

When we integrate the profiles shown in Figs. 5 and 9, much smaller values of AOT are found, e.g. 8×10^{-3} for the profile of pixel 4 on June 14, or 5×10^{-3} for the pixel 3 from orbit 48584 on June 15.

Referee comment: 27. Fig. 3: left and right panels are confused in the caption.

Authors' reply: We corrected the mistake.