Interactive comment on "Volcanic SO_2 fluxes derived from satellite data: a survey using OMI, GOME-2, IASI and MODIS" by N. Theys et al.

Nicolas Theys: Author response to referees comments

The authors gratefully thank all the reviewers for their thorough review and interesting comments which contributed to improve the manuscript.
Replies to the reviewers comments are below in italic; changes in the text are in blue in the revised version of the manuscript.

Review of anonymous Referee #2
(comments received and published: 6 February 2013)

Since this paper emphasized that these techniques mainly worked for 'dispersed and large-scale' plumes, the authors probably need to include the area sizes (in square kilometer) for the sample cases shown in this paper, and discuss how they are selected and if automatic processing is possible.

Reply: *We propose to use '*plumes of SO2' *instead of '*dispersed and large-scale plumes of SO2 '. *The techniques used in this study work best for dispersed plumes (especially for the traverse and inverse modeling techniques) but at least one method (delta-M) is also applicable to low-wind conditions. The size of the plume is also not really an issue. E.g., the traverse method has been successfully applied to ASTER observations of small-scale tropospheric SO2 plumes (Campion et al., 2012). The maximum plume area for the three events studied is of about 2, 8 and 25 Million km² for Nyiamuragira, Puyehue and Nabro, respectively. As the plume size is not critical and for the sake of conciseness , we prefer not to add this information in the text. The referee asked about the possibility of automatic processing: it is non-trivial to have automatized and accurate calculation of fluxes. For plumes with a simple geometry and single injection height, several techniques (delta-M, traverse/box estimates) could be implemented relatively easily using operational meteorological data fields.*

Also this paper needs to include the description of the SO2 mass (or the total burden) calculation for a plume, whether a threshold SO2 value is used to select the pixels or all the pixels inside a bounding box were included. How to treat the background SO2 pixels may have a significant impact on the total burden, hence the derived flux, especially when a plume is well dispersed.

Reply: *In section 1.2, we give information on SO2 column unit (molecules cm-2). The calculation of the SO2 mass is simply the multiplication of this quantity with the satellite pixel size (cm²). Selection of pixels for OMI and GOME-2 is done using threshold SO2 values as described in section 4.2. For IASI, a detection criterion is used on the measured brightness temperature difference (see Clarisse et al., 2012) but it should be noted that the noise on the IASI SO2 data is very small. The way the background pixels are treated has an influence on the derived flux via the estimates of SO2 total burden (but also to some extend via the derived SO2 e-folding time) that are underestimated for strongly diluted plumes. As stated in the paper, we have conducted sensitivity tests to estimate errors on the fluxes (typically*

50%). They include the uncertainties on the SO2 columns (threshold values and other retrieval uncertainties), SO2 losses and wind fields. We propose to use the following sentence in the text: "We conducted sensitivity tests by varying the different important parameters of the retrieval. Based on these sensitivity tests, a typical error of about 50% including the uncertainties on the SO2 columns, cut-off values used, SO2 losses and wind fields has been estimated."

Specific comments:
1. Figures 5, 10, and 12 are too small see without enlargement.
2. Rightmost panel of figure 12 seems to be mislabeled.

Reply: *The figures have now been improved in line with these suggestions.*

Review of A. Durant
(comments received and published: 7 February 2013)

(1) 31352 L3 The definitions of "explosive" and "effusive" eruptive activity should be strengthened. These types of activity relate to the erupted materials. Broadly speaking, effusive activity erupts lavas (molten rock flows) and explosive activity erupts pyroclast (rock fragments). Also "Plinian" is out of context as there are many other categories in explosive eruption classification. The presence of SO2 in a volcanic plume/cloud cannot be taken as a proxy for volcanic ash. Fire fountaining is a form of explosive activity, and in reality, there is a continuous spectrum between "explosive" and "effusive" activity.
I suggest the following modification:
"Volcanism is the surface expression of internal processes, driven by heat generated in the Earth's interior. During eruptions, solid, liquid and gaseous products are generated. The main driving force behind eruptions is exsolution of gas from magma during decompression which drives ascent through the Earth's crust. Volcanic activity may be classified into two main types:
Explosive activity: Rapid exsolution of volcanic gases in the volcanic conduit (vent) generates an ensemble of particles (tephra) through fragmentation that is ejected explosively into the atmosphere forming a plume. Heat is derived from the erupted tephra and emitted gases, and atmospheric air is entrained which increases buoyancy. Additional latent heat may be released as water condenses and freezes in the plume. Volcanic plume heights may reach altitudes well into the stratosphere and the maximum height depends on the mass flux rate (amount of material released as a function of time), the size distribution of erupted particles, and the local wind field.
Effusive activity: Driven by gas exsolution, although at lower rates than during magma fragmentation, molten rock products (lava flows) are erupted at the surface. While this style of activity does not result in particle generation, magmatic gases released are hot and may still produce a significant plume. Thermal energy is also available from the lava, however, these plumes tend only to reach mid-tropospheric levels except in some exceptional cases."

Reply: *We thank the reviewer for his suggestion. The first paragraph of the introduction has been adapted and includes the proposed modification.*

(2) 31362 L7 Does temperature also play a role here? Furthermore, how about the effect of particle surface area in the cloud (e.g., ash surfaces) to catalyse heterogeneous chemistry? And how about “wet” deposition (i.e., hydrometeor formation) and sequestration of gases in water e.g., ice? See Rose et al. [1995] for a good example.

Reply: *Temperature is of course playing a role on the kinetics of the chemical reactions. However, a major factor affecting the SO2 removal rate is the availability of atmospheric oxidants. The sentence has been rephrased: “A difficulty of using this assumption is that k is extremely variable, being sensitive to a number of poorly controlled and spatially variable factors such as plume altitude, cloudiness and atmospheric humidity (Eatough, 1994). A major factor is the availability of atmospheric oxidants in the gas (OH radical) and aqueous (H_2O_2) phases. Wet and dry deposition, heterogeneous chemistry and sequestration of gases in ice are other important processes influencing the SO_2 loss rate. The reader is referred to the literature (e.g., Rose et al., 1995; Chin and Jacob, 1996; Graf et al., 1997; Jacob, 1999; Chin et al., 2000; Lee, 2011;) for an overview of the physical and chemical processes of SO_2 removal. The literature on SO2 reactivity in volcanic plumes (review in Oppenheimer et al., 1998) provides rate constants that span three orders of magnitude, from 10^{-4} s^{-1} for an ash rich plume in the tropical boundary layer (Rodriguez et al., 2007) to 10^{-7} s^{-1} for a plume residing in the super-dry and cold stratosphere (Read et al., 1993). “*

(3) 31376 L9-13 Going back to the definition of effusive versus explosive activity, the gas plume that reached the tropopause is plausible; the type of eruption is defined by the presence of erupted tephra, or the absence of it. Also, the definition of a “Plinian” eruption is not simply a function of the height of the column, it is related to the mass eruption rate of tephra, and also on the size characteristics of the erupted material.
Please remove all reference to “Plinian” in the paper – it is not relevant to this study.

Reply: *done.*

(4) Please add a systematic and objective comparison of the different satellite sensors. This could include plots that show percentage differences between the sensors, and/or a table could be included that gives quantitative information on the following: 1. Spatial resolution; 2. temporal resolution; 3. sampling frequency; 4. height sensitivity of retrieval; 5. error estimate. This could also be an extension of Table 1.

Reply: *Intercomparison of SO2 columns is not straightforward as it depends on the conditions and hence varies from an eruption to another (altitude, latitude, clouds, etc..). Unless it is really needed, we would prefer not adding a new figure /table as the paper is already quite long. A table with quantitative information would not add much to the description in section 2 and the table might also be difficult to fill (e.g., a single error estimate per instrument is misleading as the retrieval uncertainties depend on many parameters). Moreover, such a table is available elsewhere (e.g., Thomas and Watson, 2010), hence we propose to refer to this paper.*

Technical corrections

31350 L8 Please add a reference to “ultraviolet-visible (UV-VIS) and thermal infrared (TIR)” techniques in this sentence. Also please specify the names of the satellites and techniques that are discussed in the paper.

Reply: *“ultraviolet” has been added. The names of the satellites appear in the abstract. We have not added the names of the techniques, as we believe it is not easy for the reader to understand them simply by their names.*

31352 L5-6 “The SO2 flux is often used as a proxy for the eruptive rate. . .” Better to write, “Changes in SO2 flux are often used as an eruption precursor . . .”. Also, an increase in SO2 flux may indicate that a shallow magma body has started to migrate upwards towards the surface. Please add this point.

Reply: *done.*

31353 L12 “.. are widely carried out from ground since . . .” change to “out from the surface. . .”

Reply: *done.*

31354 L1 “With measurements channels. . .” change to “The following instruments have measurement channels that correspond to the infrared and ultraviolet SO2 absorption bands:”

Reply: *changed to “The following instruments have been used, with measurement channels that correspond to the infrared and ultraviolet SO2 absorption bands:”*

31354 L15 Change “e.g.” to “For example,”

Reply: *done.*

31354 L19 This sentence is grammatically incorrect – please reword.

Reply: *We have now reformulated this as: “Estimating fluxes from emitted masses is notoriously difficult and implies more than applying (crude) scaling laws to the measured total SO2 masses.”*

31354 L23 Change to “. . .flux calculations require . . .”

Reply: *done.*

31355 L2 The use of “a fortiori” seems out of place. . . It is often used in the natural sciences to mean “even more likely”, for example.

Reply*: it has been removed.*

31355 L15 VAST, SMASH and SACS-2 could also be mentioned.

Reply: *we prefer not to refer to these projects.*

31355 L16-17 Please correct grammar.

Reply: *We have now reformulated this as: "Although the main objective of this study is the determination of SO2 fluxes, it also investigates the consistency of the SO2 products from the different sensors used."*

31356 L13 .". . . performs regular measurements"

Reply: *the sentence has been changed to "..perform frequent measurements".*

31357 L17 ". . . spectrograph that measures . . ."

Reply: *done.*

31359 L6 Please change "Because of . . ." to "The vertical sensitivity of SO2 measurements is affected by water vapour interference below 3-5 km height . . ."

Reply: *The original sentence was unclear, we have now reformulated this as: "The vertical sensitivity to SO_2 is affected by water vapor absorption and is limited to the atmospheric layers above 3-5 km height".*

31360 L10 Please put the relevant citation after each satellite sensor mentioned.

Reply: *done.*

31365 L1 "Satellite observations provide . . ."

Reply: *done.*

31366 L9 "related to the plume height . . ."

Reply: *done.*

31366 L12-22 This paragraph should come earlier, in the Introduction section.

Reply: *We agree and have moved this paragraph now to the introduction.*

31366 L15 ". . . injection altitude due to . . ."

Reply: *done.*

31367 L20 Better to use "t" instead of "T"? The latter is usually used to denote temperature.

Reply: *done.*

31374 L5-23 Please move to Methods Section 3. This is the results section.

Reply: *We believe this paragraph (specific to Nyiamuragira) is better placed in section 4.2 than in section 3.*

31379 L15 "Accurate simulation of the transport of SO2 was challenging in the first 15 h."

Reply: *done.*

31381 L17 -31382 L28 This entire section should be moved to a new section: "Discussion".

Reply: *this is a good suggestion. We have restructured the whole section 4.3 with a better introduction and a discussion section including the discussion on the plume transport and GOME-2 vs IASI comparison (cf. next comment).*

31383 L8-19 This is a review of methodology, and should be earlier in the methods section. It does not need to be repeated this late in the paper. Please move this to the relevant location. Also Section 4.3.3 should be moved to the new Discussion section as a subsection.

Reply: *We prefer to leave it there. In the methodology section, it would appear unrelated to the overall description.*
Section 4.3.3 is now in the new Discussion section.

31385 L13 Grammar: ". . .in that a sequence . . ."

Reply*: done.*

31385 L16 ". . . applicable to eruptions . . ."

Reply*: done.*

31385 L23 Please remove references to "Plinian". . . this terminology has been confused.

Reply*: done.*

31386 L16 "Flux estimates will also be improved and better constrained . . ."

Reply*: done.*

Figures 2, 7 Please add error bars to all the retrieved values.
Figures 4, 8, 9, 11 Please add error bars to the flux estimates.

Reply: *The present paper focus on different techniques to invert fluxes and on the comparison of results. A rigorous error calculation is difficult and out of the scope of this study. Adding error bars will also make the figures very hard to read.*

Figures 5, 6 (inset), 10, 12, 13, Please add geographic scale legend.
Figure 11 Text needs to be made larger; image quality poor.
Figure 14 Please increase text size

Reply: *in line with Referee's comments, the figures have been improved.*

Review of S. Carn
(comments received and published: 11 February 2013)

Although the paper contains a lot of useful new information, including the derived SO2 fluxes for the eruptions studied, and a comparison between IASI and GOME-2 SO2 data, my main criticism is that the paper is fundamentally a techniques paper. The authors compare results from the different techniques (many of which have been previously described elsewhere), and provide some validation of the methods, but do not use the SO2 fluxes they derive to address any real scientific questions. In the case of one of the eruptions studied (Nabro), with additional work their analysis could be used to further investigate the long-range transport of the SO2 emissions, which remains controversial.

Reply: *For large eruptions, ground-based measurements (when available) are not reliable because of saturation issues and the inability of measuring the whole plume from a close distance. Hence satellite measurements are really the only practical way to proceed. Since the volcanic SO_2 flux is a geophysical relevant property, even just reporting fluxes constitutes a contribution to science.*
However, in this paper we went one step further and used several different techniques and sensors to derive fluxes. This allows us to intercompare these different techniques and satellite retrievals. But also allows us to be more confident in the provided fluxes. To our knowledge, there has been no other study on multiple techniques to invert fluxes . Moreover, our study is important as it shows that the derived flux from the four satellite sensors are consistent even with the differences in measurement sensitivity, pixelsizes and time of observations.
For Nabro, it is true that the long-range transport of the emissions remains controversial. Although it is not directly linked with fluxes, we have proposed a mechanism to explain the differences between the model and the observations.

The choice of eruption case studies is somewhat arbitrary, with the Puyehue example being perhaps the least compelling. Although Puyehue was a silicic, ash-rich, explosive eruption (distinct from the other eruptions analyzed), the analysis does not contribute anything particularly unique relative to the other case studies. The Nyamulagira and Nabro eruptions are more logical choices since continuous emissions over a period of days or weeks are more amenable to SO2 flux calculations, and such measurements can be useful for eruption monitoring.

Reply: *the choice of eruptions was precisely meant to cover different scenarios (short- vs long-lasting eruptions, plumes altitudes ranging from the surface to tropopause levels) and test the different techniques. Regarding Puyehue, our results show that it is possible to infer high temporal resolution fluxes and in contrast to the referee's comment we believe Puyehue results are among the most convincing ones of the paper.*

However, a shortcoming of the paper is that is only discusses large eruptions, whereas SO2 fluxes are more commonly used for monitoring of 'passive' or non-eruptive volcanic plumes.

Reply: *We agree that SO_2 fluxes are usually reported in passive or non-eruptive volcanic plumes. But we believe that exactly for this reason our work on large eruptions is relevant as it complements existing studies. We believe our work is also important for the following reason: without computing time series of flux and further integrating them over time, it is impossible to assess the total mass of SO2 released by a large, long lasting eruption. We have made this point clearer in the text.*

For this reason I recommend that the authors cite the following, complementary paper (currently in press), on a similar topic, but focused on monitoring lower tropospheric emissions with similar techniques:

Carn, S.A., N.A. Krotkov, K. Yang, and A.J. Krueger (2013), Measuring global volcanic degassing with the Ozone Monitoring Instrument (OMI), Spec. Publ. Geol. Soc. Lon., 380, (in press).
The reviewer can supply a pre-print of this article on request if needed.

Reply: *the proposed paper provides useful information and description on techniques to infer SO2 fluxes. Although it only shows time series of OMI-based SO2 masses (not fluxes), we have added a reference to this paper.*

The paper suffers from poor organization and is often hard to follow. Methods and results are intermingled in several places (see comments below) and the paper needs reorganizing in a more logical manner. The use of English could also be improved throughout.

Reply: *The paper has been reorganized following the recommendations of the three referees and the English has been improved and corrected at several places.*

In view of the above my overall recommendation is that the paper, after some improvements, would be better considered for publication in Atmospheric Measurement Techniques (AMT), unless the authors can modify it in such a way as to use their results to address a specific scientific question(s).

Reply: *We disagree. In line with the other two referees, we believe this paper is well suited for ACP. The scope of ACP is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest. We cannot find a single ACP paper which is not technical to some extent. Our paper is definitely not only a technique paper. It combines measurements made in the UV and thermal IR, some modeling and it also gives ideas on possible geophysical applications.*

Specific comments:

31351: the discussion of 'explosive' vs 'effusive' eruptions here is overly simplistic and needs refining; in fact there is a continuous range of volcanic eruption styles of which these are 'end-members'. Both effusive and explosive eruptions involve exsolution of volcanic gases due to decompression, and 'fire fountains' can be considered a form of explosive activity involving fragmentation of the magma (albeit with larger fragments than those generated during more silicic eruptions). Weak explosive eruptions produce lower column heights than large effusive eruptions. I think the key point is that SO2 is a marker of any 'magmatic' eruption and perhaps the major control on column height is the eruptive mass flux (which determines the heat flux).

Reply: *a similar comment has been made by another referee (A. Durant). The paragraph has been re-written.*

31352, L16: scrubbing of SO2 can often lead to a complete absence of SO2 emissions at the surface (e.g., at heavily glaciated or tropical volcanoes), and a dominance of H2S or CO2. Hence, including (4) here as an example of the use of SO2 as a marker of important processes is not really valid, since there may be no SO2 to measure. It would be more correct to use this as an example of when SO2 measurements are less useful.

Reply: *We agree. The sentence has been rephrased: "However, in certain circumstances, SO_2 measurements are less useful. This is the case when dissolution of the gas occurs in a hydrothermal system located between the magma and the surface (Symonds, 2001). This process, also known as scrubbing, can complicate the interpretation of SO_2 flux data, being not related to real magmatic processes."*

31353, L23: suggest replacing 'poor detection limit' with 'high detection limit'.

Reply*: done.*

31353, L26: note that although the TOVS sensor was first launched in 1978, the technique to retrieve SO2 columns using the data was developed much later (2003).

Reply: *Indeed, the sentence is a bit ambiguous. We propose: "In the infrared, space-based sounding of SO2 was also possible with TOVS (Prata et al., 2003), with data going back to 1978".*

31354, L1: the increased sensitivity is largely due to improvements in spectral and spatial resolution, so these are not independent.

Reply*: the word 'sensitivity' has been withdrawn.*

31354, L3: the list of sensors could be updated to include the hyperspectral UV OMPS sensor on the Suomi-NPP satellite.

Reply: *we prefer to leave the list as it is. We could add OMPS but then we need to add a bunch of other satellite sensors making the list too long.*

31354, L9: 'strongest sources' is a little ambiguous here, since there is no indication of the detection limit. Also, the authors should clarify that they are referring to daily satellite measurements, since time-averaging of daily data can be used to detect much weaker SO2 sources.

Reply: *a reference to 'daily satellite measurements' has been added.*

31354, L22: Carn and Bluth, GRL, 2003 should also be cited here. They used TOMS SO2 data to calculate SO2 fluxes from Nyamuragira.

Reply: *done.*

31355, L15: note that OMI is also used in the NASA-NOAA near real-time SO2 monitoring system (http://satepsanone.nesdis.noaa.gov/pub/OMI/OMISO2/index.html).

Reply: *this reference has been added in section 2.2 (description of OMI).*

31357, L28: the origin of the OMI row anomaly is incorrectly described here. It is due to a blockage affecting the nadir viewing port of the sensor, rather than a sensor defect.

Reply: *changed in the text.*

31358, L5: the authors should describe the criteria used to decide if the data were 'useful' – if data are flagged as affected by the row anomaly, it is unwise to use them for scientific analysis.

Reply: *OMI data are routinely displayed (in SACS) simply by removing data belonging to cross-track positions affected by the row anomaly. However, the anomaly effect is not constant along the full orbit. In the vicinity of Nyiamuragira and for certain days , SO_2 data from affected cross-track positions have been found of similar quality than for non-affected pixels. Differences in SO2 column values were found small and it was apparent that the corresponding pixels belonged to the volcanic plume. Therefore we have used these pixels in the analysis. Nevertheless, adding these pixels only has a modest effect on the total SO2 burden (differences less than 10%).*

31360, L13: base -> basis

Reply: *done.*

31362, L14: . . .consider the SO2 mass contained. . .

Reply: *done.*

31362, L16: it could be clarified here that the dimensions of the ‘box’ are usually determined using a trajectory model or radiosonde wind profile.

Reply: *done.*

31364, L10-13: is this statement regarding ‘reliable SO2 fluxes’ based on actual data (e.g., comparison of satellite flux measurements with independent data from ash-laden plumes; if so a reference should be provided) or is it just conjecture? Please clarify.
Furthermore, I would not expect SO2 depletion to be negligible 100s of km from a volcano.

Reply: *this sentence is misleading so we have removed it.*

31365, L3-5: it should be noted that coverage from LEO satellites also depends on latitude, so the UV sensors can provide increased temporal resolution at high latitudes.

Reply: *it is true. We propose to remove the sentence* ‘IASI, with its twice daily global coverage, will provide a higher time resolution than OMI and GOME-2, e.g. which are constrained to operate only during daytime.’ *which brings nothing important in the context of section 3.1.*

31365, L12: with multiple satellites and global coverage, coverage of even very large plumes should not be an issue (with the exception of unforeseen data gaps)? Of course, for very large plumes the assumption of constant k (loss rate) becomes more unrealistic.

Reply: *It is true, complete plume coverage could be achieved using multiple sensors. However, an excellent consistency of the corresponding SO2 retrievals is a prerequisite for any SO2 flux calculation using these data (merged). We propose to replace the sentence “A prerequisite to this method is that the whole plume must be covered. This might be an issue for very big plumes” by “A prerequisite for this method is that the whole plume must be covered. This might be an issue for very large plumes using data from a single satellite instrument with limited spatial coverage ”.*

31365, L22: I don’t think there is any reason why time-series mass flux curves should be bell-shaped. In my experience they are usually quite asymmetric, with an initial rapid increase in SO2 flux followed by a slower decline.

Reply: *the proposed function (Eq. 7) is asymmetric by definition and reflects the observed fast increase in SO2 and then slow decrease (Figs 7,8,9). We think the confusion comes from the terminology “Bell shape curve”. We propose “Skewed shape curve” instead.*

31366, L25: in addition to the other papers cited in this paragraph, the following paper should also be cited, since it refers to an eruption of Nyamulagira (as does this paper):
Hughes, E.J., L.C. Sparling, S.A. Carn, and A.J. Krueger (2012). Using horizontal transport characteristics to infer an emission height time-series of volcanic SO2. J. Geophys. Res., 117, D18307, doi:10.1029/2012JD017957.

***Reply* :** *The papers listed in this paragraph are directly linked to the inverse modeling approach used in the present study. It is not clear why Hughes et al (2012) should be cited. This section deals with the description of the inverse modeling technique which was not applied to the eruption of Nyamulagira .*

31369: I am a little confused by the Puyehue-Cordon Caulle example as the first part of the analysis (delta-M method) appears to focus on the initial SO2 cloud that made several circuits of the globe. Calculating an SO2 flux for a drifting plume seems pointless if no additional SO2 is being emitted – and it is not clear if significant (if any) new SO2 emissions from the volcano were detected by IASI after the initial 2-3 days of the eruption? After June 7 the derived SO2 flux in Fig. 2 seems to tend towards zero, as expected. Focusing on the first few days of the eruption using the traverse method analysis (31370, L16) seems more logical.

Reply: *IASI still detected SO2 close to Puyehue on the 8th of June but much less than for the initial plume; the corresponding flux in Fig. 2 (right plot) is within the noise level on the flux data. We prefer to have both subplots (SO2 mass and flux time series) drawn with the same x-axis limits to illustrate the SO2 e-folding time calculation from the tail of the SO2 mass time series (for the drifting plume with no additional SO2 emissions). It also shows that the fluxes tends indeed towards zero as expected.*

31369, L22: increases in SO2 mass have often been observed in satellite data following large eruptions – can the authors suggest a reason for this? Signal saturation in the fresh plume and/or emission of H2S (later oxidized to SO2) have been suggested for other eruptions.

Reply: *This is a difficult question to answer. In our study, any increase in SO2 mass is interpreted as an increase in SO2 emission but it is true that there could be other reasons for the observed increase in SO2 mass.*
For the Puyehue eruption, the measured SO2 columns have modest values and we don't expect any signal saturation issues. The algorithm of Clarisse et al. (2012) is designed to cope with very large SO2 amounts (see section 2.3). Oxidation of H2S might also play a role, but it is difficult to assess. We have looked for a signal of H2S in the IASI spectra for the first overpasses after the eruption of Puyehue but we could not find any. The detection limit of H2S is however rather high (about 25 DU; see Clarisse et al., 2011).

31371, L5: the plumes were ash-rich throughout the first few days of the eruption.

Reply: *it is true but the impact on SO2 retrievals is likely to be more pronounced for the first overpass (highest ash concentration). We use this sentence instead: "Note that for the first overpass, larger (and also more scattered) values are observed; this is probably due to the impact of ash (high concentrations in the early plume) on the retrieved values ".*

31374, L3: how was the effect of the OMI row anomaly mitigated? The data gaps would affect the delta-M method. This is mentioned later (31375, L3) but should be discussed earlier in section 4.2, along with the use of GOME-2 data. Also, it is not stated which altitude is assumed for the SO2 and/or if the OMI SO2 columns were interpolated to this assumed altitude.

Reply:
**The treatment of the OMI row anomaly is given above.*
**It is already mentioned in section 3.3 that data gaps will affect the delta-M method.*
**The altitude assumed for SO2 is mentioned at the beginning of section 4.2. However, we have made the text clearer by adding that the altitude is the 'center of mass altitude'.*

31375, L11: could the OMI-GOME-2 differences also be due to the temporal offset between the morning (GOME-2) and afternoon (OMI) satellite overpasses? It is not stated if the same SO2 loss rate was assumed for OMI and GOME-2. Could differences in the assumed SO2 altitude (OMI vs. GOME-2 retrievals) also be an issue?

Reply: *in principle a temporal offset should not be a real issue in the calculation of fluxes. One possibility that could explain the OMI-GOME2 differences is the treatment of clouds which is different. Also, the meteorological/cloud conditions between morning and afternoon are probably different and it can also be a cause of discrepancy between the OMI and GOME-2 data. We have changed " .. (probably because of differences in the treatment of clouds in the retrievals).."* by *" .. (probably because of differences in the treatment of clouds in the retrievals and in meteorological conditions)..".*
Different SO2 e-folding times have been used (22.5 h for OMI and 24.9 h for GOME-2). This point has been clarified in the text. Note that, in practice, using one value or the other has a small impact on the retrieved fluxes.
We have used the SO2 middle tropospheric altitudes for simplicity. We have made sensitivity tests and differences due to SO2 altitude cannot explain the observed OMI-GOME2 differences (see also Fig. 1).

31375, L20: does this error analysis refer to OMI or GOME-2 or both?

Reply: *both. We have clarified this point in the text.*

31375, section 4.3: no references are given here for the details of the Nabro eruption – citations are needed.

Reply: *The section on Nabro (introduction, discussion) has been revised (see revised manuscript). Note that when Nabro erupted there was no proximity information so there are only a handful of references available (see text).*

31376, L9-11: I suggest omitting the categorization of the eruptions – Puyehue was not classically Plinian (it also produced a rhyolitic lava flow), nor was Nyamulagira 'pure effusive' (since lava fountains were involved). Although details of the eruption remain unclear, the Nabro eruption was clearly not purely effusive either. Note also that the 1981-82 Nyamulagira 'effusive' eruption produced a SO2 plume that reached the tropopause (Krueger et al., 1996), so this is not exceptional.

Reply: *following similar comment from another review, we have remove the (too strict) categorization of the eruptions in the text.*

31376, L13: the UV satellite images alone cannot reveal the 'multi-layered' nature of SO2 plumes; they are not altitude-resolved.

Reply : *this is true. The sentence is better expressed by: "The comparison of satellite UV and TIR SO2 images revealed inhomogeneous multi-layered SO2 plumes dispersed over large distances, making the SO2 fluxes calculation a complex problem to solve. "*

31376, L18-25: perhaps it would make sense (and save some space) for comparison purposes to present all the inversion settings used for all the eruptions studied in a table?

Reply: *it would be a table quite technical and only relevant for some readers.*
Unless it is really needed, we would prefer not adding a new table as the paper is already quite long.

31377, L6: what is the source of the plume altitude information (15-18 km)?

Reply: *Vernier et al. (2013) show evidence of a volcanic plume in this altitude range using CALIPSO (Fig 1). Fromm et al. (2013) also show satellite evidence for gases and aerosols intrusion at or above tropopause level.*

31377, L8: please explain 'fine tuning'. Also, an SO2 e-folding time of 2 days at the surface seems long – is there a reference for this?

Reply: *'Fine tuning' is not the right expression. The value of the SO2 e-folding time at the surface has been varied until the best match was found between the measurements and the simulations. The value obtained (2 days) is longer than the one obtained at Nyiamuragira (~1day) and it is somehow what we should expect from a relatively drier atmosphere. There is a large scatter on the SO2 lifetime values for tropospheric plumes and the value we derived here empirically is not shocking. Note e.g. that Beirle et al. (2012) determined a lifetime of SO2 of 2-3 days in the Kilauea plume (in a dry atmosphere). We admit the value of 2 days we give here is not a consolidated value but we have made a sensitivity test by using 1.5 days instead of 2 days and the fluxes estimates (Fig. 11) were fairly close to the original values (differences<15%). The sentence has been changed by "The e-folding time parameterization as a function of altitude was estimated by interpolation between the values at surface level, taken as 2 days, and the one at 15–18 km (5 days). The value of the SO2 e-folding time at the surface has been varied until the best match was found between the measurements and the simulations."*

31377, L25: 'limited sensitivity to SO2 in the lower troposphere' – this seems contradictory to an earlier statement (L19) that GOME-2 has sensitivity down to the surface.

Reply: *we mean 'reduced sensitivity' instead of 'limited sensitivity'. It has been changed in the text.*

31378, L22: again, the authors need to state the source of the plume altitude cited here (15-18 km).

Reply: *see above.*

31379, L7-14: in this case, why was the 6 h time step used at all? Was the inversion also performed with a 12 h time step? These issues with the technique should be highlighted earlier (section 3.4).

Reply: *we agree it should be mentioned earlier and this has been changed in the text. The algorithm using the 6h time step was setup before this feature was understood. We have also made the inversion with a 12h time step and the inverted fluxes were similar overall. Whether a 12h or a 6h time step should be used is questionable. It is also altitude-dependent. E.g., the UTLS plumes are better reproduced using a 6 h time step than 12h because the dispersion patterns are more discernible (because of faster transport). Therefore we preferred to use the 6h time step and calculate daily averages at the end.*

31379, L17: 'misfit effects' and other sources of uncertainty in the inversion technique should be described in section 3.4.

Reply: *we agree it should be mentioned earlier too (it has been changed in the text).*

31380, L5-6: details of the eruption (20 km long lava flow) are given here with no citation of the source. In fact, satellite evidence suggests that the extent of the Nabrolava flow was already significant prior to June 17, so this statement is incorrect.

Reply: *satellite pictures of the lava flow of Nabro can be visualized at http://earthobservatory.nasa.gov/. We added this link in the paper and withdraw the reference to June 17 (the flow was indeed already large before that date)*

31380, L7: this information on GOME-2 operations should also appear earlier, before the results.

Reply: *it is already mentioned in section 2.1, but we now repeat it also in section 4.3.1.*

31380, L24: MODIS measurements are introduced here, but the use of MODIS should be described prior to the 'results' section. I recommend that the sensors used in the analysis of each eruption should also be summarized in a table (along with inversion settings and other key information).

Reply: *we agree the use of MODIS needs to be described before the 'results' section. As mentioned before, we are not in favor of adding a new table unless it is absolutely needed. However, the sensors used in the analysis of each eruption are now summarized in the introduction as follows: "The motivation for this collaborative study is an effort to estimate volcanic SO2 fluxes using satellite measurements of dispersed and large-scale plumes of SO2. We make use of the SO2 products from the high spectral resolution OMI, GOME-2 (UV), IASI (TIR) and multispectral resolution MODIS (TIR) instruments. These are currently used in an automated mode to provide alerts for aviation safety (as a proxy for the presence of volcanic ash) or for volcano monitoring, in the Support to Aviation Control Service (SACS; Brenot et al.,* *2013), the European Volcano Observatory Space Services (EVOSS; Ferrucci et al., 2012) and the Support to Aviation for Volcanic Ash Avoidance (SAVAA; Prata et al., 2008) projects. We combine and compare four different approaches and investigate the time evolution of the total emissions of SO2 for three volcanic events (different in type) occurring in 2011: Puyehue-Cordon Caulle, Chile (using IASI), Nyamulagira, DR Congo (using OMI and GOME-2) and Nabro, Eritrea (using IASI, GOME-2 and MODIS)."*

31381, L6: 'volcanic water vapor particles' – does this refer to gas, liquid, solid (ice) or all three? Also, it seems unlikely that water vapor of volcanic origin can be distinguished from ambient atmospheric water vapor.

Reply: *The term 'water vapor particles' refers to 'condensed water vapor'. As we don't know what is the water vapor phase at the plume altitudes or whether different phases coexist, we decided – in first approximation - to use the optical properties of water vapor in the liquid phase. Further analysis show that the use of settings for the solid phase give results that lies within the accepted SO2 retrieval errors (about 40% in these critical cases). Further discussion of this effect is the topic of a paper in preparation and is out of the scope of the present paper. However, to avoid misunderstandings, we have used 'condensed volcanic water vapor' instead of 'volcanic water vapor particles' in the text. We are clearly able to discriminate water vapor of volcanic origin from ambient water vapor (as we are able to discriminate meteorological clouds from ambient water vapor) because it gives meaningful lower TOA radiance in the overall TIR spectral range.*

31381, L19: in their discussion of the initial Nabro eruption plume, the authors need to cite work by Bourassa et al., Science, 2012 (and subsequent technical comments) on the transport of the Nabro volcanic cloud. This is cited later (31382, L27) but appears almost as an afterthought, whereas previous work should always be cited first. Bourassa et al. (2012) invoke upward transport in the Asian Monsoon circulation to explain the large stratospheric impact of the eruption, and the peculiarities of the transport mechanism could partly explain the poor results of the inversion. Indeed, the authors should explore whether their results shed any light on the transport mechanism (which remains controversial).

Reply: *Bourassa et al., 2012 and the technical comments from Vernier et al., 2013 and Fromm et al., 2013 are now cited in the introduction of section 4.3. (see revised manuscript). We agree with the referee that the transport of the Nabro plume is controversial and we have made it clearer in the text. Our analysis shows that the simulations could not fit the observations very well if one assume a passively advected plume. While we believe this is already an important finding on its own, we believe a detailed discussion on the injection altitude and potential lifting mechanisms is out of the scope of the paper, which is focused on SO2 fluxes inversion.*

31382, L3: 'passive' may be a poor choice of word here – in volcanological terms this refers to a non-eruptive plume. By 'passive' do the authors mean a plume transported solely by the ambient wind field, and not by other processes? Could the term 'weak plume' be used instead?

Reply: *Yes. We mean a plume transported by the ambient wind field. We use the term 'passively advected plume' instead.*

31383, L10: 'non-nil' = 'non-zero'?

Reply: *Yes. It has been changed in the text.*

31383, section 4.3.3: since the authors state here that the inversion technique is only applicable if the IASI and GOME-2 SO2 columns are consistent, then this entire section should logically appear prior to application of the inversion technique (to Nabro in particular), in order to validate its use (i.e., before section 4.3.1).

Reply: *we believe it will still not be a stand-alone section because the IASI vs GOME-2 comparison requires the information on the SO2 plume heights provided by the a posteriori FLEXPART simulations that would then only come later. Therefore we propose not to move this section. Following the recommendation of one referee (A. Durant), the section "*4.3.3 Intercomparison of GOME-2 and IASI*" is renamed "Discussion" and includes the discussion on the transport of the Nabro cloud and Intercomparison of GOME-2 and IASI results.*

Figures: several of the figures (e.g., Fig. 11) would require enlargement (relative to the review copy) as the text is hard to read.

Reply: *done.*

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