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_2 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_2 emissions. Volcanic plume altitudes and SO2 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 SO2 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 "SO2 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_2 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_2 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_2?

Thanks for the suggestion, we changed 0.1-0.2K into '0.1-0.3K in the SO_2 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_2)? 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_2SO_4 and ash. SEVIRI is sensitive to both ash and SO2 since its channel around 8.7 micron is within the SO2 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_2 measurements are sensitive to both anthropogenic SO_2 in the lower troposphere as well as overpasses of volcanic plumes. Is your IASI SO_2 retrieval scheme also sensitive to the lower troposphere? I was thinking that IR nadir sounders are most sensitive to SO_2 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_2 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_2 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?

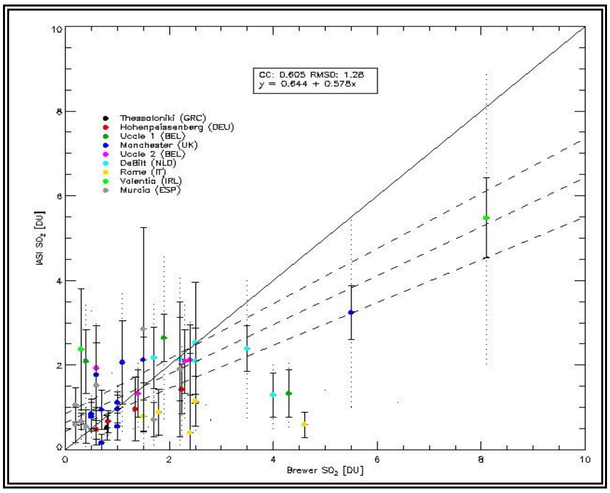


Figure 1: same as Fig 5 but considering 300 km radius.

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, l23-27: 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_2 amount and altitude. These maps are regridded 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_2 amount and altitude. IASI pixels of overlapping orbits are averaged together. These maps are gridded into 0.125◦ lat/lon boxes.'

p24656, l20-23: How do these SO2 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_2 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_2 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_2 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_2 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.

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_2 retrievals. Wrong plume altitudes may lead to significant differences in SO_2 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_2 retrieval as assumptions on plume altitude may lead to significant differences in SO_2 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 SO2 emissions in terms of the Asian Mon-soon circulation or direct injection. Fig. 11 shows that most SO2 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_2 into stratosphere twice.

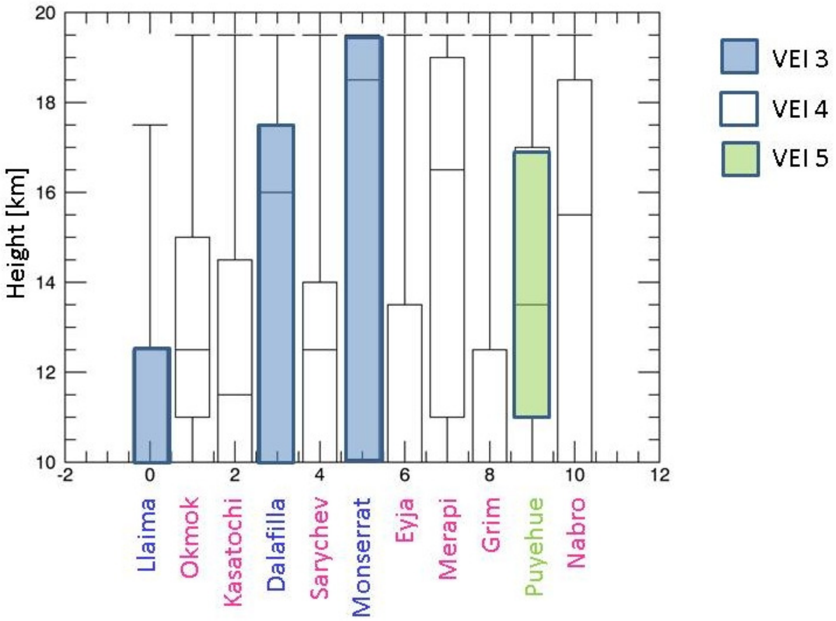
On the other hand, summary profiles can be useful to identify where the majority of SO_2 was present and, as seen in fig 11, we can observe that most of the SO_2 emitted by Nabro was mainly confined below 6km.

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_2 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_2 mass falls into, the horizontal lines within the box are the altitudes of the maximum of SO_2 mass. The altitudes where the 50% SO_2 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_2 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, 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

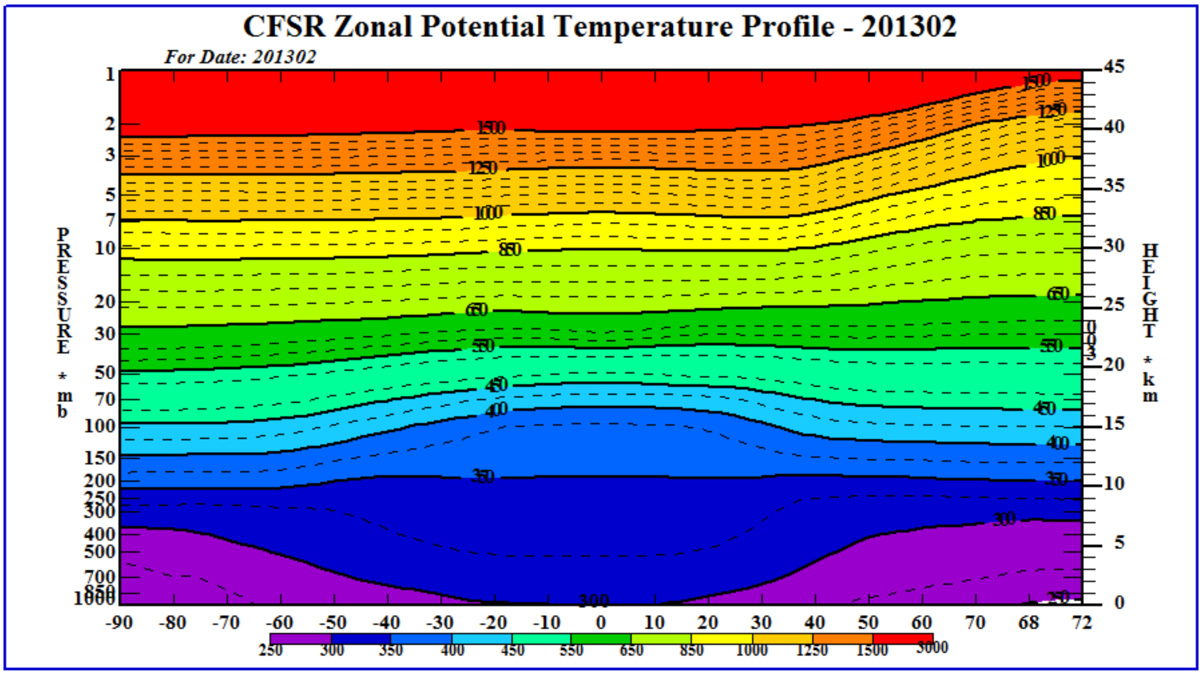


Fig. 3: It seems the CALIPSO measurement tracks used for comparison were not well chosen as they have only limited overlap with the SO_2 plume (as shown by IASI)? Perhaps CALIPSO tracks located more to the west would have been better? What was the rationale for your choice?

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_2 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_2 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_2, 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_2 amount (see Carboni et al 2012).

Fig. 6: Does gray color indicate that the retrieval failed because of an SO2 column density larger than 100 DU? It seems there are a number of data gaps (white color) in the SO_2

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_2 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_2 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_2 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 SO2 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_2 (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_2 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 Eyjafjallajokull 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. Ejyafjallajokull 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 SO2 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.

Referee 2

In their manuscript “The vertical distribution of volcanic SO2 plumes measured by IASI”, Carboni et al. present results from retrievals of volcanic plume SO2 content and vertical distribution performed on IASI data. They present a large data set covering several volcanic eruptions, provide some validation of SO_2 layer height with CALIPSO measurements and of SO2 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_2 product and for data users, it would be relevant to know how this product compares with other published IASI SO_2 products, at least in terms of SO_2 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_2 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_2 and the BIRA GOME2/MetopA SO_2 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, 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_2 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 SO2 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_2 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_2 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 SO2 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_2 plumes. Valentia is the highest value and it is clearly encouraging that we agree well with it. The scatter for lower SO_2 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

- p 24655, l 21: something is wrong with this sentence, please check
- p 24655, l 22: into a 0.125 => into 0.125
- p 24658, l 25: release => released
- p 24660, l 17: to Kasatochi => as Kasatochi
- p 24661, l 11: and implicate => and implicated
- Figure 5: to a different Brewer ground station => to different Brewer ground stations
- Figure 8: Add explanation of tropopause lines in caption
- Figure 10: Etna plots shows => Etna plots show

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The vertical distribution of volcanic SO_2 plumes measured by IASI

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Abstract

Sulphur dioxide (SO_2) is an important atmospheric constituent that plays a crucial role in many atmospheric processes. Volcanic eruptions are a significant source of atmospheric SO_2 and its effects and lifetime depend on the SO_2 injection altitude. The Infrared Atmospheric Sounding ~~Instrument~~ Interferometer (IASI) on the ~~Metop~~ METOP satellite can be used to study volcanic emission of SO_2 using high-spectral resolution measurements from 1000 to 1200 cm^{-1} and from 1300 to 1410 cm^{-1} (the 7.3 and 8.7 µm SO_2 bands). The scheme described in Carboni et al. (2012) has been applied to measure volcanic SO_2 amount and altitude for fourteen explosive eruptions from 2008 to 2012. The work includes a comparison with independent measurements: (i) the SO_2 column amounts from the 2010 Eyjafjallajökull plumes have been compared with Brewer ground measurements over Europe; (ii) the SO_2 plumes heights, for the 2010 Eyjafjallajökull and 2011 Grimsvötn eruptions, have been compared with CALIPSO backscatter profiles. The results of the comparisons show that IASI SO_2 measurements are not affected by underlying cloud and are consistent (within the retrieved errors) with the other measurements. The series of analysed eruptions (2008 to 2012) show that the biggest emitter of volcanic SO_2 was Nabro, followed by Kasatochi and Grímsvötn. Our observations also show a tendency for volcanic SO_2 to be injected to the level of the tropopause during many of the moderately explosive eruptions observed. For the eruptions observed, this tendency was independent of the maximum amount of SO_2 (e.g. 0.2 Tg for Dalafilla compared with 1.6 Tg for Nabro) and of the volcanic explosive index (between 3 and 5).

1 Introduction

Sulphur dioxide (SO_2) is an important atmospheric constituent, important in many atmospheric processes (Stevenson et al., 2003; Seinfeld and Pandis, 1998; Schmidt et al., 2012). Volcanic eruptions are a significant source of atmospheric SO_2, with its effects and lifetime depending on the SO_2 injection altitude. In the troposphere these include acidification of rainfall, modification of cloud formation and impacts on air quality and vegetation (Ebmeier et al., 2014; Delmelle et al., 2002; Delmelle, 2003; Calabrese et al., 2011). In the stratosphere SO_2 oxidises to form a stratospheric H_2SO_4 aerosol that can affect climate for several years (Robock, 2000). Volcanoes contribute about one third of the tropospheric sulphur burden ($14 \pm 6\,\mathrm{Tg\,S\,yr^{-1}}$; Graf et al., 1997; Textor et al., 2003). The annual ~~amounts~~ amount of volcanic SO_2 emitted is both poorly constrained and highly variable. The uncertainty in released SO_2 arises from the stochastic nature of volcanic processes, very little or no surface monitoring of many volcanoes, and from uncertainties in the contribution of volcanic sulphur emitted by quiescent (non-explosive) degassing. The effects of SO_2 in the atmosphere depend not only on the amount released but also the altitude of the plume. Altitude information is important for atmospheric chemistry, as SO_2 reactions and depletion times change with height and atmospheric composition, particularly as a function of water vapour concentration (Kroll et al., 2015; Glasow, 2010; McGonigle et al., 2004; Mather et al., 2003).

Most volcanic eruptions are accompanied by the release of SO_2 to the atmosphere, and data both on the quantity emitted, and the height at which it is injected into the atmosphere are valuable indicators of the nature of the eruption.

It is important to monitor volcanic SO_2 plumes for air safety (Brenot et al., 2014; Schmidt et al., 2014) as sulphidation of nickel alloys cause rapid degeneration of an aircraft engine (Encinas-Oropesa et al., 2008). SO_2 is also used as a proxy for the presence of volcanic ash, altough the collocation of SO_2 and ash depends upon the eruption so this approach is not always reliable for hazard avoidance (Sears et al., 2013). Mitigation strategies to avoid volcanic SO_2 and ash are currently based on ground monitoring and satellite measurements to access the location and altitude of the proximal volcanic plume, followed by the use of dispersion models to forecast the future position and concentration of the plume (Tupper and Wunderman, 2009; Bonadonna et al., 2012; Flemming and Inness, 2013). The outputs of the models are strongly dependent on the assumed initial plume altitude mainly because of the variability of the wind fields with altitude. The dispersion models may use an ensemble of possible initial altitudes to identify all places where the plume can potentially arrive.

Hence improving the accuracy of the initial plume altitude reduces the uncertainty in the location of the transported volcanic SO_2.

Limb viewing satellite instruments, such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and the Microwave Limb Sounder (MLS), can estimate the vertical profile of SO_2 (Höpfner et al., 2013; Pumphrey et al., 2014) in the upper troposphere and lower stratosphere (UTLS), but generally cannot observe ~~into~~ in the lower troposphere. Nadir measurements, by both spectrometers and radiometers, have been used to retrieve SO_2 column amounts, assuming the altitude of the plume, e.g., the Total Ozone Monitoring Spectrometer (TOMS, Carn et al., 2003), the Moderate Resolution Imaging Spectroradiometer (MODIS, Watson et al., 2004; Corradini et al., 2009), the Ozone Monitoring Instrument (OMI, Krotkov et al., 2006; Yang et al., 2009; Carn et al., 2009), the Atmospheric Infrared Sounder (AIRS, Prata and Bernardo, 2007), the Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER, Pugnaghi et al., 2006; Campion et al., 2010), the Global Ozone Monitoring Experiment 2 (GOME-2, Rix et al., 2012), and the Spinning Enhanced Visible and Infrared Imager (SEVIRI, Prata and Kerkmann, 2007).

Nadir spectrometer measurements in the ultraviolet (OMI, GOME-2) and infrared (AIRS, the Tropospheric Emission Sounder, TES, IASI) can be used to infer information on the SO_2 plume altitude. This is based on the fact that the top-of-atmosphere radiance of a plume, with the same amount of SO_2 but at another altitude, will have a different spectral shape within the SO_2 absorption bands.

UV spectra have been used to retrieve information on SO_2 altitude by Nowlan et al. (2011) using an optimal estimation retrieval applied to GOME-2 measurements. Yang et al. (2010) used a direct fitting algorithm to retrieve the SO_2 amount and altitude from OMI data. Both of these techniques were applied to the 2008 Kasatochi eruption which injected a relatively large amount of SO_2 high (circa 12 km) in the atmosphere. A new scheme to retrieve altitude from GOME-2 has been developed by Van Gent et al. (2015) and applied to Icelandic eruption case studies (Koukouli et al., 2014).

Volcanic SO_2 retrievals from satellite data in the thermal infra-red (TIR) part of the spectrum are based on two regions of SO_2 absorption around 7.3 and 8.7 µm. The strongest

SO_2 band is at 7.3 µm. This is in a strong water vapour (H_2O) absorption band and is not very sensitive to emission from the surface and lower atmosphere. Above the lower atmosphere this band contains information on the vertical profile of SO_2. Fortunately differences between the H_2O and SO_2 emission spectra allow the signals from the two gases to be decoupled in high resolution measurements. The 8.7 µm absorption feature is in an atmospheric window so it contains information on SO_2 from throughout the column. In Clerbaux et al. (2008) the SO_2 absorbing feature around 8.7 µm was used to retrieve the total SO_2 amount and profile from TES data by exploiting this instruments ability to resolve the change in SO_2 line-width with pressure. The 7.3 µm spectral range has been used by Clarisse et al. (2014) in a fast retrieval of SO_2 altitude and amount. Their approach, based on Walker et al. (2012), assumes that the SO_2 is located at different altitudes, choosing the altitude that gives the best spectral fit.

In this paper, the Carboni et al. (2012) retrieval scheme (hereafter C12) has been applied to study the vertical distribution of SO_2 for the major eruptions during the period 2008–2012 and for some minor low tropospheric eruptions such as an Etna lava fountain.

The aims of this paper are to test the retreived SO_2 amount and altitude against other datasets (we used CALIPSO backscattering profile to test the altitudes and Brewer ground measurements for the column amounts) and to study the volcanic plumes from different eruption types and in different locations.

2 IASI data

There are currently IASI sensors on-board the European weather satellites METOP-A and METOP-B. METOP-A has been operating since ~~2007~~ October 2006 and was the first of three satellites scheduled to operate for a total of 14 years. METOP-B has been operating since September 2012. The satellites cross the Equator on the descending node at a local time of around 9:30 a.m.

IASI is a Fourier transform spectrometer covering the spectral range 645–2760 cm^{-1} (3.62 to 15.5 µm) with spectral sampling of 0.25 cm^{-1} and an apodized spectral resolution of 0.5 cm^{-1} (Blumstein et al., 2004). It has a nominal radiometric noise ~~(NeDT at 280)~~ of 0.1~~–0.2(650–1750).~~-0.3K in the SO_2 spectral range considered, according to Hilton et al. (2012) (fig 3).

The field-of-view (FOV) consists of four circular footprints of 12 km diameter (at nadir) inside a square of 50 km $\times$ 50 km, step-scanned across track (with 30 steps). The swath is 2100 km wide and the instrument can nominally achieve global coverage in twelve hours.

Observations are co-located with the Advanced Very High Resolution Radiometer (AVHRR), providing complementary visible/near infrared measurements. IASI carries out nadir observation of the Earth simultaneously with the Global Ozone Monitoring Experiment (GOME-2) also on-board METOP. GOME-2 is a UV spectrometer measuring SO_2 in the UV absorption band and is used for both Differential Optical Absorption Spectroscopy (DOAS) (Rix et al., 2012) and optimal estimation retrievals (Nowlan et al., 2011) of SO_2. More information about IASI can be found in Clerbaux et al. (2009). The IASI level 1C data (geolocated with apodized spectra) used here were obtained from both the British Atmospheric Data Centre (BADC) archive and EUMETSAT Unified Meteorological Archive Facility (UMARF) archive.

3 SO_2 retrieval scheme

The retrieval scheme follows Carboni et al. (2012) where the SO_2 concentration is parametrised as a Gaussian profile in pressure (with 100 mb spread). Using IASI measurements from 1000 to 1200 cm^{-1} and from 1300 to 1410 cm^{-1}, an optimal estimation retrieval (Rodgers, 2000) is employed to estimate the SO_2 column amount, the height and spread of the SO_2 profile, and the surface skin temperature.

The forward model, based on Radiative Transfer model for TOVS (RTTOV) (Saunders et al., 1999) but extended to include SO_2 explicitly, uses ECMWF temperatures interpolated to the measurement time and location. The retrieval technique uses an error covariance matrix, based on a climatology of differences between the IASI measurements and SO_2-free forward modelled spectra. Any differences not related to SO_2 between IASI spectra and those simulated by a forward model are included in the covariance matrix. Note that the SO_2 retrieval is not affected by underlying cloud. If the SO_2 is within or below an ash or cloud layer its signal will be masked and the retrieval will underestimate the SO_2 amount, in the case of ash this is indicated by a cost function value greater than two.

The retrieval is performed for every pixel where the SO_2 detection result is positive (Walker et al., 2011, 2012). The scheme iteratively fits the forward model (simulations) with the measurements, to seek a minimum of a cost function. The solution, when the measurements do not contain enough information to retrieve all the parameters in the state vector, is strongly affected by the assumed a priori values. When the SO_2 amount decreases, the spectral information decreases, and ~~is it~~ it is not possible to retrieve both SO_2 amount and altitude. This often happens at the edge of the plume. In this work we use 400 ± 500 mb as the a priori value for plume altitude and 0.5 ± 100 DU for column amount. In addition, only quality controlled pixels are considered; these are values where the minimisation routine converges within 10 iterations, the SO_2 amount is positive, the plume pressure is below 1000 mb and the cost function is less then 10.

Rigorous error propagation, including the incorporation of forward model and forward model parameter error, is built into the system, providing quality control and error estimates on the retrieved state. Retrieved uncertainties increase with decreasing altitude, nevertheless it is possible to retrieve information in the lower troposphere and monitor volcanic degassing. In the case of two or multiple SO_2 layers the forward model assumption of a single Gaussian layer is a source of error (and this error is not included in the pixel by pixel error estimate). In this case the retrieved altitude is an effective altitude between the two (or more) plume layers; in particular the altitude will be the one that is radiatively closer to the measured spectra and the altitude will be the radiatively equivalent altitude.

The altitude of the SO_2 plume strongly modulates the retrieval error as the contrast between plume temperature and surface temperature is a critical factor. The error in SO_2 amount decreases with an increase in plume altitude. Typical uncertainties are 2 DU for a plume centred at 1.5 km and less then 1 DU for plumes above 3 km.

When using the result from this setting of the retrieval scheme the following caveats should be considered:

1. The retrieval is valid for SO_2 column amounts less then 100 DU (limit used in the computation of RTTOV coefficients) and altitudes less then 20 km (due to the used Gaussian profile with 100 mb spread). For higher eruptions one should use a thinner Gaussian profile, for column amounts bigger then 100 DU new RTTOV coefficients will be needed. At present both of these conditions produce results that do not pass the quality control.

2. Quality control and orbit gaps can produce artefacts (see Sect. 6).

3. Care should be taken when using the altitude to infer if a plume is above or below the tropopause. In this case, it is desirable to combine or confirm these measurements using other measurements. This is because there are altitudes with the same temperature above and below the tropopause, and the SO_2 signal could be similar. Nevertheless, the IASI spectrum contains altitude information from the overlapping of SO_2 with other gas absorption.

4. The global annual mean variability between real atmospheric profiles and ECMWF profiles is included in the error covariance matrix, and then propagated into the retrieval errors. However we cannot exclude some extreme events where the difference between ECMWF profiles and the real profile is at the tail of the annual global statistical distribution. In this case, local and seasonal covariance matrices could be used. For the retrievals presented here a global covariance matrix has been used, computed using days from all four seasons.

5. Volcanoes also emit water vapour. If this emission is larger than the water vapour variability considered in the error covariance matrix, the results could be affected by errors that are not included in the output errors.

Comparisons with the ULB IASI SO_2 dataset, as well as UV-Vis instruments such as GOME2/MetOp-A 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/MetOp-A SO_2 and the BIRA GOME2/MetOp-A SO_2 retrievals (Koukouli et al., 2014). For the Etna continuous outflows with INGV MODIS/Terra, RAL MODIS/Terra, ULB IASI/METOP-A, 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 Bárdarbunga eruption, were compared with the BIRA OMI/Aura data (Schmidt et al., 2015).

4 Altitude comparison with CALIOP

Vertical profiles of aerosol and clouds are provided by NASA's Cloud Aerosol Lidar with Orthogonal Polarization instrument (CALIOP, Winker et al., 2009) carried on the CALIPSO satellite. CALIOP's vertical/horizontal resolution is 0.06/1.0 km at altitudes between 8.2 and 20.2 km. CALIPSO is part of the A-train with an equatorial overpass time of ~ 13.30. CALIOP and IASI have coincident measurements around $\pm 70^{\circ}$ latitude and moving from this latitude towards the equator causes the time difference between the two measurements to increase.

CALIOP backscatter profile have been averaged (to 3 km along-track, 250 m vertical) and CALIOP observations of volcanic plumes have been identified using SEVIRI false colour images based on the infrared channel at 8.7, 11 and 12 µm (Thomas and Siddans, 2015). CALIOP is sensitive to aerosol and water droplets that scatter sunlight; in the case of volcanic eruptions these aerosols are H_2SO_4 and ash. SEVIRI is sensitive to both ash and SO_2 since its channel around 8.7 µm is within the SO_2 absorption band.

The criteria used to define a coincidence between CALIOP and IASI pixels were: a distance of < 100 km and a time difference of < 2 h. With these relatively strict criteria, the selected coincident data are only from the Icelandic Eyjafallajökull and Grimsvötn eruptions. For the other eruptions considered (Puyehue-Cordón Caulle, June 2011; Nabro, June 2011; Soufrière Hills, February 2010) the differences in acquisition time between the CALIOP and

IASI measurements are more than 2 h. It may be possible to analyse these eruptions in the future by using the wind field to account for the movement of the plume with time.

Figure 1 shows the comparison between CALIOP and IASI for the Eyjafjallajökull plume on the 7 May 2010. This is the CALIOP track presented by Thomas and Prata (2011) in which they identify both ash and SO_2 plumes in the southern part of the track (less then 55° N) and SO_2 only plumes in the northern part of the track (the scattering feature around 5 km between 55–60° N where the scattering signal is possibly from H_2SO_4 aerosol resulting from the oxidation of volcanic SO_2).

Ash and SO_2 are not necessarily advected together (Thomas and Prata, 2011; Sears et al., 2013), but in these case studies (Figs. 1–4), there is good agreement between IASI and the backscattering features within the IASI error bars. This may be due to the presence of H_2SO_4 associated with the SO_2 plume. An important aspect to note is that in nearly all of the coincidences for the Eyjafallajökull case (with the exception of a few pixels on 14 May) the volcanic plume is above a lower meteorological cloud. This is identifiable as the scattering layer at around 1 km height in Fig. 1 and between 1 and 4 km in Figs. 2 and 3. This comparison against CALIOP in the presence of water cloud confirms that the retrieval is not affected by underlying cloud. This is because the variability that such cloud introduces is represented by the error covariance matrix which includes radiance differences between the clear forward model and cloudy IASI spectra. An accurate retrieval of altitude is also an important factor for the estimate of SO_2 amount, this is because the thermal SO_2 signal can be up to one order of magnitude different between different altitudes. A good comparison of altitude with CALIPSO then gives confidence in both altitude and column amount.

Figure 4 shows the Grimsvötn plume at the beginning of the eruption. There is separation between the higher part of the plume (dominated by SO_2) moving higher and spreading north, and the lower part of the plume (dominated by ash, but still with some SO_2 associated with it) moving south-west. It was this part of the plume that moved over Europe in the following days and caused airspace closure.

5 Comparison with Brewer ground data

During the Eyjafjallajökull eruption in April and May 2010, the volcanic plume overpassed several European ground measurement stations equipped with Brewer instruments. Brewer instruments are UV spectrophotometers, principally dedicated to measuring the total ozone column but also capable of determining total column SO_2 (Kerr et al., 1981). Extraction of the SO_2 signal from the UV measurements is performed as a second step after the ozone quantity retrieval due to the much lower SO_2 absorption features strength (De Muer and De Backer, 1992). Depending on the amount of atmospheric SO_2 affecting the instruments, the Brewer spectrophotometer has been shown to be sensitive both to anthropogenic SO_2 loading in the lower troposphere (Zerefos et al., 2000) as well as to an overpass of a volcanic plume, which produces a strong SO_2 signal within the ozone absorption bands. These ground-based measurements have lately been used in Rix et al. (2012) in a comparison with the GOME-2/~~Metop-A~~ METOP-A SO_2 retrievals and in the validation work of the European Space Agency, ESA, projects (SMASH and SACS2[1]) on the 2010 and 2011 Icelandic eruptions (Koukouli et al., 2014). The World Ozone and Ultraviolet Radiation Data Centre (WOUDC) is an active archive facility that includes quality-assured Brewer ground-based O_3 measurements and also provides SO_2 daily averages. The dataset for April and May 2010 was downloaded from the WOUDC archive (http://www.woudc.org/) for all European sites with Brewer instruments.

The reported SO_2 amount in non-volcanic conditions (e.g. over Europe before 15 April 2010) from Brewer stations varies considerably which can mostly be attributed to the small signal-to-noise ratio of the SO_2 signal compared to the ozone signal. This variability may also be due to insufficient reported data quality control with respect to SO_2 retrieval, since most stations focus on the quality of the ozone measurements and not their by-products. Negative values can also be present in the daily average datasets, a result of the nominal Brewer algorithm's inability to resolve small atmospheric SO_2 amounts. Therefore, only

[1]"Study on an End-to-End system for volcanic ash plume monitoring and prediction" (SMASH) and "Support to aviation control service 2" (SMASH2).

a subset of the available Brewer sites in the WOUDC database were selected for this study. Stations were not selected if they reported a majority of negative values (such as Reading, UK; La Coruna, Spain, and so on) for the two months considered (April and May 2010), or had negative values less than −1 DU (such as Madrid, Spain; Poprad-Ganovce, Slovakia). These negative values point to the small amount of the volcanic gas reaching the specific locations. Despite recording several negative values, the Valencia, Spain, site was considered since it was presented in Rix et al. (2012), after checking that the measurements coincident with the presence of the IASI plume were statistically significantly larger than the average background measurements.

All the positive values of the selected ground stations (listed in Fig. 5) have been compared with the IASI measurements of the SO_2 plume. The SO_2 estimates available from WOUDC are daily averages. In order to compare these datasets with the IASI observations, all the satellite pixels of the morning and evening orbits within a 200 km radius from the Brewer site have been averaged. The choice of radius was made since at an average wind speed of 6 $m\,s^{-1}$ an air parcel will travel 250 km in 12 h, so with this spatial criteria we are including in the satellite averaging all pixels that might be overpassing the Brewer location within a temporal frame of 12 h (daytime). At the same time, the averaged IASI error and standard deviation have been computed. The results are presented in the scatter plot in Fig. 5. The linear fit is computed considering the IASI average error and a fixed error of 0.5 DU for the Brewer measurements. Note that the variability of the IASI SO_2 amount within 200 km is often much bigger ~~then~~ than the IASI error bar. A correlation coefficient of 0.76 with root mean square differences of 1.16 DU has been found. This result is encouraging for the IASI retrieval, even if this initial comparison alone cannot be considered a comprehensive validation exercise because (1) it is difficult to assess the quality of Brewer SO_2 daily average values; (2) the comparison is restricted to the Eyjafjallajökull eruption, an eruption where the SO_2 plume covered a small range of altitudes (between 2 and 5 km) and a relative small loading amount.

From Fig. 5 it can be noted that for loadings up to and around 2 DU both types of observation appear to depict the same atmospheric SO_2 loading, which, depending on the location of the site, might be both of anthropogenic and volcanic provenance. For values between 2 and 3 DU there appears to be a slight underestimation by IASI ~~instrument~~ of around 0.5 DU, well within the statistical uncertainty. For higher loadings still, the Brewer instruments report higher SO_2 values than the satellite.

The overestimation of low amounts can be explained because the a priori SO_2 amount used by IASI retrieval is 0.5 DU and in case of no information content from the measurments (as happen when there is no detectable SO_2) the output of an optimal estimation retreival is the retreival is the a priori value. One factor that can explain the underestimation (by IASI of $SO_2 > 3$ DU) is that this IASI retrieval is sensitive to SO_2 values higher ~~then~~ than the climatological SO_2 amount considered in the IASI data ensemble used to compute the error covariance matrix. In the case of the Eyjafallajökull eruption the north Atlantic and European region of April and May 2009 were used to compute the error covariance matrix, so the retrieval could be insensitive (biased) to the average values of SO_2 amount in that region, which includes the European background value of SO_2.

6 Total mass and vertical distribution

The SO_2 retrieval algorithm has been applied to several volcanic eruptions in the period 2008–2012. For each eruption the IASI orbits are grouped into twelve hour intervals in order to have ~~, twice-daily maps, maps~~ two maps, each day, of IASI retrieved SO_2 amount and altitude.

IASI pixels of overlapping orbits are averaged together. These maps are ~~regridded intoa~~gridded into 0.125° lat/lon boxes.

An example of maps for the Nabro eruption is shown in Fig. 6. Note that with only METOP-A the IASI data have a gap between the orbits at tropical latitudes, this gap is filled with the ~~Metop-B~~ METOP-B launch in September 2012. It is also possible to have gaps in SO_2 coverage due to pixels that did not pass quality control. The regridding routine fills gaps by a triangular interpolation of neighbouring pixels.

Figure 6 shows an example of how the regridding routine fills in missing data. However, because of the possible creation of artefacts, the regridding should be used carefully. For example, in the case of a plume covering the edge of one orbit but with no plume present in the adjacent orbit, regridding can fill up the gaps between the orbits with a bigger plume than would be reasonable to expect. For the case studies presented here, all the regridded IASI maps have been inspected "by eye" to check that no particularly significant artefacts have been introduced.

The total SO_2 mass present in the atmosphere is obtained by summing all the values of the regularly gridded map of SO_2 amounts. In particular every grid-box column amount is multiplied by the grid-box area to obtain the SO_2 mass, and all the grid-box masses are summed together to obtain the total mass of SO_2 for each IASI image. The total mass errors are obtained in the same way from the grid box errors, i.e. all the box errors are summed to produce the total mass error. This is an overestimation of the error, but considering the mean squared error as total error will be an underestimation. This is due to the presence of systematic errors within the retrieval. The systematic errors are included within the error estimate but cannot be considered independent of each other~~, it~~. It is more likely that, if present, the systematic error will become a bias in the region and time considered. The time series of these total masses, together with the errors, are presented in Fig. 7 for the studied volcanic eruptions.

Using the IASI dataset it is possible to follow the plume evolution of several volcanic eruptions. Within the eruptions considered Nabro produced a maximum load of 1.6 Tg of SO_2, followed by Kasatochi (0.9 Tg), Grímsvötn (0.75 Tg), Copahue (0.72 Tg) and Sarychev (0.60 Tg). Using the time series created by this dataset it might be possible to estimate the SO_2 lifetime (not included in this work).

From the eruptions presented in Fig. 7, there is a wide spread of error-bars, depending on the SO_2 amount, altitude and atmospheric conditions. In general, plumes tend to spread and dissipate with time, covering a larger area (i.e. more IASI pixels) with smaller quantities. This produces smaller SO_2 signals and consequently bigger retrieval errors. This is the main cause of the general tendency of increasing error bars with time.

It is also possible to estimate the SO_2 mass present between two altitude levels. Doing this every 0.5 km between 0 and 20 km gives a vertical distribution of SO_2 every ~ 12 h. These results are shown, in chronological order, in Figs. 8–11. Note that the colour-bars for different volcanic eruptions have different values, going from smaller values for Etna and Llaima eruptions to a maximum for Nabro. From these figures, it is possible to observe the temporal evolution of the SO_2 plume as a function of altitude. These plots have to be interpreted carefully and studied together with the maps of the amount and altitude (values and errors) because here the retrieved errors in altitude are not accounted for, and for low amounts of SO_2, error in altitude can be significant. Within the IASI spectra there is enough information to retrieve altitude from small/medium eruptions such as Etna and Nyamuragira/Nyiragongo.

The tropopause heights have been computed from ECMWF temperature and pressure profiles, using the lowest layer with a lapse rate below $2\,K\,km^{-1}$ and lapse rate of $2\,K\,km^{-1}$ in all layers within 2 km above this, for every pixel of the volcanic plume. The three tropopause lines, shown in Figs. 8–11, are the mean, and the mean plus or minus the standard deviations, within the IASI plume pixels in the 12 h maps. The reported values of the tropopause are computed using the location of the volcanic pixels only. Ejyafjallajokull 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_2 being sometimes detected or not from one day to the next (coupled to the wide range of latitudes spanned by the plumes). Within the plume, the tropopause heights can differ by many kilometres especially for plumes that cover a wide latitude range. As an example, the Kasatochi plume has been analysed between 30 and 90° N. Over this range of latitudes the tropopause height varies between 8 and 18 km. Given this caveat the lines of tropopause mean and standard deviation heights are indicative, e.g. a plume that is below the three lines is likely, but not necessary, confined to the troposphere, a plume that is above the three lines is likely confined in the stratosphere, and SO_2 that is between the lines could be in either the troposphere or stratosphere.

In the following Sects. 5.1–5.5 we describe the eruptions in Figs. 8–11 divided by geographic area. A summary of these comments together with other relevant literature and volcanic explosivity index (VEI) from Smithsonian is given in Table 1.

Three principal factors affect plume height: (i) the energetics of the eruption, (ii) the dynamic effect, (iii) retrieval artefacts in the case of a multilayer plumes. Here we attempt to discriminate between these factors as follows:

a. The energetics of the eruption. A crude parameter for eruption intensity and plume height is the "volcanic explosivity index" or VEI. 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. We reported this, when availabe, in Table 1.

b. The ~~dynamic~~ atmospheric effect. In the following sections we group the eruptions by geographic area in order to consider together eruptions that may have similar conditions of water vapour. Moreover we consider the tropopause altitude together with the plume altitude.

c. Retrieval artefact in the case of ~~a~~ multilayer plumes. We do not have a way to identify this in a fresh plume but we indicate (with a black triangle) when the old plume overpasses near the volcano again (with the possibility of presence of both the old overpassing plume and the new emitted plume, at two different altitudes). The vertical distribution plots in Figs. 8–11 present the studied eruptions in chronological order and indicate the presence of a new plume connected with the volcano with red triangular symbols at the bottom of the column.

6.1 Southern Chile: Llaima, Puyuhue-Cordon-Caulle and Copahue

In the Southern Hemisphere we analysed three eruptions that originated from volcanic activity in Chile. The wind direction was similar for each eruption so the SO_2 was transported to the east (towards South Africa).

Llaima (period analysed: 2–6 January 2008), is presented in Fig. 8. This is the smallest eruption in terms of SO_2 amount (with a maximum atmospheric load of 0.04 Tg) and after the first day, the plume was disconnected from the volcano.

Copahue (22–27 December 2012), in Fig. 11, ~~release~~ released a higher amount of SO_2 (up to 0.72 Tg of atmospheric load) and there was a continuous plume from the volcano, indicating that the volcano emitted SO_2 for at least seven days. Both of these eruptions remained confined to the troposphere.

The Puyehue-Cordon-Caulle eruption of 5–30 June 2011 was the most significant eruption in southern Chile since 1991. The eruption commenced with the formation of a very significant eruption plume, that was tracked around the globe, and sustained, lower-level activity that continued intermittently during the observation period. This eruption was between Llaima and Copahue in terms of SO_2 amount (maximum load of 0.13 Tg), but higher than both in terms of VEI (with a value of 5), and with a much higher altitude (Fig. 11). The first part of the eruption produced a higher plume in terms of amount and altitude, and a lower plume connected with the volcano was present intermittently for all of the period observed. The maximum amount of SO_2 loading found was 0.13 ± 0.06 Tg on 8 and 9 June. Theys et al. (2013) present the SO_2 amount from the Clarisse et al. (2012) IASI scheme, assuming an altitude of 13 km, and reported values around 0.14 Tg (and higher) for the periods 6–9 June, with a maximum on 7 June. The fresh part of the plume (over south America and the western Atlantic ocean) is found to be significantly lower using C12 (between 2 and 5 km of altitude) than the altitude of 13 km assumed by Theys et al. (2013), and the SO_2 amount for a plume at 5 km will be underestimated using the assumption of higher altitude. Due to the opposite effect, the presence of SO_2 higher then 13 km in the first two days may explain the discrepancy in the SO_2 load (Fig. 2 from Theys et al., 2013 reports higher SO_2

than C12 but assumes lower altitude than C12). This shows how important it is to have good altitude information for the SO2 retrieval as assumptions on plume altitude may lead to significant differences in SO2 total mass estimates.

6.2 Northern Pacific Ocean: Okmok, Kasatochi and Sarychev

Okmok, Kasatochi and Sarychev eruptions (Figs. 8 and 9) all affected the boreal atmosphere and injected SO_2 to altitudes around the tropopause (and higher).

Okmok (12–20 July 2008) injected SO_2 around 12 km and the plume spread south and east from the volcano, over the Pacific Ocean. The main plume reached the US and Canada, and there was an intermittent presence of a small and low plume connected with the volcano.

Kasatochi (7–22 August 2008) injected the majority amount of SO_2 within the first three days and the plume spread around the Northern Hemisphere, going east, north and west from the volcano. The SO_2 amount reached a maximum seven days after the start of eruption, indicating continuous injection from the volcano. Within the first ten days of the eruption it affected the latitude bands 30 to 90° N. The SO_2 amounts for the first days of the eruption are in the ranges reported from Corradini et al. (2010) for AIRS and MODIS estimate. (Corradini et al., 2010 report AIRS and MODIS 7.3 µm SO_2 mass loadings between 0.3 and 1.2 Tg, while the MODIS ash corrected 8.7 µm SO_2 masses vary between 0.4 and 2.7 Tg). The altitude retrieved using C12 for Kasatochi is consistent with 12.5 ± 4 km reported by Karagulian et al. (2010), but the total SO_2 load is around 30 % lower. Karagulian et al. (2010) use an IASI retrieval from ν_3 band (1362 cm^{-1}) and $\nu_1 + \nu_3$ band (2500 cm^{-1}). The altitudes from C12 are also comparable with the range reported in CALIPSO and OMI data from Wang et al. (2013) (Wang et al., 2013 used the OMI retrieval of altitude and amount and GEOS-Chem models to estimate that the forcing by Kasatochi volcanic sulphate aerosol became negligible six months after the eruption).

Sarychev is located at a similar latitude ~~to~~ as Kasatochi, but in the Kurile islands south of Kamchatka. The IASI SO_2 scheme, C12, (similar to that previously reported by the Université Libre de Bruxelles, IASI-ULB, near real-time SO_2 alert system, http://cpm-ws4.ulb.ac.

be/Alerts/index.php, Rix et al., 2009; Haywood et al., 2010), retrieves a small tropospheric plume in the two images from 11 June 2009, followed by a higher plume on 12 June. The SO_2 loading increased in the following days (with a big injection the 15 June 2009) reaching a maximum on 16 June (0.6 Tg). Then the SO_2 load remained approximately constant for 16–18 June, before decrasig after that. The plume went in two directions, one branch spreading across the Pacific Ocean to North America and crossing Canada, reaching the Atlantic Ocean on 22 June and the Spanish coast of Europe on 24 June; the second branch went north, crossing Siberia up to the Siberian Sea and turning east to Greenland.

6.3 Africa: Dalafilla, Nabro

Two volcanic eruptions from the the Ethiopian Rift, Dalafilla in November 2008 and Nabro in June 2011, had a lower plume around 4–6 km and a higher part of the plume around the tropopause, but they were nearly one order of magnitude different in terms of SO_2 amount. Dalafilla produced a maximum SO_2 atmospheric loading of 0.2 Tg while Nabro produced 1.6 Tg. This could be the result of volcanic effects (for example Dalafilla was a very short-lived and vigorous fire-fountaining eruption from an extended fissure that produced, in some parts low mass eruption rates and in other parts much higher eruption rates. Another possible explanation of how a medium/small eruption such as Dalafilla (VEI 3) reached the tropopause altitude is illustrated in Tupper et al. (2009) and ~~implicate~~ implicated the effect of a moist atmosphere. Tupper et al. (2009) showed that volcanic emissions with different eruption rates in moist atmosphere both reached the tropopause; in dry atmosphere they reach different altitudes.

The first IASI observation showing the Dalafilla eruption was at 05:00 UTC on 4 November 2008. The plume is divided into a lower and a higher part. These spread north and east from Ethiopia, covering the Arabic peninsula, and arrived over north India and the Himalayas on the 5 November. They spread then over China and a diluted plume arrived over south Japan on 6 November. IASI detected SO_2 near the volcano in every image in this period. According to Meteosat-9 false color RGB images, the eruption started between

12:45 UTC and 13:00 UTC on 3 November 2008, with high-level SO_2 plumes and an ash plume (mixed with ice). The low-level SO_2 plume, started three hours later[2].

The IASI images of SO_2 amount and altitude are consistent with the measurements over northern India reported by Mallik et al. (2013). In particular Mallik et al. (2013) report: (i) high concentration of column SO_2 from ground measurements where the time of exceptionally high SO_2 amount is consistent with the IASI plume arriving over the Indian location; (ii) CALIPSO observations of a scattering feature around 4–5 km altitude on 6 November that is consistent with the lower part of the plume reported here by IASI; (iii) OMI maps, obtained assuming a fixed altitude, with a position of the SO_2 plume similar to IASI. With the use of C12 IASI scheme it is possible to discern that there is both a lower plume in the troposphere (as reported by Mallik et al., 2013) and a significant part of the plume at altitudes around the tropopause.

Nabro produced the largest amount of SO_2 in any volcanic plume observed by IASI with a maximum of up to $1.6 \pm 0.3\,\mathrm{Tg}$ of SO_2.

Numerous previouse studies have documented this eruption, with many of these focusing on whether Nabro inject SO_2 directly into the stratosphere or whether the injection was associated with monsoon circulation (Bourassa et al., 2012, 2013; Sawamura et al., 2012; Vernier et al., 2013; Fromm et al., 2013, 2014; Clarisse et al., 2014; Biondi et al., 2015). A more detailed study of the Nabro Eruption, also using the C12 IASI retrieval scheme, is reported in Fromm et al. (2014), and concluded that “Nabro injected sulphur directly to or above the tropopause upon the initial eruption on 12/13 June, and again on 16 June 2011”. Here we include the Nabro summary of the C12 IASI dataset. The Nabro plume is retrieved on 13 June, with a plume over north-east Africa between 14 and 18 km height. On the 14 the plume arrived over the Middle East and over central Asia on 15 June. The eruption formed two plumes at different altitudes, the higher one that reached the stratosphere and a lower one that remained confined to the troposphere with less then 10 km altitude. The higher plume is further separated into two segments, a “north” one (15 km and above) and a “south” one, a bit lower. Over all these days the plume was still attached to the volcano,

[2]http://www.eumetsat.int/website/home/Images/ImageLibrary/DAT_IL_08_11_03_A.html

indicating continuous injection. On 17 June there is a lower altitude plume and a new high altitude part going over north-east Africa.

In Fromm et al. (2014) two comparisons have demonstrated the consistency of IASI altitude with other measurements: (1) morning and afternoon IASI data of the 14 June are compared with the lidar ground data at Sede Boker (Israel) and an SO_2 profile from MLS (Fromm et al., 2014; Fig. 8); (2) the night time IASI measurements of 17 June are compared with the ~~Caliop~~ CALIOP and MLS measurements (Fromm et al., 2014; Fig. 9).

6.4 Iceland: Eyjafjallajökull and Grímsvötn

Eruptions of Eyjafjallajökull in April and May 2010 and Grímsvötn in May 2011 have been examined. A deeper analysis of the IASI SO_2 plume from the Eyjafjallajökull eruption is presented in Carboni et al. (2012) and here we only report the time series of vertical distribution.

The first IASI observation of the Eyjafjallajökull plume is in the evening of 14 April 2010. The SO_2 plume altitudes retrieved in successive observations up to 20 April are below 10 km, mainly confined below 5 km. Small amounts of SO_2 between 16 and 20 April are between 5 and 10 km in altitude. From 20 to 30 April SO_2 is always below 5 km. There is a little increase of SO_2 amount and altitude on the 1 and 2 May, and a more pronounced increase of both SO_2 amount and altitude from the 5 May. The retrieved altitude has a maximum during the 14–17 May period, where values reach around 10 km.

Grímsvötn's plume was first observed by IASI on the morning of 22 May 2011. From the afternoon of the 22 it is possible to see an SO_2 rich plume going north, with a segment going north-east, and a lower plume going south/south-east. The denser part of the higher SO_2 plume moved west and arrived over Greenland, and out of the analysed area on 24 May. This segment re-entered the analysed area on 26 May in the afternoon (this is why the last column of the plot has an apparent increase in SO_2). The lower altitude plume, moving in south/south-east direction, is reported to be more ash rich (Moxnes et al., 2014) and travelled toward Europe, arriving over the northern UK on the 24 in the morning, and over Scandinavia in the afternoon. These observations of the Grímsvötn eruption do not completely represent the total atmospheric SO_2 due to the fact that a part of the plume is missing.

6.4.1 ~~Minor frequent events: Etna and Congo~~

6.5 Minor frequent events: Etna and Congo

The activity of Etna and Nyamuragira/Nyiragongo during the period was limited to smaller eruptions and lava-fountains. Here we do not report an exhaustive list of these episodes, but a few examples to show how these emissions can be observed by IASI. The Smithsonian Institution reported that Nyamuragira erupted on 2 January 2010 and 6 November 2011. From IASI data it is not possible to distinguish between Nyamuragira and Nyriagongo emissions, but SO_2 plumes are observed from 3 to 12 January 2010 and from 7 to 15 November 2011. The SO_2 plumes spread over the equatorial area of Africa from Congo up to Sudan and Chad, with decreasing SO_2 loading away from the volcano. The main plumes were confined to the troposphere and disappeared after a few days.

The Etna events considered are the 23–25 November 2007 eruption and the 2011 lava-fountains: 11–13 January, 17–19 February, 9–12 April, 11–13 May, 8–10 July, 18–20 July, 24–26 July, 29 July, 1 August, 4–7 August, 11–13 August, 19–21 August, 28–30 August, 7–10 September, 18–20 September, 27–30 September, 12–17 November. Analysis was performed for the lava fountain period plus one day either side. The Etna eruption in 2007 produced the highest SO_2 atmospheric loading for the period analysed (more then 0.1 Tg). The lava-fountains are associated with smaller amounts of SO_2 and with large error. Etna emissions are approximately ten times smaller then those from the Congo. In particular, the SO_2 for 2011 Etna fountains have been validated within the SAMSH-SACS2 project and the SO_2 amount measured by IASI are consistent with ground observation (Spinetti et al., 2014).

7 Conclusions

In this paper, the IASI SO_2 volcanic plume altitudes and amount from a recently developed SO_2 retrieval algorithm (Carboni et al., 2012) are presented. IASI has significant advantages for the monitoring of extended volcanic eruptions, because it can be used to track the intensity of the eruption, both in terms of gas amount and gas plume height.

Comparisons of IASI altitudes against CALIPSO profiles and IASI SO_2 column amounts against Brewer ground measurements have been performed. These show that IASI retrieved values are consistent with the satellite and ground measurements. Despite the ability of IASI to retrieve the plume altitude, it could be difficult to distinguish, with only IASI data, if the eruption injects into the stratosphere or into the high troposphere. Further work integrating and comparing IASI with other measurements, such as lidar or limb measurements (e.g. Michelson Interferometer for Passive Atmospheric Sounding ~~(MIPAS)measurements~~ MIPAS), are needed to better assess the ability of IASI to determine injection into the stratosphere.

The IASI scheme has been used to follow the vertical distribution of SO_2 as a function of time (twice daily), for different eruption types (e.g. VEI ranging between 1 and 5) and different latitudes. This work demonstrates that while VEI is a convenient and rapidly estimated proxy for eruption “scale”, it is a poor index of the potential height to which volcanic SO_2 is injected.

There is a tendency for volcanic SO_2 plumes to reach a point of buoyancy near the tropopause. All of the eruptions in the tropics (except Nyamuragira), reached the tropopause. In the mid latitudes, the eruptions of Eyjafjallajökull, Llaima, Copahue and Etna remained confined in the troposphere.

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

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Table 1. Summary of studied eruptions, in chronological order, together with other relevant literature, volcanic explosivity index (VEI) and a short descriptions of the events from the Smithsonian Institution Global Volcanism Programme website (http://www.volcano.si.edu).

Volcano name	Analysed dates	Lat/Lon/ Elevation	Comments on IASI SO_2 plume	References to other work	Volcanic event description (Smithsonian weekly reports)
Llaima (Chile)	2–6 Jan 2008	38.692° S 71.729° W 3125 m	Small eruption (maximum of 0.04 Tg of SO_2). Plume connected to the volcano only on the first day. Confined in troposphere.	McCormick et al. (2013): OMI SO_2 study.	VEI 3. Strombolian eruption began 1 Jan; ash plumes to 12.5 km on 2 Jan, and to 3.7 km on 3 Jan.
Okmok (Aleutian islands)	12–20 Jul 2008	53.43° N 168.13° W 1073 m	SO_2 injected around tropopause and the plume spreads south (pacific ocean) and east (north America) from the volcano. Presence of intermittent smaller and lower plume connected with the volcano. Main plume altitude increases with time following the tropopause altitude.	Spinei et al. (2010): OMI SO_2 validation against ground measurements over north America; Prata et al. (2010): SO_2 and ash from AIRS.	VEI 4. Strong explosive eruption began 12 Jul, with ash to at least 15 km. Eruption intensity declined, with ash plumes to 6 km on 17 Jul. Eruptions on 19 Jul sent ash to 9 km.
Kasatochi (Aleutian islands)	7–22 Aug 2008	52.177° N 175.508° W 314 m	SO_2 amount reached the maximum after seven days indicating continuous injection from volcano. Within ten days SO_2 plume affected all latitudes between 30–90° N. Reached to tropopause and stratosphere.	Krotkov et al. (2010): Dispersion and lifetime of the SO_2 using OMI; Yang et al. (2010): OMI retrieval of SO_2 amount and altitude; Karagulian et al. (2010): SO_2 and ash from IASI; Corradini et al. (2010): Comparison of different IR instruments (AIRS, MODIS); Kristiansen et al. (2010): inverse transport modelling; Bitar et al. (2010): lidar measurements, injected material into the troposphere and lower stratosphere; Prata et al. (2010): SO_2 and ash from AIRS; Nowlan et al. (2011): GOME-2 optimal estimation retrieval of amount and altitude; Wang et al. (2013): Volcanic sulfate forcing uning OMI SO_2.	VEI 4. Explosive eruption began on 7 Aug, with ash plumes to 14 km; continuous eruptions from 8–9 Aug, with ash plumes to 9–14 km. Subsequent observations obscured by cloud.
Alu-Dalaffilla (Ethiopia)	4–7 Nov 2008	13.792° N 40.55° E 613 m	Plume is divided in two parts from the beginning (one lower in troposphere and one higher into tropopause /stratosphere). SO_2 near the volcano in every image (continuous emission). Nearly one order of magnitude smaller than Nabro (in amount of SO_2) but reached comparable height.	Pagli et al. (2012): Ethiopian Rift deformation paper, studying the source of Alu-Dalafilla Eruption. Mallik et al. (2013): Enhanced SO_2 concentrations observed over northern India in Nov 2008.	VEI 3 – Mainly effusive eruption. Strong fissure eruption began on 3 Nov, for a few hours, waned rapidly and ended on 6 Nov. (Pagli et al., 2012). No direct observations of the eruption.

Table 1. Continued.

Volcano name	Analysed dates	Lat/Lon/ Elevation	Comments on IASI SO_2 plume	References to other work	Volcanic event description (Smithsonian weekly reports)
Sarychev Peak (Kuril islands)	11–26 Jun 2009	48.092° N 153.2° E 1496 m	Started with a small tropospheric plume building up with increasing SO_2 load, maximum on 16 Jun (0.6 Tg). Reached tropopause and stratosphere.	Theys et al. (2009): Volcanic BrO from GOME-2. Haywood et al. (2010): Climate model (HadGEM2), IASI SO_2, OSIRIS limb sounder and CALIPSO lidar measurements to investigate the distributions of SO_2 and radiative impact of sulphate aerosol; Carn and Lopez (2011): validation of OMI SO_2; Doeringer et al. (2012): sulphate aerosols and SO_2 from the Atmospheric Chemistry Experiment (ACE) measurements; Fromm et al. (2014); on correction for OSIRIS dataset and using SO_2 from C12.	VEI 4. Eruption began 11 Jun, with ash plumes to 7.5 km on 12 Jun, and a strong ash plume to 12 km on 14 Jun. Further explosions with ash to 10–14 km continued through 18 Jun.
Soufrière Hills (Montserrat)	10–15 Feb 2010	16.72° N 62.18° W 915 m	When it was first observed the SO_2 plume is divided into two parts: a higher one around 16–19 km (tropopause/stratosphere) and a lower one in the troposphere. The lower plume disappeared (non detectable) after one day, while the higher one spread east, south east.	Prata et al. (2007) on 2006 eruption. Carn and Prata (2010): satellite measurements for 1995–2009; Christopher et al. (2010) include SO_2 measurments for 1995–2009. Wadge 2010: Lava production for the period 1995–2009. Christopher et al. (2014): SO_2 degassing, for 1995– 2013, and relation with magma supply; Nicholson et al. (2013), include daily mean SO_2 flux.	VEI 3. Large dome collapse on 11 Feb, with pyroclastic flows and an ash plume to 15 km.
Eyjafjallajökull (Iceland)	14 Apr–24 May 2010	63.63° N 19.62° W 1666 m	SO_2 plume confined in troposphere. Up to 20 Apr the plume was below 10 km (mainly confined within 5 km); From 20 to 30 Apr plume < 5 km. Increase in both (amount and altitude) from 5 May, with maximum altitude (around 10 km) from 14 to 17 May.	Stohl et al. (2011): ash emission height from inversion with dispersion model FLEXPART. Marzano et al. (2011): radar measurements of plume altitude. Stevenson et al. (2012): tephra deposit. Petersen et al. (2012): impact in atmophere and plume altitude. Carboni et al. (2012): IASI SO_2. Boichu et al. (2013): chemistry-transport model + IASI SO_2 to invert SO_2 flux.	VEI 4. Explosive eruptions from the summit, with ash plumes from 3–10 km (Gudmundsson et al., 2012).
Merapi (Java, Indonesia)	4–11 Nov 2010	7.542° S 110.442° E 2968 m	High plume up to tropopause and stratosphere. (Problems: old plume overpassing the volcano).	Surono et al. (2012): estimate a total of 0.44 Tg of SO_2, using IASI -AIRS assuming a plume altitude of 16 km. Saepuloh et al. (2013); Picquout et al. (2013); Pallister et al. (2013); Luehr et al. (2013); Jenkins et al. (2013); Innocenti et al. (2013b, a); Cronin et al. (2013); Costa et al. (2013); Budi-Santoso et al. (2013)	VEI 4. Major explosive eruption, with a climactic phase that began on 3 Nov with ash plumes to 18 km. Strong explosions continued through 5–6 Nov (17 km ash plumes), declining to < 8 km by 12 Nov.

Table 1. Continued.

Volcano name	Analysed dates	Lat/Lon/ Elevation	Comments on IASI SO_2 plume	References to other work	Volcanic event description (Smithsonian weekly reports)
Etna (Sicily, Italy)	23–25 Nov 2007 + 2011 fountains	37.734° N 15.004° E 3330 m	All episodes considered are confined to the troposphere.	Aiuppa et al. (2010); combine seismic (volcanic tremor and long-period seismicity), deformation (GPS), and geochemical (volcanic gas plume CO_2/SO_2 ratios) measurements to interpret trends in the recent (2007–2008) activity of Etna volcano. Patane et al. (2013); study the 2011 Etna episodes using multi-parameter approach (sesmicity, ground deformations and geochemistry, SO_2 flux, CO_2 flux, CO_2/SO_2 ratio). Viccaro et al. (2014); Behncke et al. (2014); Birth, growth and products of the new SE crater. Scollo et al. (2014); Eruption column height estimation (using calibrated TIR camera) of the 2011–2013 Etna lava fountains. Guerrieri et al. (2015): ash and SO_2, retrieval and flux estimate, from SEVIRI. Spinetti et al. (2014), satellite SO_2 validation with ground measurements (including this IASI SO_2 scheme).	
Nyamuragira/ Nyiragongo (DR Congo)	3–12 Jan 2010 and 7–15 Nov 2011	1.408° S 29.2° E 3058 m/ 1.52° S 29.25° E 3470 m	Confined to the troposphere.	Carn and Bluth (2003); Nyamuragira SO_2 from TOMS period: 1978–2002. Bluth and Carn (2008); Exceptional sulphur degassing from Nyamuragira volcano using TOMS, 1979–2005. Wadge and Burt (2011). Carn et al. (2013): monitored Nyiragongo volcano degassing with OMI, including monthly means SO_2 for 2004–2008. Cuoco et al. (2013): impact of Nyamuragira. Campion (2014): studied Nyamuragira emissions of SO_2 using OMI and ASTER. Smets et al. (2014): multidisciplinary monitoring reveals pre- and co-eruptive signals at Nyamuragira volcano associated with the Jan 2010 volcanic eruption (seismic and OMI data).	Jan 2010, VEI 1. (Nyamuragira). A flank fissure eruption began on 2 Jan 2010, and continued intermittently for 3–4 weeks. Nov 2011, VEI 2. (Nyamuragira): major flank fissure eruption began with fire-fountaining and the emplacement of a long lava flow on 6 Nov 2011. The eruption is thought to have ceased by 18 Nov. During both Jan 2010 and Nov 2011, the lava lake at Nyiragongo remained active (based on thermal alerts).

Table 1. Continued.

Volcano name	Analysed dates	Lat/Lon/ Elevation	Comments on IASI SO_2 plume	References to other work	Volcanic event description (Smithsonian weekly reports)
Grímsvötn	21–26 May 2011	64.42° N 17.33° W 1725 m	Higher part of the plume moved west and spread north reaching the tropopause/stratosphere. Lower plume travelled towards Europe (together with ash), confined to the troposphere.	Flemming and Inness (2013): Assimilation of UV satellite measurements for forecast. Moxnes et al. (2014): FLEXPART + IASI SO_2 and ash. Koukouli et al. (2014): Intercomparison of SO_2 from IASI (including C12), GOME-2 and ground data.	VEI 4. Strong explosive eruption, with an ash plume to 20 km on 21 May, declining to 10–15 km on the morning of 22 May. Minor activity on 24 May, with ash to 5–7 km.
Puyehue-Cordon-Caulle	5–30 Jun 2011	40.59° S 72.117° W 2236 m	Intermediate magnitude (between Llaima and Copahue) Reached the tropopause/stratosphere and we can follow the plume going 3 times around the Southern Hemisphere in 30 days. First part of the eruption is higher in SO_2 amount and altitude. Intermittent low plume connected with the volcano for all the period considered.	Theys et al. (2014): detection of volcanic OClO. Bignami et al. (2014): SAR deformation, MODIS ash and SO_2.	VEI 5. Major sequence of explosive and effusive eruptions, that began on 4 Jun with ash plumes to 11–14 km. For the rest of Jun, lower-level eruptive activity persisted, with intermittent ash plumes to 5–8 km. The eruption continued until Apr 2012.
Nabro	12–23 Jun 2011	13.37° N 41.7° E 2218 m	The highest emission of SO_2 for the period considered (2008–2012). Two plumes at different altitudes, the highest one reached the to stratosphere, the lower remained confined to the troposphere.	Fromm et al. (2014): Nabro injected sulphur directly to or above the tropopause upon the initial eruption on 12/13 Jun, and again on 16 Jun 2011. Bourassa et al. (2012); Sawamura et al. (2012); Vernier et al. (2013); Fromm et al. (2013); Bourassa et al. (2013); Clarisse et al. (2014); Biondi et al. (2015).	VEI 4. The eruption began with a major ash plume, to 9–14 km on 12 Jun. The activity continue dominated by lava flow, and with ash plumes to 6–8 km from 15–19 Jun.
Copahue	22–27 Dec 2012	37.856° S 71.183° W 2953 m	Maximum of 0.72 Tg of SO_2 present in atmosphere. Plume connected with volcano every day. Confined to the troposphere.	Velez et al. (2011): ground deformation using InSAR; Bonali (2013): earthquake and stress study.	VEI 2. Moderate explosive eruptions, with a mixture of Strombolian fire-fountaining and steam-driven phreatic explosions, producing frequent low-level ash plumes rising 1–2 km above the crater rim.

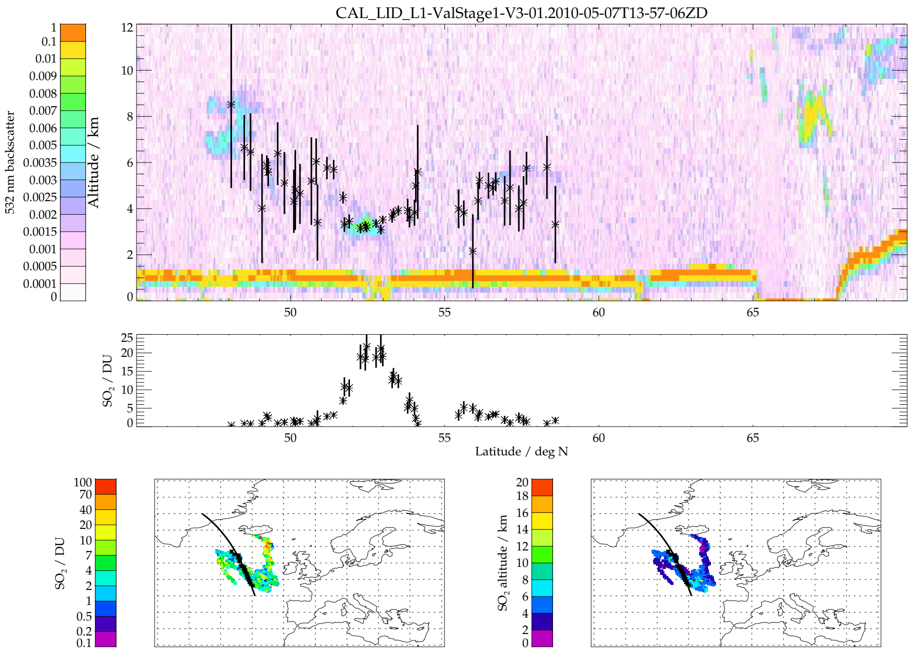

Figure 1. CALIOP/ IASI coincidences for the Eyjafjallajökull plume on 7 May 2010 overpasses within 2 h off each other. Top plot: CALIOP backscatter profile with IASI over-plotted retrieval altitude (black stars) and error-bar (black line); middle plot: the IASI SO_2 amount and error-bars corresponding to the altitude plotted above; bottom plots: map of IASI SO_2 amount (left) and altitude (right) with CALIPSO track (black line) and identifying the IASI pixel plotted above with black stars.

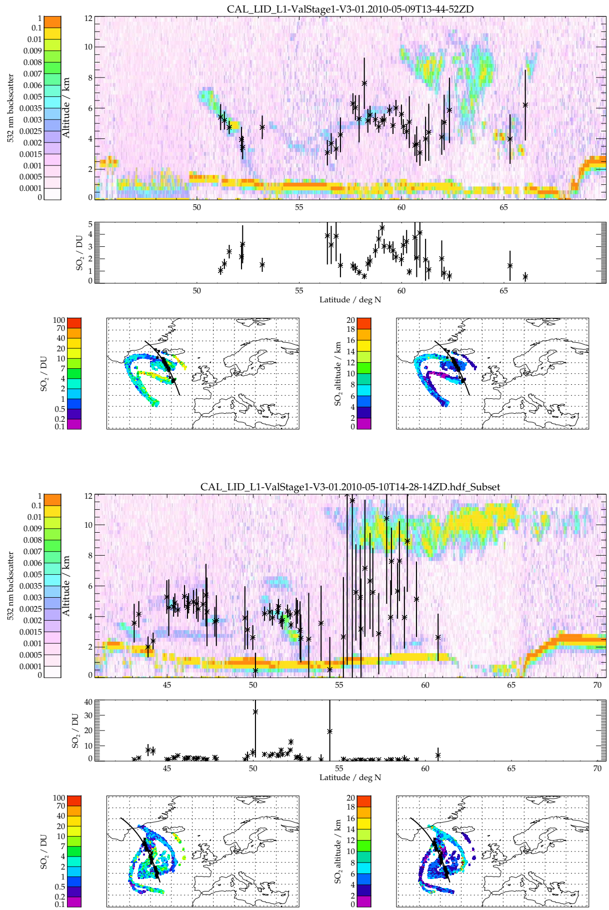

Figure 2. CALIOP-IASI coincidences for the 9 and 10 May 2010, Eyjafjallajökull plume. As Fig. 1.

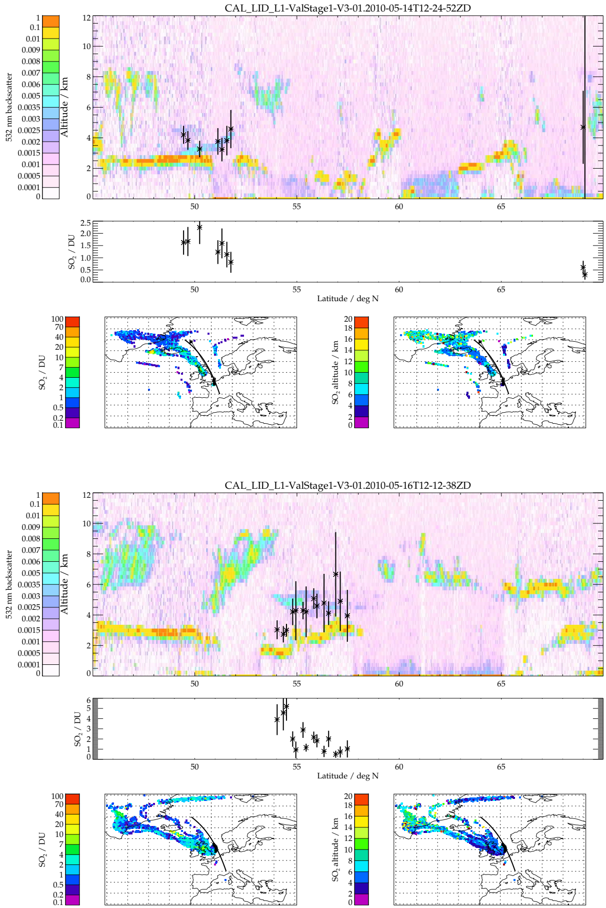

Figure 3. CALIOP-IASI coincidences for the 14 and 16 May 2010, Eyjafjallajökull plume. As Fig. 1.

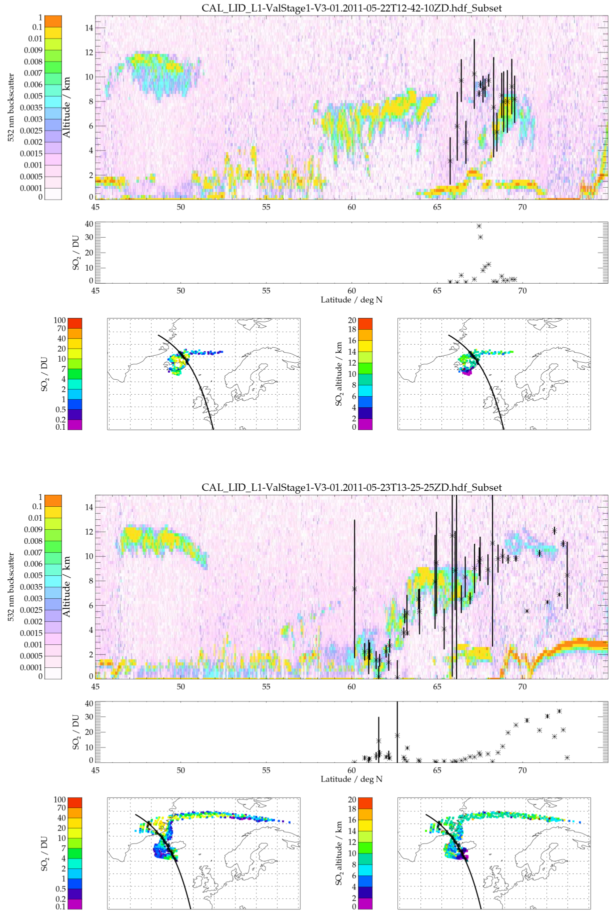

Figure 4. CALIOP/IASI coincidences for the Grímsvötn eruption on 22 and 23 May 2011. As Fig. 1.

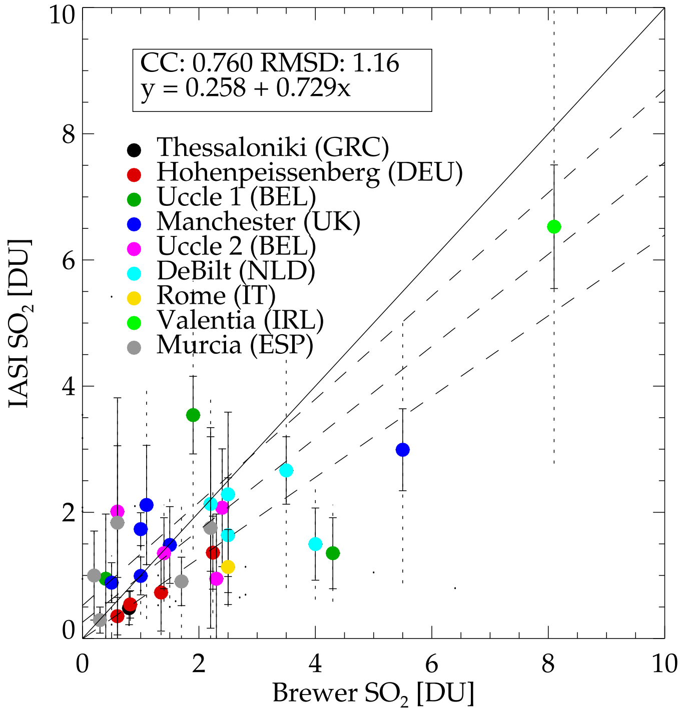

Figure 5. Scatter plot of IASI SO_2 measurements, averaged within a distance of 200 km from the ground station, vs. the daily SO_2 column amount, measured from Brewer spectrometers. Different colours correspond to ~~a~~ different Brewer ground ~~station~~stations. Black error-bars are the IASI average errors; dotted error-bars are the standard deviation of the IASI data within the selected distance. Black lines represent the ideal line $y = x$; dotted lines are the best fits with error in the best fit. The legend box shows the correlation coefficient (CC), root mean square difference (RMSD) and the best fit line.

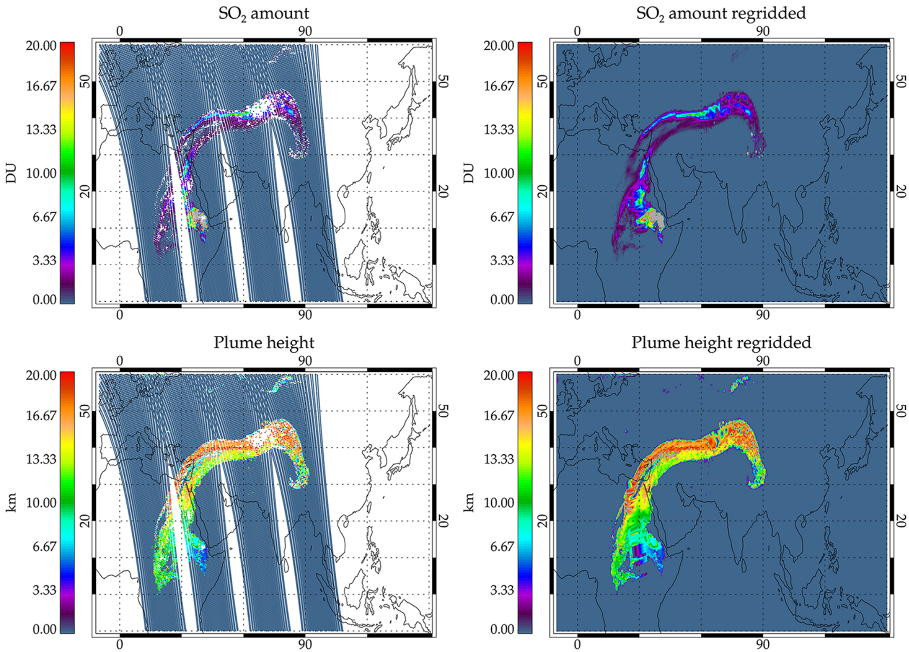

Figure 6. Maps of IASI SO_2 amount (top left) and height (bottom left) and the equivalent maps of SO_2 amount (top right) and height (bottom right) obtained after regridding. Grey colour indicate values higher than the colour bar. The IASI data are a combination of four orbits on the 15 June 2011 from 13:00 to 18:00 UTC.

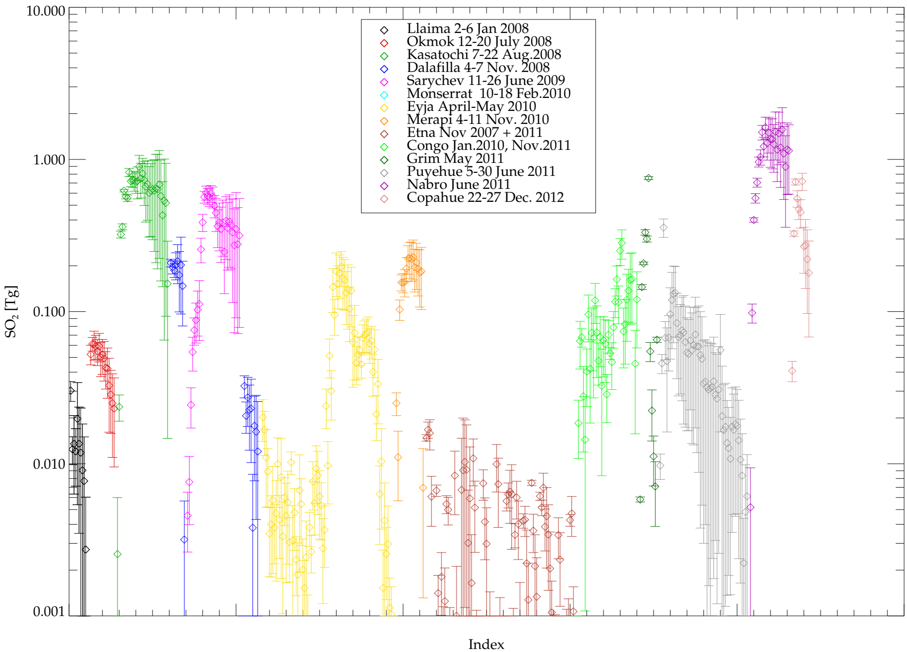

Figure 7. SO_2 mass present in the atmosphere as retrieved from IASI data. The values are the measured amount every half day and vary with volcanic emission, gas removal and satellite sampling. Points are separated by $\sim 12\,\mathrm{h}$. Data are presented in temporal order along the ordinate (x axis) but eruptions are plotted consecutively one after the other without a gap between them. The total SO_2 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_2 loads within each respective area are considered and plotted separately.

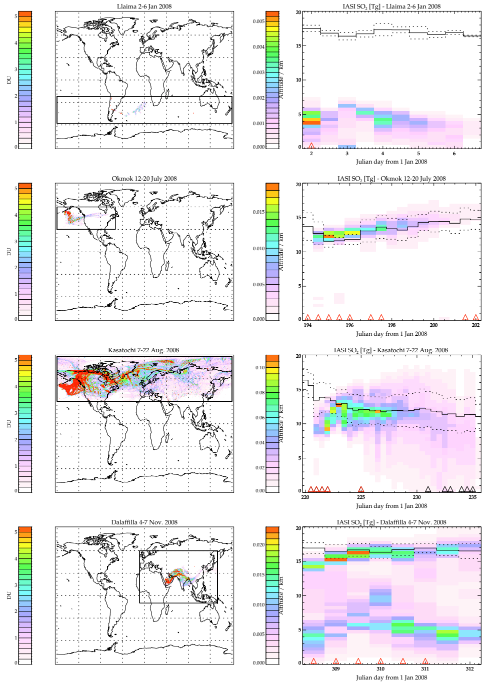

Figure 8. The left column shows the plots of the maximum SO_2 amount retrieved within the considered area (black rectangle). The right column shows the SO_2 vertical distribution for the considered volcanic eruption. In each plot the y axes are the vertical levels in km. The colour represents the total mass of SO_2 in Tg, dark-red represents values higher than the colour-bar. Every column of the plots come from an IASI map (one every 12 h). The black lines are the mean, and the mean plus or minus the standard deviations, of tropopause computed from ECMWF profiles at the location of plume pixels. Red triangles in the bottom line indicate the presence of a fresh plume connected with the volcano, black triangles indicate the presence of an old plume overpassing the volcano (this may eventually mask a newer plume). Note that the plots for different eruptions have different colour-scales and cover different time ranges.

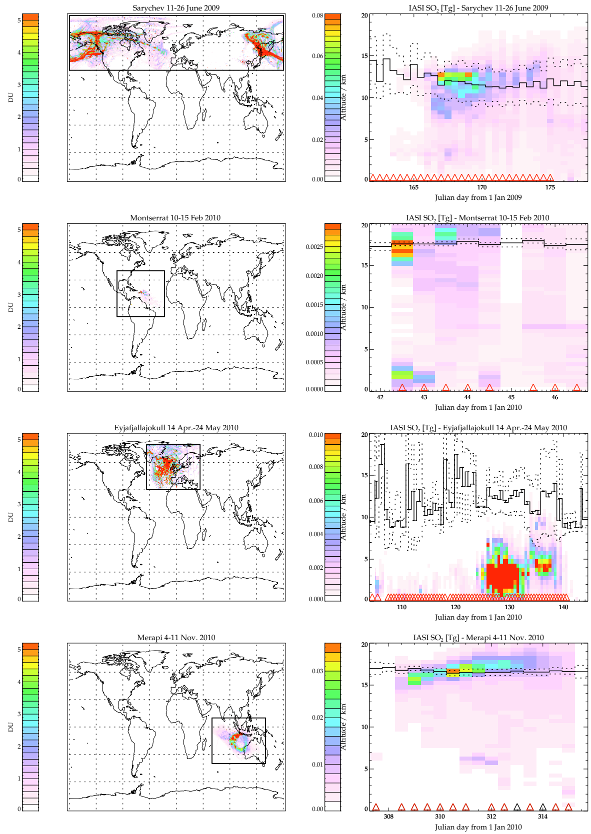

Figure 9. As for Fig. 8.

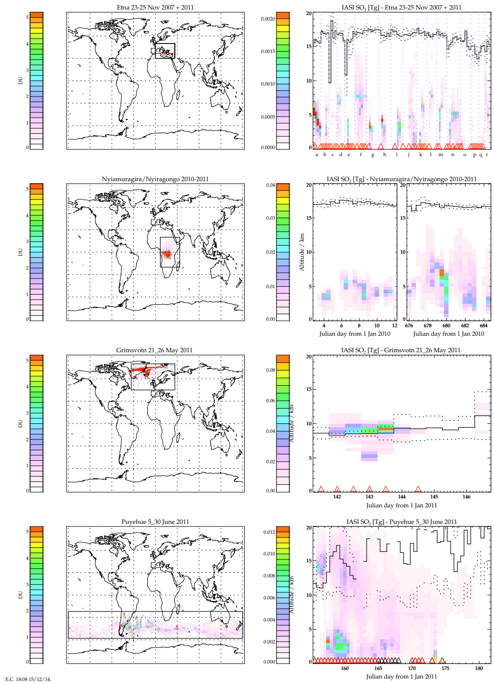

Figure 10. As for Fig. 8. The Etna plots ~~shows~~ show different eruptive episodes corresponding to x axes labels: (a) 23–25 November 2007 eruption; and the 2011 lava-fountains: (b) 11–13 January, (c) 17–19 February, (d) 10–12 April, (e) 11–13 May, (f) 8–10 July, (g) 18–20 July, (h) 24–26 July, (i) 29 July–1 August, (j) 4–7 August, (k) 11–13 August, (l) 19–21 August, (m) 28–30 August, (n) 7–10 September, (o) 18–20 September, (p) 28–30 September, (q) 11–14 October, (r) 12–17 November.

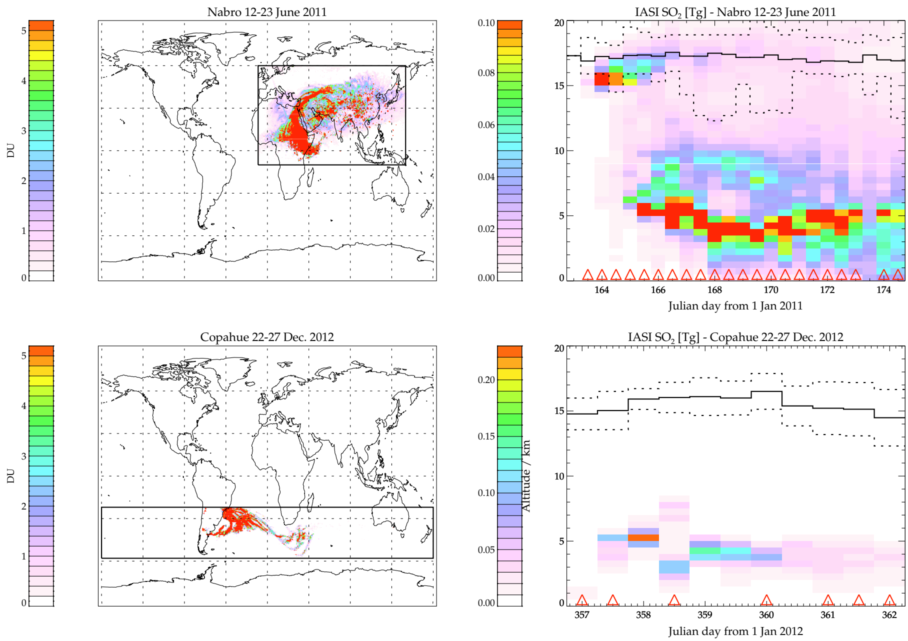

Figure 11. As for Fig. 8.