Marie Boichu's response to referees remarks and questions:

The authors wish to thank both referees for their remarks and comments. Detailed replies and modifications brought to the revised manuscript are following.

Answers to reviewer 1 :

This is a good work, suitable for publication in ACP. I have only few comments. The manuscript provides details about the method here developed and an evaluation of the results. However, I have found the presentation of the material is partly confused. It is difficult to separate the calculation of the results (i.e. the SO2 flux) from the evaluation and from the application to forecast models.

General comments

• Are the authors comparing to IASI observations? Is comparing to IASI correct, considering that the same observations are the starting point of your procedure? Shouldn't the authors use independent observation to evaluate their method?

In Fig. 4, the consistency and stability of the SO_2 flux reconstructions which are obtained through single-image inversions using independent and different IASI images represent supporting evidences that the reconstructed SO_2 emissions are robust, since independent IASI image lead to very similar patterns of SO2 emissions.

Moreover, Section 3.4, which describes the results of the forecast of the volcanic plume evolution, also demonstrates the robustness of the method using independent IASI observations. Indeed, the SO_2 flux, which is used to initialise the forecast, is reconstructed from the inversion of IASI data collected on 7 May 2010. The corresponding forecast at + 2 and + 3 days are compared with independent IASI data, collected on 9 and 10 May 2010 respectively, which were not used to reconstruct the forecasted SO_2 flux. The broad agreement reached between simulations and observations further demonstrates the robustness of the inverse modelling approach.

• How exactly would this method be applicable to other eruptions? The authors make some assumptions, for instance, on the injection height. Do they expect that major changes should be done for other eruptions? Which assumptions should be changed?

The inverse modelling approach developed in this paper could indeed be applied to the study of other eruptions. Compared to standard inverse modelling approaches, we show in this study that no a-priori knowledge on SO_2 flux values is required to perform the SO_2 flux reconstruction, which means that any remote or poorly-monitored volcano can be studied. Nevertheless, the method indeed requires some information on the injection height as an input parameter to initialise the chemistry-transport simulations of the volcanic plume. These constraints can be provided by ground observations, such as Band-C radar measurements which were available for the 2010 Eyjafjallajökull eruption. If no ground observations are available, back trajectory calculations can be made using SO_2 column amount maps of the dispersed plume to reconstruct injection height, provided there is sufficient wind shear above the volcano crater to allow for emissions injected at different altitude to follow distinct trajectories. In the same line of thinking, we could consider reconstructing simultaneously both SO_2 flux and injection height by inverse modelling, if again wind shear permits it.

Moreover, state-of-the-art retrieval algorithms of infrared satellite observations assume a constant plume altitude to convert SO_2 brightness temperature differences to SO_2 column amounts, which may lead to bias in the retrieved volcanic SO_2 column amounts. Recent studies have shown how to estimate the altitude of SO_2 plumes from spaceborne hyper-spectral UV measurements (Yang et al., 2009). But more generally, it would be of interest in the near future not to separate the satellite observation processing from the inverse modelling part. Such a coupling would allow to avoid any assumption on plume altitude in the satellite retrieval algorithm, benefiting directly from the information on plume altitude delivered by the plume chemistry-transport simulations constrained by the inverse modelling analysis.

Yang, K., Liu, X., Krotkov, N. A., Krueger, A. J., & Carn, S. A. (2009). Estimating the altitude of volcanic sulfur dioxide plumes from space borne hyper-spectral UV measurements. *Geophysical Research Letters*, *36*(10).

Specific comments

• 6556 L15: parenthesis missing before Highwood.

ok

• 6561 L1: in page 6559 the authors state that they do not include SO2 oxidation. Does g include anyhow chemical processes?

Reviewer 1 spotted a mistake here. g could theoretically include chemical processes associated to SO_2 oxidation processes, as the CHIMERE model allows for the description of these mechanisms, but these chemical processes have been neglected here, as mentioned in Section 2.1. Clarification has been made on this point in the revised manuscript.

• 6561 L4: if g includes chemical processes, shouldn't they be included in this line?

Answered above

• 6562 L19: Reason number 3 should underestimate the observed plume, too, correct?

That is true that the observed plume can be also under-estimated due to the detection limit, and this sentence presented in this way is not clear.

The authors wanted to point out that parts of the plume may be missed in the observations, leading to an over-count of zero pixels which implies finally an under-estimation of the modelled plume intensity. In particular, the observed plume may appear narrower than it is in reality, due to the detection limit missing SO_2-poor borders of the plume. Therefore, zero pixels at the border of the observed plume may not indicate the true absence of the plume, leading to an over-count of zero pixels.

Text has been changed accordingly in the revised manuscript to clarify this point.

• 6564 L21: Did the plume from Eyjafjallajökull contain much water? Is it a problem for using your method in this case?

Volcanic plumes are generally water-rich and the Eyjafjallajökull plume contained indeed ~92 mol% H_2O in early May 2010, according to FTIR measurements by Allard et al. (2011). The algorithm takes into account the water vapour column (from the IASI level 2) below and above the volcanic plume. The algorithm has an inherent low dependency on the water vapour content though. Large water vapour concentrations within the plume have not been investigated, but will likely affect the retrieved SO_2 columns in a similar way as aerosols and clouds (see below), leading to an overestimation of the retrieved SO_2. Large amounts of water vapour are also known to deposit as ice crystals within the volcanic plume (evidence of ice was commonly observed in IASI spectra) and these do not significantly affect the retrieval.

A mention to this point has been added in the revised manuscript.

Allard, P., Burton, M. R., Oskarsson, N., Michel, A., & Polacci, M. (2010, December). Chemistry and fluxes of magmatic gases powering the explosive trachyandesitic phase of Eyjafjallajokull 2010 eruption: Constraints on degassing magma volumes and processes. In *AGU Fall Meeting Abstracts* (Vol. 1, p. 07).

Answers to reviewer 2 :

General comments

This article presents a method of reconstructing SO_2 emission flux during volcanic eruptions via the combination of satellite observations of the plume and atmospheric chemistry transport modelling. The focus is on the infrared instrument IASI, and the May 2010 eruption of Eyjafjallajökull, though in theory the method is applicable to other satellite-based sensors and indeed other volcanic eruptions. The authors stress that the study of smaller, more regular volcanic eruptions and persistently degassing volcanoes is important for assessing the complete impact of volcanism on Earth's atmosphere – this emphasis is very welcome. Since the method presented does not require any initial knowledge of volcanic emission flux, there are obviously many potential future applications to other remote or poorly-monitored volcanoes. The agreement between model simulations and the satellite dataset is very encouraging, and indicative of the method's robustness. Generally the article is well-written and laid out. Although I think it would benefit from more detail in a few areas, I have no hesitation in recommending this article for publication, with only a few minor revisions in response to points discussed below.

Specific comments

• The neglect of atmospheric chemical processing of the SO_2may be valid in this case, but is less likely to be so at many other volcanoes, particularly those in lower latitudes. A little more discussion of the implications of this would be welcome

Lower latitude eruptions should indeed provide more humid environments leading to significant atmospheric chemical processing of the SO_2 and shorter plume lifetime. In that context, less satellite observations will be available to constrain the atmospheric evolution of the plume, thereby decreasing the accuracy of the SO_2 flux reconstruction by multiple-image inversion. This point has been added in the text of the revised manuscript.

• Connectedly, SO_2 lifetimes are described on the order of days. Some brief comparison between this eruption and other high latitude eruptions where plume lifetime has been estimated would be nice to see.

The SO_2 loss rate is the result of various processes affecting the volcanic SO_2 plume including dry/wet deposition and SO_2 oxidation. These processes depend on many factors including relative humidity, solar irradiation, temperature, aerosol concentration and pH, and availability of oxidants, among which the relative humidity seems to play a major role. High latitudes provide dryer environments that should lead to longer SO_2 lifetime compared to low latitudes (Stevenson et al, 2003). However, for a given volcano, the SO_2 lifetime will vary widely from one eruption to another depending on internal factors, such as the magnitude of the eruption that controls the injection height, and external factors such as the meteorological conditions that prevail at the time of the eruption, etc. That is the reason why, compared to stratospheric SO_2 lifetimes, tropospheric SO_2 plume lifetimes are observed to widely vary, ranging a few orders of magnitude, from less than an hour to days/weeks (Oppenheimer et al. 1998; Stevenson et al., 2003; Rodriguez et al. 2008). Therefore, the latitude of the volcano does not seem the most important factor. The SO_2 lifetime will depend on the complex history of the plume and the atmospheric conditions encountered during its travel which can cover a large range of latitudes (parts of the Eyjafjallajökull plume reach North Africa latitude (~ 30 degree N) in May 2010).

Oppenheimer, C., Francis, P., & Stix, J. (1998). Depletion rates of sulfur dioxide in tropospheric volcanic plumes. *Geophysical Research Letters*, *25*(14), 2671-2674.

Rodríguez, L. A., Watson, I. M., Edmonds, M., Ryan, G., Hards, V., Oppenheimer, C. M., & Bluth, G. J. (2008). SO< sub> 2</sub> loss rates in the plume emitted by Soufrière Hills volcano, Montserrat. *Journal of Volcanology and Geothermal Research*, *173*(1), 135-147.

Stevenson, D. S., Johnson, C. E., Collins, W. J., & Derwent, R. G. (2003). The tropospheric sulphur cycle and the role of volcanic SO2. *Geological Society, London, Special Publications*, *213*(1), 295-305.

• The common trends in SO_2 and ash emission are interesting to see, but I would like to see more discussion of how typical this is. On 6569, line 24, the remark "During any explosive eruptive episode, both SO_2 and ash release generally tend to broadly follow the same trend" should be better supported, with citations. In which other eruptions has this been documented?

More explanations are indeed required to clarify this point.

Gases control the degree of explosivity of an eruption and the associated formation of ash. Explosive eruptions occur when magma decompresses as it rises, allowing dissolved volatiles to exsolve into gas bubbles. It is commonly assumed that magma fragmentation occurs when bubbles occupy ~70-80 vol% of the erupting mixture (Cashman et al., 2000). When fragmentation occurs, violently expanding bubbles tear the magma apart into fragments which are ejected into the atmosphere where they solidify into ash particles. Therefore, if peaks in the ash release rate are observed during a specific eruptive episode, simultaneous peaks in the gas emission rate are expected. However, gas can be released from the volcano without being accompanied by ash-rich explosions, if processes of gas/melt separation occur as magma rises to the surface.

To my knowledge, the similar behaviour of ash and SO_2 release has never been quantitatively highlighted at a volcano before, certainly due to a lack of observations to monitor ash and gas emissions. Hence, this observation of an expected behaviour represents a volcanological result in itself.

An additional discussion on this point has been added in the revised manuscript.

Otherwise, I am not sure that similar trends in ash and SO_2 are a rigorous means to validate your SO_2 reconstruction. Furthermore, closer links could be made between the satellite observations and contemporaneous observations of volcanic activity made on the ground throughout the eruption.

I disagree with this remark. If we had found a completely different evolution of ash and SO_2, we would not be able to conclude on the validity of our SO_2 flux reconstruction. But the broadly similar behaviour gives us a good confidence that we correctly captured the dynamics of the eruption. Quite the contrary, I think that this similarity between ash and SO_2 release rates can be considered as an observational proof that ash and SO_2 do present the same overall evolution, as would be theoretically expected (see previous paragraph).

Gas emissions were not continuously monitored during the 2010 Eyjafjallajökull eruption. Concerning other standard volcanological ground observations which are generally available (such as seismicity, deformation…), their comparison with gas emissions is not straightforward. Ash and SO_2 represent the two closest parameters as they are both components of the volcanic plume. Hence, comparing SO_2 flux with ash release rate is the most direct comparison that can be made for this eruption.

• It is remarked that the method could be applied to other satellite sensors – in what ways would this improve or limit the effectiveness of the method? For example, UV sensors would only see the plume once per 24 hours, rather than every 12. Conversely, an instrument like OMI could potentially detect SO_2 at lower altitudes. In common with reviewer 1, I agree that some comparison with other satellite datasets would be welcome, if available. I appreciate however that differences in overpass time, sensitivity, detection limit, spatial resolution, etc, may not facilitate straightforward comparison.

Indeed, the inverse modelling approach developed here to reconstruct the SO_2 flux can be applied to any ultraviolet (including OMI, GOME-2, etc…) or infrared (including AIRS, etc...) sensor which can provide information on SO_2 column amounts in the volcanic plume. This proof-of-concept study opens new perspectives of rigorous comparison of the volcanic SO_2 observations collected from diverse satellite sensors but also with ground-based instruments. So far, characteristics of the various satellite sensors, with different overpass time, spatial resolution, etc, impeded a rigorous comparison. Moreover, few attempts have been made to compare observations of volcanic SO_2 plumes acquired by satellite and by ground measurements. The main reason for this is that spaceborne or ground-based instruments do not measure the same physical quantity, but rather measure the integrated SO_2 column in a line of sight that is specific to the instrument. Comparison of results obtained by different spaceborne or ground-based instruments can only be achieved by the estimation of a common parameter, such as the SO_2 flux emitted at the source, which is now possible with the method developed in this paper.

Gathering the wealth of observations of the SO_2 plume from different instruments will improve the accuracy of the SO_2 flux reconstruction by inverse modelling, as the various spaceborne and ground-based instruments are complementary. To cite a few complementarities, UV sensors are indeed more sensitive to low altitude plume than IR sensors but do not provide night-time observations, ground-based instruments can detect SO_2 emissions of lower intensity than satellite instruments, etc…

However, performing such a multi-instrument analysis first requires a detailed validation study which is beyond the scope of this paper.

• Total mass of the emissions (ie. integral of the flux history) is mentioned but not quantified (6562, line 09). Some estimate of the total mass of the eruption would be welcome, and could be compared to previous estimates achieved using other methods. Total SO2 mass release is an important parameter for assessing the climatic impact of eruptions.

The integral of the flux history derived in this paper indicates that Eyjafjallajökull volcano emitted from 1 to 13 May 2010 a total mass of SO_2 of ~ 0.17 Tg (170 kt). Our estimation of the total mass of SO_2 expelled by Eyjafjallajökull in early May 2010 has been added in the revised manuscript.

However, this mass cannot be straightforwardly compared to the estimations reported in other studies, since the latter rely on a simple analysis of satellite observations. Indeed, common estimations refer to the mass of SO_2 that is present in an image at the time of image acquisition, after having checked that this satellite image captures the whole volcanic plume. Hence, this estimation includes different SO_2 contributions: the fresh SO_2 emissions that were just released by the volcano, and also the SO_2 remaining in the atmosphere which is associated to releases of SO_2 in the days preceding the satellite image acquisition.

As an example, Carboni et al. (2012) obtain a peak in the SO_2 mass of 0.17 Tg on 7 May 2010. This estimation is deduced from the use of a single IASI satellite image. Other estimates include a mass of ~0.1 Tg from GOME-2 (Rix et al., 2012) and ~0.05 Tg from OMI (Thomas and Prata, 2011).

Therefore, the estimation provided in our study is in broad agreement with previous estimations. Differences could be attributed to (1) the fact that single-image estimates miss parts of the total amount of gas released in the atmosphere (either because it is too old and therefore too diluted or already transformed into another species by atmospheric processing, or because gas release occurring after the acquisition of the image is not yet observed), (2) different sensitivity of the different sensors (e.g. IR sensors might miss parts of the plume located at low altitude), or (3) differences in the processing of the images, in particular when comparing the results obtained from IASI in our study and those from Carboni et al. (2012).
Note that an error on the SO_2 flux scale was noticed in the various figures displaying the reconstructed SO_2 flux time-series in the previous ACPD version. This has been corrected in the revised manuscript.

Carboni, E., Grainger, R., Walker, J., Dudhia, A., & Siddans, R. (2012). A new scheme for sulphur dioxide retrieval from IASI measurements: application to the Eyjafjallajökull eruption of April and May 2010. *Atmos. Chem. Phys*, *12*, 11417-11434.

Rix, M., Valks, P., Hao, N., Loyola, D., Schlager, H., Huntrieser, H., ... & Inness, A. (2012). Volcanic SO2, BrO and plume height estimations using GOME-2 satellite measurements during the eruption of Eyjafjallajökull in May 2010. *Journal of Geophysical Research: Atmospheres (1984–2012)*, *117*(D6).

Thomas, H. E., & Prata, A. J. (2011). Sulphur dioxide as a volcanic ash proxy during the April–May 2010 eruption of Eyjafjallajökull volcano, Iceland. *Atmos. Chem. Phys*, *11*(14), 6871-6880.

• Cloud cover masking the SO2 plume is mentioned as a potential source of error (6564, line 05) but is not discussed further. Does any information on cloud cover at the time of IASI overpass exist?
As shown by Clarisse et al. (2012), the presence of aerosols below the volcanic plume, including cloud water droplets, has a limited impact (up to 2%) on retrieved SO_2 abundance as long as the plume is not completely obscured by their presence. Hence, the existence of underlying clouds marginally affects the retrieved SO_2 flux. The presence of cloud water droplets above the plume may give rise to 45% overestimation of the SO_2 abundance (Clarisse et al., 2012), depending on the cloud aerosol load. This bias on the SO_2 retrieval is expected to affect the estimated SO_2 flux. However, it is unlikely that the volcanic SO_2 plume will permanently travel below clouds. Therefore, the multiple-image inversion is expected to reduce the impact of this bias as the procedure will incorporate various satellite observations of the same parcel of SO_2 at different ages, in cloudy- but also in non-cloudy scenes. Finally, the presence of very thick meteorological clouds extinguishing infrared radiations can mask any underlying SO_2 plume. But again these are not expected to follow the SO2 plume for longer periods of time.

• Similarly, on 6565, line 01 onwards: has the impact of ash in the plume on the IASI retrieval been quantified or accounted for on each day? Are the days where large ash releases are also observed less reliable?
Ash was not simultaneously retrieved with SO_2 in the IASI retrieval presented here, but the standard state-of-the-art IASI SO_2 product, described in Clarisse et al. (2012), was performed. Sensitivity studies on the impact of the presence of ash in the plume on the IASI SO_2 retrieval have been published. If the ash-rich plume does not become opaque to infrared radiations, it is shown that the presence of a thick layer of ash in the plume can cause an 20% overestimation

of the retrieved SO_2 column amount (Clarisse et al., 2012). This uncertainty mainly depends on the plume ash loading and will decrease when the plume gets further from the volcano as ashes sediment. Consequently, 20% should represent an upper bound of the resulting uncertainty on the SO_2 flux, as the multiple-image inversion is based on satellite observations capturing a SO_2 parcel of the plume at various distances/ages from the volcano. This bias in the SO_2 retrieval, related to the presence of ash, may have a moderate effect on the amplitude of the peaks in the SO_2 flux time-series but will not affect their timing. The agreement that is reached between observed and modelled SO_2 plume intensity (Fig. 5) demonstrates that this effect seems minor.
A more detailed discussion on this uncertainty has been added in Section 3.3 in the revised manuscript.

Technical comments
• 6557, line 24: consider "repeatedly disrupt"
• 6557, line 25: consider "for" instead of "during"
• 6558, line 15: Due to the limited scope of the forecasting demonstrated herein, perhaps consider "may yield"
• 6559, line 24: consider "has provided, since 2006,"
• 6560, line 14: alternative to "restituted"?
• 6561, line 21: sentence beginning "No a-priori" is confusing
• 6564, line 09: consider "regular" instead of "redundant"
• 6564, line 10: consider "travels" instead of "transits"
• 6568, line 03: consider "after travelling" instead of "after a long travel of"
• 6569, line 17: "in detail" not "in details"
• 6570, line 14: Following on from a point made earlier use of "strong similarity" needs more support, and seems contradictory to "broad similarities" (6570, line 07)
*• 6573, line 25: "should make **it** possible"*
Corrections associated to these comments have been included in the revised manuscript.