Replies to the comments of Reviewer #1

General comments:

The paper by Clarisse et al. contains two major subjects. First, a new method for effective retrieval of SO2 plume heights from IASI measurements is introduced and compared with external data, and, second, the evolution of SO2 plume heights from the Nabro eruption is discussed. The presented work is a timely and valuable contribution which adds new data helping to understand this interesting event. Further the method seems to be well fitted for standard processing of huge amounts of data as in the case of the IASI instruments. My main comments are directed to a better understanding of the method itself, its limits and uncertainties:

We would like to thank the referee for his/her careful reading, review and useful questions and comments, which definitely helped to improve the paper. We have addressed all comments below and revised the manuscript where needed.

- It is mentioned that the altitude-resolution of SO2 stems mainly from the interfering water-vapour lines (P31168 L20). How can it be explained then, that there is such a low sensitivity on the real atmospheric situation as stated on P31169 L28?

We did not observe large differences when we did the altitude retrieval using only one set of Jacobians, i.e. definitely not in the broad categorization low/mid/upper troposphere. We did not look in great detail in the low-mid tropospheric plumes, but differences were observed exceeding the +/- 2km uncertainty of the retrieval. We have therefore removed the sentence saying that the results were 'very similar' and also changed the next sentence on the recommendation for operational applications, which now reads: 'For operational applications Jacobians would need to be precalculated for such boxes and different time-periods of the year'

In addition, as we explain below, water vapour interference is not the only component responsible for the altitude sensitivity. Pressure and temperature dependence on the SO_2 line shapes also contributes. This also helps explaining the not-so-large dependence on the atmospheric state.

- On P31168 L18 it is mentioned that 'large differences in the Jacobians can be observed up to an altitude of 15 km'. Further, also in Fig. 1 the Z(h) functions are rather smooth, especially those for which the maximum is above 10 km (the blue curve). One would guess that this leads to a larger error in the height determination than suggested by the comparisons with CALIOP. Could you reason why this might not be the case?

This was indeed also surprising to us. One possible explanation is that the error on Z is very small (easily less than 10% for a few DU); and so while the curve appears to be smooth in a normalized graph, in absolute value it still offers enough resolution in practice.

This does not take away the fact that the resolution below 15 km is better, which we also demonstrate below with a forward simulation (see next point).

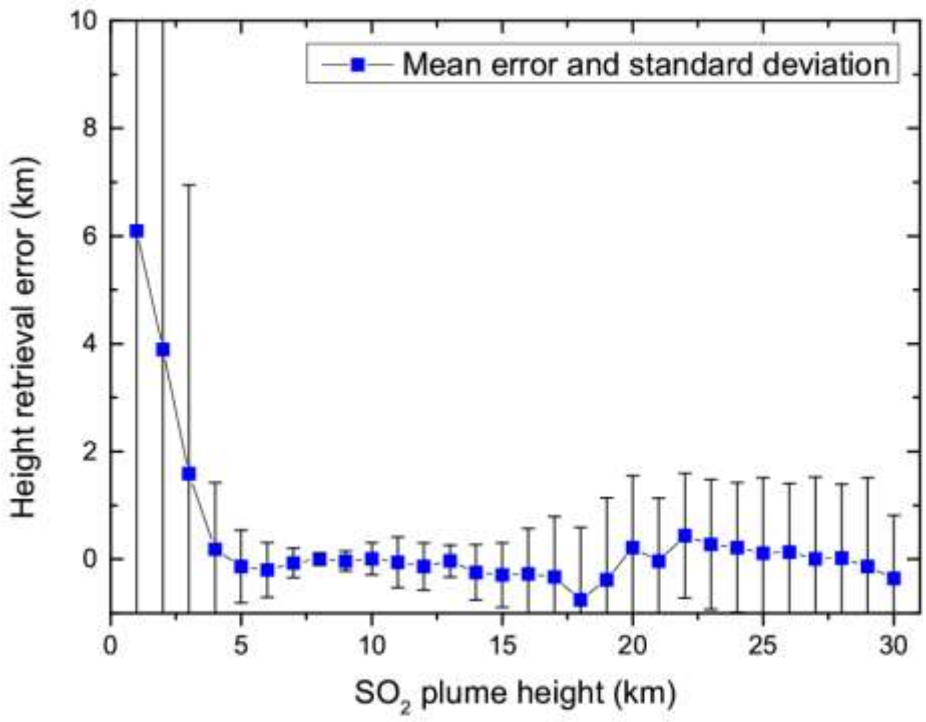
- Since in the stratosphere the water vapour is rather low, the altitude resolution of the method for stratospheric plumes (e.g. above 18 km in the tropics) should also be rather bad (if there is even any). Could you make any simulations to show that the method can resolve the altitude of plumes situated clearly above the tropical tropopause? Otherwise one could argue that stratospheric plumes cannot be resolved and even, might be put by the retrieval to an altitude at or just below the tropopause (where a lot of the retrieved SO2-heights lie).

Thus, can you really state from your observations how much of the early plume is directly injected into the stratosphere or is the method just not sensitive enough to make such a statement. This should be clearly expressed in the conclusions of the paper.

Thank you for suggesting testing the algorithm with simulations. We have now carried out such a simulation and discussed the results in the revised manuscript. The figure belonging to the discussion has also been reproduced here.

'To test the theoretical accuracy of this method for different altitudes, 10000 forward simulations of 5 DU SO_2 clouds between 1 and 30 km were carried out. To make the simulation as realistic as possible, spectral noise was added to these spectra. This noise was generated from the multivariate normal distribution with the mean bias and covariance matrix used for the quantitative SO_2 column retrievals (see end of this section). Calculating spectra in this way is a realistic way of simulating real observed spectra because biases in the forward model are removed, and because instrumental noise is added. However, note that the atmospheric parameters used in the simulation are the same as the ones which were used to construct the Jacobians. Using this method we hence obtain upper bounds on the accuracy of the algorithm.

The results are summarized in Fig. 2. As expected from the Z(h) profiles shown in Fig. 1, the best accuracy is achieved between 5 and 15 km with error bars below 500 meter. Below 3 km the tropical atmosphere is almost opaque in the spectral range of interest due to water vapour, and the algorithm therefore loses its accuracy drastically (a dryer atmosphere would allow to penetrate lower down). Above 18 km, the error bar is almost constant at around 1.5 km. At these altitudes, the water vapour column in the atmosphere is low, and the fact that such a good sensitivity is achieved is related to pressure and temperature dependence of the SO_2 lines (see also Clerbaux et al, Geophys. Res. Lett., 2008, 35, L22807). Although hard to see with the naked eye on apodized IASI spectrum, the simulation demonstrates that altitude information is contained in the spectrum even in the stratosphere.'



Specific comments:

P 31162 L 14 ‘Evidence is presented that emissions in the first 15 week of the eruption also contributed to the stratospheric sulfur input.’: This sounds like direct emissions into the stratosphere. Perhaps add ‘via slow ascent’.

We believe that the line below explains this sufficiently: ‘This includes a second eruption between 15 and 17km on the 16th and continuous emissions in the mid-troposphere of which some were also entrained and lifted within the anticyclonic circulation.’

P 31165 L 27 ‘to retrieve vertically resolved SO2 columns’:
Columns with vertical resolution seem inconsistent. Perhaps use ‘profiles’ or ‘partial column amounts’.

Yes we agree. However, also a profile is not entirely correct in this context, since all SO2 is assumed to be located in a narrow altitude band. We therefore replaced the word ‘columns’ by ‘clouds’ since this conveys better what is actually retrieved.

P 31167 L10 Eq. (2) and lines before:
Please clarify if xˆ here is not height-dependent, i.e. one number and not a vector.

This has now been clarified in the beginning of that section by explicitly referring to a total column retrieval.

P31169 L20 ‘using average atmospheric conditions’:
Which input data have been used to determine these conditions?

These were calculated from the level 2 IASI data (from EUMETSAT) of several days in June 2011. This information has now been added in the manuscript.

P31168 L25: Do you need other Jacobians in case of clouds (I assume they look different since they shield the H2O-features from below) and if not, could you explain the reason?
P31170 L1 ‘covariance matrix S we used one million random IASI spectra’: Has there been a selection with regard to cloud contamination? If not, why is it not necessary?

Clouds were not treated separately. As pointed out in the manuscript when the comparison is drawn with CALIPSO (where the location of clouds can be seen), the algorithm seems to perform no worse in the presence of meteorological clouds (below or above the plume),

The likely reason is that clouds have a broadband effect on the spectra, while in the spectral band of interest most of the spectral information on SO2 and H2O is contained in sharper spectral feature originating from spectral lines. By including cloudy spectra in the covariance matrix, they become part of the spectral noise in the height/detection algorithm. This argument is made in the manuscript (end of page 31168, beginning 31169) In this context, see in also the discussion of clouds in Carboni, E.; Grainger, R.; Walker, J.; Dudhia, A. & Siddans, R. A new scheme for sulphur dioxide retrieval from IASI measurements: application to the Eyjafjallajökull eruption of April and May 2010 Atmos. Chem. Phys., 2012, 12, 11417-11434.

We have now added two sentences in the algorithm description to clarify that clouds are not treated separately (both for the height and SO_2 detection and the SO_2 column retrieval).

P31170 L5-10:
From the description of the quantitative column retrieval, the SO2 column is retrieved as it would be situated only at the layer belonging to the previously retrieved maximum height. Is it then the case that only the Jacobians from these altitude are used? How does the retrieval result depend on these Jacobians? How do the averaging Kernels of the result look like?

Futher, I assume that the retrieval is linear or do you use explicit forward calculations? In the first case: what is the error due to the linear fit? If an iterative fit is done, could you describe more in detail the forward-model and used input data (e.g. atmospheric profiles). Could you give in any case some error estimation?

The retrieval uses an (iterative non-linear) optimal estimation scheme (but with a generalized covariance matrix) as explained in the end of section 2. The forward model is fed collocated IASI L2 data (water and temperature profiles). These two facts are now explicitly mentioned in the manuscript.

As altitudes are assumed at the retrieval stage, it becomes a one-dimensional retrieval (with a scalar as an averaging kernel). For the vast majority of retrievals the choice of a prior is irrelevant, since the covariance matrix is chosen very large and since pixels are pre-filtered for having a large SO2 signal.

Estimating errors from the retrieval is very difficult, since by far the most important error term comes from errors in the assumed altitude, for which the current scheme also does not explicitly quantifies the uncertainty (which is the reason why a comprehensive validation was offered for the retrieved altitudes). The error on the actual SO2 retrieval, assuming perfect knowledge of the height is low (estimated below 25% for plumes above the lower troposphere and columns above 2 DU for a similar retrieval scheme presented in Carboni et al., ACP, 12, 11417–11434, 2012).

P31176 L13:
To get a better overview of how well the plume altitude fits with CALIOP aerosol height it would be good to show a summary of all matches from the single plots in form of a scatter-plot, like Fig. 10.

In preparation of the initial manuscript, this was attempted; however the manual attribution of many of CALIPSOs features makes this technically difficult. In addition, a plot like that would not represent the CALIPSO data independently, since IASI data is needed to locate the volcanic plumes.

P31177 L3:
Could you name a source for the MLS data and a reference?

A recent reference on MLS SO2 does not seem to be available, however the official product description can be found at https://mls.jpl.nasa.gov/data/v3-3_data_quality_document.pdf. We have now added a sentence with the origin of the data (in the caption of the relevant figure for clarity):

'MLS data is from the v3.3 EOS MLS Level2 which was downloaded from the NASA Goddard Space Flight Center, Data and Information Services Center (http://disc.gsfc.nasa.gov/).'

Technical comments:

P31166 L15 Eq (1) and further mathematical expressions:
The ACP convention is that for vectors bold italic fonts should be used. Could you change it everywhere in the manuscript.

Corrected

P31167 L20: 'residues' -> 'residuals'

Corrected

P 31174 L16 ‘east side’: -> ‘west side’ (?)

Corrected

P31177 L7: ‘height altitude Nabro plume’ -> ‘high altitude Nabro plume’ (?)

Corrected

P31177 L17: ‘becomes’ -> ‘become’

Corrected

P31185 Fig. 2: Could the position of the volcano be indicated more clearly.

Corrected

P31194 Fig. 11: Overlapping tick labels in middle row.

Corrected