

Dear ACP Editor and Anonymous Referees,

Please find below our answers to the 2 Anonymous Referees. In 'black' the Referee comments, in 'blue' our response.

We would like to thank the Referees for their constructive comments.

## **Referee 1**

### General Comments

In this new study Carboni et al. applied an SO<sub>2</sub> retrieval scheme for IASI introduced in a paper in 2012 to fourteen minor and major volcanic eruptions in 2008 to 2012. Column density maps illustrate the horizontal distribution and altitude-dependent time series the vertical distribution of the volcanic SO<sub>2</sub> emissions. Volcanic plume altitudes and SO<sub>2</sub> total mass estimates derived from the retrievals are important pieces of information, in particular for modelling studies. Plume heights are validated with CALIOP satellite measurements and SO<sub>2</sub> total mass is validated with data from ground-based Brewer instruments for selected case studies.

The paper presents an interesting topic and is within the scope of ACP. The presentation is generally clear, but I found that a number of specific questions and issues remains open. I would recommend the paper to be published once the specific comments listed below are carefully addressed. Also, as a general comment, I got the impression that some time should be spent on improving the textual flow of the manuscript and language editing to improve readability.

### Specific Comments

short title: Just wondering if the short title of the paper could be replaced by "SO<sub>2</sub> vertical distribution of volcanic plumes" to make it more specific?

Good suggestion, done.

p24646, l19-23: Here it is mentioned that nadir spectrometer measurements can be used to infer information on SO<sub>2</sub> plume altitude, but no details are given. How good does this work? Can you provide references?

A short description of which satellite spectrometers have been used to retrieve SO<sub>2</sub> in previous literature is presented between line 24 of page 24646 and line 17 on the following page (24647).

p24648, l9-10: Hilton et al. (2012, Fig. 3) show that the IASI nominal and actual NeDT may be as large as 0.3-0.4K in the spectral range from 650-1750/cm. The range of 0.1-0.2K mentioned in your paper may be applicable for the 7.3 and 8.7 micron wavebands of SO<sub>2</sub>?

Thanks for the suggestion, we changed 0.1-0.2K into '0.1-0.3K in the SO<sub>2</sub> spectral range considered, according to Hilton et al 2012 (fig 3)'.

To clarify our reasoning a bit, this paragraph was intended to introduce the IASI instrument. This is not the error that goes into the covariance matrix that we are using in the retrieval.

p24651, l16-19: It is mentioned that SEVIRI imagery is used to identify volcanic plumes in the CALIOP data. I was wondering what CALIOP and SEVIRI are actually sensitive to? I guess CALIOP is sensitive to sulphate aerosols (rather than SO<sub>2</sub>)? Please clarify also for SEVIRI.

We added this in the paper: 'CALIPSO is sensitive to aerosol and water droplets that scatter sunlight; in the case of volcanic eruptions these aerosols are H<sub>2</sub>SO<sub>4</sub> and ash. SEVIRI is sensitive to both ash and SO<sub>2</sub> since its channel around 8.7 micron is within the SO<sub>2</sub> absorption band.'

p24652, l24-28: I have trouble seeing this separation of the plume as there are a number of features visible in the plots. Perhaps this could be made more clear by adding arrows or other markers in the plots?

We added two arrows indicating both the lower and the higher parts of the plume, as well as a description in the caption.

p24653, l8-12: Here it is mentioned that the Brewer SO<sub>2</sub> measurements are sensitive to both anthropogenic SO<sub>2</sub> in the lower troposphere as well as overpasses of volcanic plumes. Is your IASI SO<sub>2</sub> retrieval scheme also sensitive to the lower troposphere? I was thinking that IR nadir sounders are most sensitive to SO<sub>2</sub> in the mid/upper troposphere and lower stratosphere. Assuming that there are some differences in vertical sensitivity of the different measurements (Brewer versus IR nadir sounder), do these pose limits to this comparison?

The vertical sensitivity of IR sounders really depends on the gas studied and the spectral range used. In this SO<sub>2</sub> retrieval scheme we use simultaneously all the IASI channels in 2 spectral ranges: one around the 7.3 micron (within the water vapour absorption, more sensitive to mid/upper troposphere, and used in IASI ULB algorithm), and the second one around the 8.7 micron where SO<sub>2</sub> is in a window region, sensitive down to the surface (and not used in IASI ULB algorithm). We included both spectral ranges in the retrieval so as to be sensitive down to the surface. The error increases moving closer to the surface (due to reduced temperature contrast) and this is represented in the error bars.

p24654, l16-19: What is the reference for the 6 m/s average wind speed used to calculate the influence radius? This value looks a bit small. How do the correlations between the Brewer instruments and IASI (Fig. 5) change if the influence radius is increased?

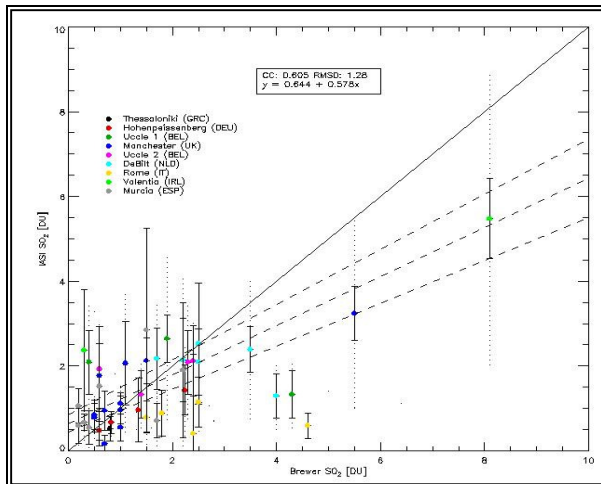


Figure 1: same as Fig 5 but considering 300 km radius.

Relaxing the coincidence criteria, i.e. increasing the allowed distance between Brewer and IASI, results in worse comparisons. Going from 200km to 300km radial distance changes the correlation from 0.76 to 0.60 and the best fit line slope is also decreasing from 0.73 to 0.58. The 200km radius was chosen as a trade-off between making sure the same atmosphere is viewed between satellite and ground while keeping the number of collocations large enough for statistically-significant results to be extracted.

p24655, I23-27: Triangular interpolation is used to fill data gaps, but what happens if IASI measurement tracks are overlapping at high latitudes? How do you consider the different measurement times of the satellite orbits?

The overlapping orbits crossing the area are re-gridded together. We consider all pixels (including those of overlapping orbits) and average them together into the grid boxes.

'For each eruption the IASI orbits are grouped into twelve hour intervals in order to have, twice-daily maps, maps of IASI retrieved SO<sub>2</sub> amount and altitude. These maps are re-gridded into a 0.125° lat/lon boxes.'

changed into: 'For each eruption the IASI orbits are grouped into twelve hour intervals in order to have two maps, each day, of IASI retrieved SO<sub>2</sub> amount and altitude. IASI pixels of overlapping orbits are averaged together. These maps are gridded into 0.125° lat/lon boxes.'

p24656, I20-23: How do these SO<sub>2</sub> total mass estimates agree with other studies? What are the uncertainties of these estimates? From the data presented in Fig. 7 it might be possible to estimate SO<sub>2</sub> lifetimes, which would be very interesting for modellers, I think.

Comparisons with values reported by other studies are discussed for individual eruptions in session 6. The uncertainties of the SO<sub>2</sub> total masses are reported as error bars in all points of Fig 7. See also answer to referee 2 general comment 1.

The referee is right, it might be possible to estimate the lifetime from this dataset. However, the dataset has to be handled carefully; it would really help to identify the days in which there was no new injection of SO<sub>2</sub> from the volcano. This would add a significant amount of effort and has not been performed in this study.

Added the following sentence in the text: 'Using the time series created by this dataset it might be possible to estimate the SO<sub>2</sub> lifetime (not included in this work).'

p24657, l12-14: The WMO definition of the tropopause does not depend on the pressure profile. Do you mean you used log-pressure altitudes calculated from ECMWF pressure/sigma levels to estimate the tropopause altitudes rather than considering geopotential heights?

We compute the tropopause, given temperature and pressure profiles and using the hydrostatic equation to give altitude from these. From this we can calculate lapse rate as used in the WMO definition i.e.

'The first tropopause is defined as the lowest level at which the lapse rate decreases to 2 deg K per kilometer or less, provided also that the average lapse rate between this level and all higher levels within 2 kilometers does not exceed 2 deg K.'

World Meteorological Organization. 1992. *International Meteorological Vocabulary*, 2nd ed. Geneva. ISBN 978-92-630-2182-3.

[http://www.wmo.int/pages/prog/lsp/meteoterm\\_wmo\\_en.html](http://www.wmo.int/pages/prog/lsp/meteoterm_wmo_en.html).

This citation has been added to the paper.

p24658, l4-6: It might be good to recap the meaning of different VEI values at this point. Which plume altitudes are expected/found for a VEI of 1-5? Is stratospheric injection for the different VEI values considered to be likely or not?

We added this in text of the paper: 'The VEI is a semi-quantitative index of eruption size, which for contemporary eruptions can be used as a 'threshold' to determine the likelihood of stratospheric injection (Newhall and Self, 1982). Eruptions of VEI 4 and larger are expected to have strong plumes and be associated with significant stratospheric injections. Eruptions of VEI 3 are intermediate in size, with eruptive ash plumes that rise 5 – 15 km above the vent. Based on analysis of eruption statistics, 25 – 30% of VEI 3 eruptions may reach the stratosphere (Pyle et al., 1996). Eruptions of VEI 2 and smaller are not expected to reach the stratosphere.'

And added these references:

Newhall, C.G., and Self, S. (1982). The volcanic explosivity index (VEI) – an estimate of explosive magnitude for historical volcanism. *J. Geophys. Res.* 87, 1231–1238

Pyle, D.M., Beattie, P.D. and G.J.S. Bluth, (1996) Sulphur emissions to the stratosphere from explosive volcanic eruptions, *Bulletin of Volcanology*, 57, 663-671

p24658, l7-10: At this point it is likely not clear to the reader what you mean by "dynamic effect". The explanation at this point seems a bit short and vague.

Changed 'dynamic effect' with 'atmospheric effect'

p24659, l11-21: This discussion is a bit long, but I took as a key point that it is very important to have good plume altitude information for the SO<sub>2</sub> retrievals. Wrong plume altitudes may lead to significant differences in SO<sub>2</sub> total mass estimates. Perhaps you could add a sentence at the end of this paragraph to conclude and stress this point, as it provides strong motivation for this work?

Thanks, good point. We added this sentence:

'This shows how important it is to have good altitude information for the SO<sub>2</sub> retrieval as assumptions on plume altitude may lead to significant differences in SO<sub>2</sub> total mass estimates.'

p24662, l25-29: It would be interesting to see how you rate your findings regarding the discussion of the transport of Nabro SO<sub>2</sub> emissions in terms of the Asian Monsoon circulation or direct injection. Fig. 11 shows that most SO<sub>2</sub> is located below the tropopause, i.e., it might not be a clear case of direct injection into the stratosphere as suggested by Fromm et al.?

The vertical distribution of Fig 11 is a 'summary plot' that simply reports the average altitude and spread of tropopause computed for all the plume locations every 12 hours. These averaged tropopause lines are computed over different latitudes where the tropopause altitude varies significantly, and to use this summary plot in order to identify if the injection was in the stratosphere or not can be misleading. The analysis in Fromm et al 2014 was more specific, using our IASI data but also involving other instruments, and showed that Nabro directly injected SO<sub>2</sub> into stratosphere twice.

On the other hand, summary profiles can be useful to identify where the majority of SO<sub>2</sub> was present and, as seen in fig 11, we can observe that most of the SO<sub>2</sub> emitted by Nabro was mainly confined below 6km.

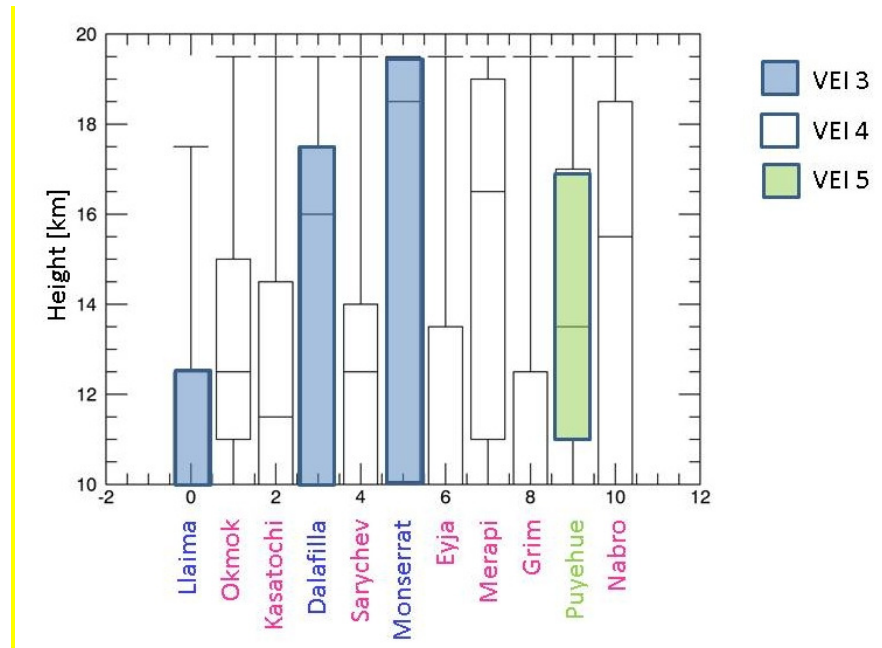
p24664, l25-27: Stating that IASI is "consistent" with CALIPSO and the Brewers instruments is good, but I think you should try to be more precise. How good/uncertain are the plume altitudes and total mass estimates from your retrieval scheme?

The uncertainties in our retrieval change pixel by pixel with the atmospheric and plume conditions (altitude and amount). All these are taken into account in the Optimal Estimation scheme and reported as error in any retrieved values. In this paper we visualize the uncertainty as error bars in the various plots.

p24665, l7-11: The conclusion that your paper demonstrated that the VEI "is a poor index of the potential height to which volcanic SO<sub>2</sub> is injected" is not evident to me. Perhaps you could add a table or a scatter plot showing the VEI and plume heights for the different eruptions to demonstrate that there is no good correlation.

It is not straightforward to define the altitude of one eruption. We show below a plot obtained for the eruptions with VEI 3, 4 and 5. The black lines show the range of altitudes retrieved, the boxes depict the altitude range where the 50% of the SO<sub>2</sub> mass falls into, the horizontal lines within the box are the altitudes of the maximum of SO<sub>2</sub> mass. The altitudes where the 50% SO<sub>2</sub> mass is contained and where the maximum mass is retrieved are obtained by

averaging of our vertical distributions (in fig. 8-11) over time, except from Puyehue where only the first 48 hours are considered, as the rest of the Puyehue eruption is mainly injecting lower than 10 km. Different colours are representing different VEI . Eruptions like Monserrat and Dalafilla, both VEI 3, are 'higher' than Puyehue, VEI 5.



We believe this show that the VEI and plume height are uncorrelated. If the editor feels this plot should be in the paper we will add it.

p24665, l12-15: Is it to be expected that many volcanic SO<sub>2</sub> plumes reach a level near the tropopause? What would be the physical mechanism for this?

A typical temperature for an air mass coming from an eruption (pyroclastic flow) is around 700 Celsius = 973 K. Using, as example of potential temperature profile, the following figure from here: [http://www.cpc.ncep.noaa.gov/products/stratosphere/theta/theta\\_info.shtml](http://www.cpc.ncep.noaa.gov/products/stratosphere/theta/theta_info.shtml), the plume can arrive higher than 30 km. Following mixing with the atmosphere the cooler volcanic plume arrives at equivalent potential temperature of about 450 K. Because the hot plume is mixing with the cold air, it follows that the plume does not reach the potential temperature altitude of the throat temperature

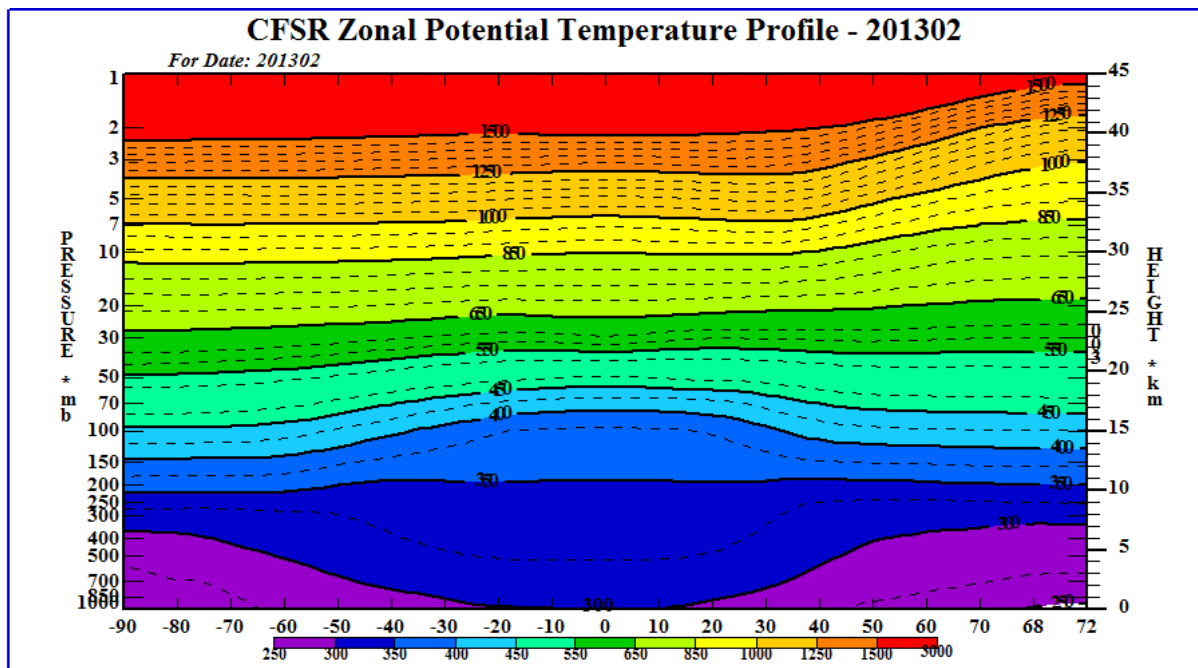


Fig. 3: It seems the CALIPSO measurement tracks used for comparison were not well chosen as they have only limited overlap with the SO<sub>2</sub> plume (as shown by IASI)? Perhaps CALIPSO tracks located more to the west would have been better? What was the rationale for your choice?

The CALIPSO tracks to the west have more than 2 hours difference to the IASI measurements and are rejected by the coincidence criteria applied.

Fig. 4: Some data points from the SO<sub>2</sub> retrieval seem to have very large uncertainties in plume altitude (up to +/- 8 km). I guess these large uncertainties are related some kind of retrieval problem? I wanted to suggest to remove these points from Fig. 4 (as well as Figs. 1-3) as they do not seem to tell us a lot? The comparison with CALIOP seems to be meaningful only if the SO<sub>2</sub> retrieval delivers a plume altitude with reasonable accuracy (e.g. with an uncertainty less than +/- 2 km or similar).

Large uncertainties are usually associated with low amounts of SO<sub>2</sub>, and since the uncertainty in the altitude retrieval is of interest to many, we have opted to leave these values in the plot.

It is a plot to compare the IASI retrievals with CALIPSO, but it can also give to the reader a visual example of how the uncertainty in IASI altitude varies point by point with atmospheric and plume conditions. Eliminating the points with more than 2 km error can give the false impression that IASI can retrieve altitude with less the 2km error, whereas this really depends on the SO<sub>2</sub> amount (see Carboni et al 2012).

Fig. 6: Does gray color indicate that the retrieval failed because of an SO<sub>2</sub> column density larger than 100 DU? It seems there are a number of data gaps (white color) in the SO<sub>2</sub>

column density and plume height maps near 70 W and 45 N, which are filled by zero rather than interpolation from neighbouring pixels? Zooming in on the plume could help to check this.

The left column present the outputs of the retrieval (amount and altitude), and the right column presents the regridded data (amount and altitude). The grey colours have values higher than the colour bar (we added this in the caption now). Yes there are gaps in the left column due to orbit gaps or retrievals that do not pass the quality control. There are several reasons for not passing the control, for e.g. the forward model cannot fit the measurements or the iterative routine did not converge within 10 iterations, etc.

Fig. 7: This figure might be a bit confusing as the emissions of some volcanic eruptions are overlapping in time (e.g. Grimsvötn, Puyehue, and Nabro in May and June 2011), but are plotted as separate events here. Full time ranges including actual days are given for some eruptions in the plot key (which seems helpful), but are missing for others. What defines the time span of data points shown for eac volcano in this plot?

For instance, the time span for the Puyehue is much longer than for the Nabro (whereas the total SO<sub>2</sub> mass is much lower for the Puyehue than for the Nabro)?

We added the data interval in the legend and added this explanation in the caption: 'The total SO<sub>2</sub> amount reported here is computed using the geographic area associated with the eruption. For eruptions which overlap in time (e.g Grimsvötn, Puyehue, and Nabro in May and June 2011) the SO<sub>2</sub> loads within each respective area are considered and plotted separately.'

The time interval is what we have analysed. No particularly criteria were involved beyond checking that we include the main "interesting" phases of each eruption and the plume evolution.

Figs. 8-11: The SO<sub>2</sub> column density maps are all limited to a maximum value of 5 DU, which seems pretty low compared to actual maximum values that occurred in the case studies. Different color bar ranges for each plot or a log-scale with fixed range for all plots may help to retain this information. The plots of the vertical distribution present the total mass of SO<sub>2</sub> (in Tg). However, the total mass in each box will depend on the vertical and temporal binning. The box sizes (in particular the vertical binning) should be mentioned in the caption. Alternatively, SO<sub>2</sub> density (Tg/km/day) rather than total mass could be shown.

We have changed the colour bars of the maps and added the vertical binning in the caption for the vertical distribution.

Fig. 10: The tropopause data for the Puyehue case study (bottom row) shows very large fluctuations and has data gaps. Is this considered to be realistic? Are there any problems with the estimation of the WMO tropopause height in this case study? (Very large fluctuation of the tropopause height is also visible for the Eyjafjallajökull case study in Fig. 8.)

We added this sentence in the section explaining the tropopause lines (p24657 l20):  
'The reported values of the tropopause are computed using the location of the volcanic pixels only. Eyjafjallajökull and Puyehue eruptions cover the latitude range between 30-80 N



and -20 -60 S respectively, so spanning a large range in tropopause heights. Day to day variation are sometimes large due to small amounts of SO<sub>2</sub> being sometimes detected or not from one day to the next (coupled to the wide range of latitudes spanned by the plumes).

#### Technical Corrections

The following technical comments have been corrected, thanks.

p24644, l5: "Instrument" -> "Interferometer"

p24646, l8: "observe into" -> "observe in" (?)

p24648, l3: "2007" -> "October 2006"

p24648, l21: "1 C" -> "1C"

p24649, l19: "is it" -> "it is"

p24653, l13: "Metop" -> "METOP" (also in other places of the paper)

p24655, l7: remove "instrument" (?)

p24655, l22: "into a 0.125" -> "into 0.125"

p24656, l17: "other, it" -> "other. It"

p24658, l11: "of a multilayer" -> "of multilayer"

p24662, l29: "Caliop" -> "CALIOP"

p24665, l4: "lidar or limb Michelson... (MIPAS) measurements" -> "lidar or limb measurements (e.g., MIPAS)" (?)

Figs. 1-4: The plot titles of the CALIPSO plots (e.g., "CAL\_LID\_L1-ValStage1-V3-01.2010...") could be replaced by more readable style.

Section 6.4.1 should be Section 6.5, I guess?

#### References

Hilton, F., R. Armante, T. August et al., Hyperspectral earth observation from IASI: Five years of accomplishments. *Bulletin of the American Meteorological Society*, 93, 347–370, 2012.

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#### Referee 2

In their manuscript "The vertical distribution of volcanic SO<sub>2</sub> plumes measured by IASI", Carboni et al. present results from retrievals of volcanic plume SO<sub>2</sub> content and vertical

distribution performed on IASI data. They present a large data set covering several volcanic eruptions, provide some validation of SO<sub>2</sub> layer height with CALIPSO measurements and of SO<sub>2</sub> columns with ground-based Brewer observations and describe in detail results for a set of large volcanic eruptions.

The paper is well written but in some places, minor English corrections needed. It is clearly structured and reports on an impressive and relevant data set which is of interest to the community and fits well into the scope of ACP. I therefore recommend this manuscript for publication in ACP after taking into account the comments and suggestions given below.

#### General comments

1 My only real concern with the manuscript is that this is by no means the first IASI SO<sub>2</sub> product and for data users, it would be relevant to know how this product compares with other published IASI SO<sub>2</sub> products, at least in terms of SO<sub>2</sub> columns. There is some brief discussion of comparisons in the text but ideally, Fig. 7 or parts of it should contain data points from other IASI retrievals as well. It would be good if some direct comparison could be added here or in another figure.

The objective of this paper is to compare observations of a number of volcanic eruptions. The retrieval and its accuracy have been discussed in other papers.

We added this section to the paper:

‘Comparisons with the ULB IASI SO<sub>2</sub> dataset, as well as UV-Vis instruments such as GOME2/MetopA and OMI/Aura, have been performed and are cited in the relevant eruptions sections. We simply note here that for the Grímsvötn eruption our data were compared to the ULB IASI/MetopA SO<sub>2</sub> and the BIRA GOME2/MetopA SO<sub>2</sub> retrievals [Koukouli et al., 2015];; for the Etna continuous outflows with INGV MODIS/Terra, RAL MODIS/Terra, ULB IASI/MetopA, DLR GOME2/MetopA and ground-based Flame network measurements [Spinetti et al., 2015] and finally, for the Eyjafjallajökull eruptions, with the DLR GOME2/MetopA, the BIRA OMI/Aura and the AIRS data [Carboni et al., 2012; Koukouli et al., 2014] . More recently data for the Bradabunga eruption, were compared with the BIRA OMI/Aura data [Schmidt et al., 2015];’

Reference added:

Koukouli et al., SACS2/SMASH Validation Report on the Eyjafjallajökull & Grímsvötn eruptions, [http://sacs.aeronomie.be/Documentation/LAP-AUTH-SACS-ValidationReport\\_FINAL.pdf](http://sacs.aeronomie.be/Documentation/LAP-AUTH-SACS-ValidationReport_FINAL.pdf), last accessed: 17 December 2015.

Schmidt, A., S. Leadbetter, N. Theys, E. Carboni, C.S. Witham, J.A. Stevenson, C.E. Birch, T. Thordarson, S. Turnock, S. Barsotti, L. Delaney, W. Feng, R.G. Grainger, M.C. Hort, Á. Höskuldsson, I. Ialongo, E. Ilyinskaya, T. Jóhannsson, P. Kenny, T.A. Mather, N.A.D. Richards and J. Shepherd, Satellite detection, long-range transport, and air quality impacts of volcanic sulfur dioxide from the 2014-2015 flood lava eruption at Bárðarbunga (Iceland), *Journal of Geophysical Research*, 120, 9739–9757, 2015. (doi:10.1002/2015JD023638)

2. I'm also not fully convinced that the lengthy description of the individual events in section 6 is needed but on the other hand it does provide quick information for people interested in a specific eruption.

We believe these descriptions are important as they place the SO<sub>2</sub> plume in context of the eruption sequence. As the referee will see from the references these descriptions are not readily available from the literature.

3. As this is an interesting data set it would be good to indicate how it can be accessed.

We added this sentence:

'The dataset can be made available by contacting the author Elisa Carboni <elisa.carboni@physics.ox.ac.uk>'

4. The result that most of the eruptions inject SO<sub>2</sub> into the tropopause region is interesting and somewhat surprising. Can you give any possible explanation for this finding?

See resp. to referee 1 referring to p24665, l12-15

Is it in line with other observations of the height of volcanic emissions?

This is the most comprehensive examination of plume altitude that we are aware of. But it makes perfect sense in terms of the troposphere which is the region of the atmosphere which can be convectively unstable.

Do you think there is a risk that this result is biased by your SO<sub>2</sub> plume height retrieval?

No, because the retrieval is based on the thermal signal received by the instrument and is mainly sensitive to the temperature of the SO<sub>2</sub> layer. However, errors may arise if the ECMWF temperature profile used as input is not appropriate. In principle, the retrieval can 'swap' between layers with the same temperature above and below the tropopause, but the presence of water vapour absorption seems to add information content on the altitude of the plume and helps to discern between altitudes above and below the tropopause. In any case, the point in this retrieval where we are most confident is exactly when the retrieval finds the temperature minimum i.e. the tropopause level.

5. The validation of SO<sub>2</sub> column amount with Brewer observations is useful but the good correlation really hinges on one single point (Valentia).

The referee is right and I wish as well that we have more data-points. Currently, these are the only ground-based assessments that we can work with and they do not coincide with strong SO<sub>2</sub> plumes. Valentia is the highest value and it is clearly encouraging that we agree well with it. The scatter for lower SO<sub>2</sub> amounts is to be expected due to errors in both Brewer and IASI.

Technical comments•

The following technical comments have been corrected, thanks.

p 24645, l 7: amounts => amount

p 24654, l 23: then => than

p 24655, l 9 and following: I do not understand your explanation for the overestimation of low amounts – please clarify

p 24655, l 12: then => than

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p 24655, l 21: something is wrong with this sentence, please check

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p 24655, l 22: into a 0.125 => into 0.125

•

p 24658, l 25: release => released

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p 24660, l 17: to Kasatochi => as Kasatochi

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p 24661, l 11: and implicate => and implicated

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Figure 5: to a different Brewer ground station => to different Brewer ground stations

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Figure 8: Add explanation of tropopause lines in caption

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Figure 10: Etna plots shows => Etna plots show