Anonymous Referee #1

This paper presents a retrieval scheme of atmospheric SO_2 from measurements of the high resolution infrared sounder IASI. With respect to existing retrieval methodology, the approach has two novel aspects:

1. Use of a generalised error covariance which includes more than just the instrumental noise. It includes for instance the covariance due to modelling errors and due to interfering trace gases and clouds. In this way, these do not need to be retrieved or taken into account otherwise in the retrieval. The advantage is also that of a more accurate error budget at the end of the retrieval.

2. Explicit retrieval of altitude. Most other retrieval schemes that retrieve altitude information attempt to retrieve a profile. This often leads to plumes with an unrealistic broad profile. In this paper, SO_2 is explicitly confined to a layer of certain width and this implies that the plume has been injected at a certain altitude (ie is of volcanic origin).

This approach obviously has certain advantages.

I think the retrieval algorithm is scientifically sound, important and interesting. However, as outlined below, the presentation of this paper leaves much to be desired, and needs a thorough revision prior to publication.

Comments related to the presentation:

- Spelling and grammar. It is surprising to read so many grammar and spelling mistakes, especially as many of the authors are native speakers. Given this fact, it does not show a lot of respect to leave it to the referees or copyeditor to correct them. Just some examples:

We apologise for the level of English. The manuscript has been carefully revised accordingly. All of the specific points mentioned have been addressed.

3. So again, the whole manuscript needs to be revised very carefully for spelling and grammar mistakes.

A revision of spelling and grammar has been done.

- Figures:

Figure 1: the axis labels (especially of the colour bars) need to be much larger. The caption mentions ?aiting functions

A new fig. 1 with bigger labels has been included. The figure now occupies a full a4 page rather than the smaller ACPD page dimension.

Figure 3: this is very hard to read. The first row should be removed I think, as it is confusing. The first row essentially shows the colour bar. The use of so many different colours makes it hard to deduce anything quantitative from the other rows (and this and next figures are not quantitatively described in the text either). What would really help is to convert the S02 absolute errors in relative errors (in %). Using a smaller number of colours should also make it easier to read (eg. in 5 bins <1% <5% <25% <100% and >100%). For the altitude, I suggest converting the pressure altitudes in height differences (km). For the error of

the surface temperature and DF I suggest using a smaller colour scale (eg 0 to 2 and 1 to 4 respectively).

The figure has been modified as follows:

- The first row has been removed,
- Pressure values have been converted to km (in all plots in the paper).
- Colour scales for surface temperature and DF have been modified as suggested.
- Both absolute and relative errors are now shown.

We believe absolute errors for SO2 amount are useful: The error in column density is tends to a fixed value at low optical depths and it is useful to see this value clearly. It is an important aspect of the algorithm that the minimum error is ~0.2 DU and useful to see how this varies will altitude.

We have also added a more quantitative description of the results in the main text.

Figure 4: same comments as figure 3. Rather than the retrieved values, it would be much better to plot either the relative or absolute differences (relative difference for SO2, absolute for the other two). This would also make direct comparison with the other rows of figure 3 much easier. It would be good to also here, convert the pressure altitudes to heights (differences).

As above, pressure values have been converted to km.

Fig 3 represents the total errors (including random errors and smoothing errors).
The point of the Fig 4 is to show where the retrieved values are different from the true state because there is insufficient information, illustrating the tendency towards the *a priori* state in these circumstances. Plotting differences from the 'truth' will show an estimate of the smoothing error (i.e. one component of the error shown in Fig. 3). All these difference values are smaller than the errors shown in Fig. 3.
However we agree that the figure is somewhat confusing and the purpose of the plot had not been made clear in the text.
We have now replaced the figure with one showing the values of the averaging kernel. This directly shows the balance between prior and measurement information as a function of SO2 amount and layer altitude. The text has been modified to explain the significance of the figure.

Figure 5: same comments, please convert everything to differences (absolute or relative). What are the numbers inside the plot of the cost?

The plot has been modified to show absolute difference for SO2 and altitude in km.
The optical depth range for ash has been extended to span thin to opaque conditions.
The numbers inside the cost plot showed the value of a convergence flag. These are not relevant so they have been removed.

Figures 6-8: I could not see any value over 10 DU. The figures would look better with a reduced colour bar 0-10 DU (or even smaller).

There are several parts of the plume with SO2 up to 20 DU, especially in May.

The colour scale has been modified and the plots redrawn to improve their clarity.

Specific comments:
- Page 11862, line 5: nu1 is at 8.7 and nu3 at 7.3

done.

- Page 11862, line 22: 0.02 Tg instead of 0.2 Tg
done

- Page 11864, line 13: I wouldn? use the word affected here, since it is a good thing; more radiation is more signal and therefore higher sensitivity.

Changed to
“The 4.0\,\unit{\mu}m absorption feature ($\nu_1+\nu_3$) is weak and the reflected solar radiation is significant during the daytime.”

- Introduction: here other SO2 retrieval algorithms from high resolution infrared sounders should be reviewed (from IASI but also AIRS and TES).

Agreed. The text has been extended to include references to (in particular):
AIRS: Prata and Bernardo 2007, Carn et al. 2005;
TES: Clerbaux et al 2008;
IASI: Clarisse 2012

- Page 11865, section 2: some of the IASI specifications are inaccurate, such as the swath, footprint, 50x50 ?quare radiometric noise (please specify the spectral region here).

'It has a nominated radiometric accuracy of 0.25.58 K. The IASI field-of-view (FOV) consists of four circular footprints of 20 km diameter (at nadir) inside a square of 50 ? 50 km, step-scanned across track (30 steps). It has a 2000 km wide swath and nominally can achieve global coverage in 12 h (although there are some gaps between orbits near the Equator).'

changed to

'It is specified to have a radiometric accuracy of 0.25-.58 K and a field-of-view (FOV) of 12 km at nadir. It has a 2200 km wide swath and achieves near uniform global coverage in 12 h (although there are some gaps between orbits near the Equator).'

- Page 11866, line 11: this is unclear, please revise

The section describing RTTOV has been reworded to make the approach clearer.

- Page 11872-3: please specify the precise spectral retrieval range.

Added:
'In this work the retrieval have been performed using all the IASI channels between 1000-1200 and 1300-1410 cm^{-1}.'

- Page 11874: Line 5: what do you mean with 'We do not subtract the bias?"

In this retrieval we used the same Se and a priori settings as in the retrieval applied to real measurements. However, in the retrieval simulation, the bias spectrum b was not used (since it represents the systematic forward model error which is not relevant for measurements simulated using that same model).

The error analysis paragraph has been rewritten, and in particular Fig. 4 is substituted with the averaging kernel plots. This part of the text has been removed (see following comment).

- Page 11874, section 5: the presented analysis is only representative for real situations when also Se errors are added to the simulated spectra. It was not clear to me if this has been done or not. If not, I suggest redoing the analysis, but this time adding noise generated randomly from the Gaussian distribution specified by the covariance matrix Se and its corresponding mean. A function for drawing randomly from a distribution like this is available in most statistical software packages.

We apologise that the text and Figs 3 and 4 did not make sufficiently clear the points we wanted to make. We want to show the total retrieval error (Fig 3) and the influence of the prior (Fig 4). The approach suggested by the reviewer is a way of estimating the noise component of the total error which is plotted in Figure 3. If we repeated this approach many times and plotted the standard deviation of the error found this would show the sqrt(diagonals) of noise covariance Sn, which will equal the values shown in Fig 3 when the averaging kernel is 1 (noise errors will be smaller than the total otherwise). As discussed above we have now replaced Fig.4 with a plot of the averaging kernel which we believe to be clearer way of showing the sensitivity (where relevant) to prior information.

It is recognised that computing the noise error in the way suggested would also serve to test the retrieval convergence, however that the retrieval converges is not in doubt from the performance when applied to real date.

- Page 11874, section 5: One type of error not covered in this analysis is the effect of errors of the temperature profiles on the retrieved SO_2 parameters. While errors of the temperature profile that propagated to e.g. water vapour line strengths are accounted for by using the generalized Se, they are not taken into account when it comes to the line strengths of SO_2. This could be checked separately by adding synthetic noise to the temperature profile prior to the retrieval. This effect should perhaps be mentioned in the text.

We agree with the referee, this error was not included in our analysis.
We have now done a test (for all the SO_2 amount and altitude as fig 3) adding 1K of systematic error at every level of ECMWF temperature profile. The resulting errors are negligible compare to the total error reported in Fig 3.
A comment on this has been added in the text.

- Page 11875-6. Although the description of Figure 3 and Figure 4 is quite long, some accompanying for would be really helpful, especially for SO_2 and the altitude. E.g, for plumes above 3 km and below 7 km, we expect an error between .. and .., E.g. for plumes between 1 and 10 DU we expect an altitude error between .. and .. km. etc.. When the figures are remade following the suggestions above, it should also be easier for the reader to figure this out byhim/herself.

Typical numbers and ballpark ranges for errors have been added to the text.

- Page 11877, section 5.1: at what wavenumber is the aerosol optical depth reported? Which refractive indices were used?

Ash and water cloud optical depth are both referred to 550 nm.
Ash refractive indices are from Dan Peters measurements (personal communication).
These points have been added to the text.

- Page 11877, section 5.1: there is a large difference between the effects of water and ash clouds. In both cases, can you show a spectrum with a large AOD to demonstrate these differences? From the errors in the SO_2 retrievals, it is hard to believe that the ash simulation is realistic (see also next point). Also, something should be said with respect to the chosen spectral retrieval range (nu1 should be more sensitive to ash than nu3).

Ash simulations with AOD (550nm) up to 2 (at 550nm) were intended to represent a plume dispersed far from the vent. However this does not give an opaque plume (AOD =2 is not a really thick ash), hence the weak impact on SO_2 compared to more optically thick cloud. We have extended Fig 5 with ash AOD up to 20 to cover opaque conditions. A masking effect, similar to cloud, is now seen.

We have added a new plot with standard atmosphere, ash and water cloud spectra, with and without SO_2, and the difference between 'clean' and SO_2 affected spectra. The plot shows a thick water cloud (550 nm OD=10) and an ash plume with OD=3. These have comparable transmission in the v1 range (both ash and cloud covered the SO_2 signal), but ash has less impact in the v3 band (here cloud masks the SO_2 signal much more then ash).

The text has been extended to cover these points.

- Figure 5: it appears that ash above the SO_2 layer has no impact on the retrieved loading. This is not logical and not clear from text. Thick ash clouds will completely wipe out the SO_2 signal (even in the nu3 band) and obviously lead to errors in the retrieval. Could you investigate or clarify?

See above.

- Page 11877, section 5.1: the whole section really needs a rewrite, as it could be formulated much clearer.

Section rewritten.

- Page 11879, line 15: please give a reference for these numbers.

References added:

European Commission; The Impact of the Volcanic Ash Cloud Crisis
on the Air Transport Industry, SEC(2010) 533, 27 April, Brussels, 2010.

Mazzocchi, M., Hansstein, F. and Ragona, M.: The 2010 volcanic ash
cloud and its financial impact on the European airline industry, CESifo
Forum 11: 9200, 2010.

- Page 11880: line 17: this is an unusual way of calculating the total mass, which will lead to large errors. At nadir, IASI has a circular footprint of 12 km diameter and therefore the total mass of a pixel at nadir will be overestimated by a factor of five. At the swath? maximum, when the footprint is elliptical (20x39), the mass will be underestimated.
Also, IASI has gaps between the different scan lines and arrays of 4 pixels.
Just summing up the surface of the footprints will lead to large underestimates. At high latitudes on the other hand, IASI orbits overlap and by summing up the different footprints, the same so2 will be counted multiple times leading to so2 overestimates.
An approach to calculate the total mass which doesn? have all these disadvantages (albeit also not perfect) is to interpolate the IASI data onto a regular latitude/longitude grid.

The mass has be recalculated as suggested (by interpolating and re-gridding to a regular lat lon grid of 0.125 degree intervals and the total mass is computed multiplying the grid box column density * grid box area.)
Fig 9. shows the new mass values and text has been adapted to explain the altered approach.

M. Watson (Referee)
This paper details the development and testing of a new algorithm to retrieve SO2 burdens from the IASI sensor. The authors use published observations of the 2010 eruption of Eyjafjalljokull as a case study to compare with their results. I?e seen this work presented a few times and I am familiar with the algorithm and the early results.
The retrieval is physically sound and has the capacity to significantly improve our ability to retrieve SO2 from hyperspectral sensors. The paper has two issues, both of which are serious but easy to address.

The first issue is a lack of understanding of the volcanological literature. It is inappropriate, for example, to suggest that Prata, 2009, was the first paper to demonstrate SO2 is the third most abundant volcanic gas. Throughout the m/s there are problems where the authors use recent papers as a proxy for better citations.

They also (understandably) lean on the remote sensing literature a little too much. This is bearable until the description of the eruption and the context of the results. The authors need to read the special issue on Eyja carefully and reassign their results accordingly. This section should be rewritten in the light of this work.

The second serious issue with the work is the presentation the English is poor in places and the figures, especially the axis labels are unreadable in several figures. A really good proof read would have made the reviewer experience much better. I have tried to capture as many of the editorial issues as possible but confess to losing the will to live at some point...
Reviewer 1 raises interesting and valid points. In addition, please consider the following...

11862 5: wavelength/wavenumber relationship between two absorption bands flipped

corrected

20: Not sure this should be counted as phase 1 see later: : : 22: 0.2 is not an order of magnitude lower than 0.14

0.2 was an error, corrected with 0.02

Plus check with other Ejya literature confirm the 'phase' division for Ejya eruption (Stevenson JGR et al 2012, Peterson JGR 2012)

5: Implies was the first to make this observation.
Better would be Symmonds et al., 1994
changed Prata (2009) with Symmonds et al., 1994

8-9: This was not really the finding of Thomas and Prata, 2011 quite the opposite in fact. It is very often not a good idea to use SO2 as a tracer for ash.

'Moreover often the SO2 volcanic plume can be used as a proxy for with volcanic ash (Thomas and Prata, 2011), especially within a few hours of release when the effect of wind shear and gravitation have not divided the ash plume from the SO2.'
Changed in
'Moreover, the volcanic SO2 plume can be used as a proxy for volcanic ash within the first few hours of its release, when the effect of wind shear and gravitation have not separated the ash plume from the SO2. However this needs to be done with extreme caution as several eruptions show separation of the two species (Prata and Kerkmann 2007, Prata 2010, Thomas and Prata 2011, Sears 2012).'

12: Carn et al., 2003 is a better description of the TOMS archive.
The referee is right, Prata 2003 is referring to TOVS. A Carn citation has been added for TOMS.

Sentence changed to:
During the past thirty years, satellite observations using spectral bands containing SO2 absorption in the ultraviolet (UV) or the infrared (IR) have been used to monitor explosive volcanic activity e.g.\ Total Ozone Mapping Spectrometer, TOMS, data from 1979; TIROS Operational Vertical Sounder, TOVS, data from 1978 (Carn et~al.,~2003, Prata et al. 2003)..

11864 3: (and elsewhere): ground measurements should be ?round-based measurementsunless you are measuring the ground

corrections added

6: Add recent paper by Henney et al, 2012 and rewrite.

Added these citations in the following sentence:
Some recent work (Henney et al, 2012, Merucci et al 2011)
compares ground-based SO_2 concentrations with satellite data.
However ground-based data is generally unavailable during explosive
eruptions when large quantities of SO_2 are released.

16: co-located 20: 7.3 is A?o?3 21: insert an ?between in and brightness
22: 8.6 is A?o?1 23: containS 27: I? give Realmuto (1994, 1997, 2000) credit for this observation.

Correction replaced with Realmuto (1994, 1997, 2000).

11865 9: Change ?sto ?as been'
11866 12: Recast the sentence ending ?eference forward model
14: ?everalis vague, please provide more detail.

The section describing RTTOV has been reworded to make the approach more clear.

11868 1-4: The sentence that starts ?he spectra of both: : :is impenetrable, please recast.

The section have been rewritten.

11869 2: degreeS 13: define DISTORT
added:
DIscrete Ordinates Radiative Transfer (DISORT, Stamnes et al 1988)

11872 9: Recast sentence starting ?he new error: : :.

Section rewritten

11875 17: remove carriage return
18 and onwards: The section that starts ?he Fig 3. : : :is very poorly written and should be thoroughly checked and adapted until it makes sense. It? quite an important paragraph that covers a lot of ground, detailing the sensitivity and issues with the retrieval.

Paragraph rewritten

11876 3: ?roughshould be ?hrough
4-5: proof read and formalise these lines

'Assuming a known plume altitude, and converting pressure values into km

trough the standard atmospheric profile, the values of the minimum error obtained are around two DU for plume centred at 1.5 km and go to 1 DU (and less) for 3 km altitude (and more) arriving to 0.25 DU at 11 km.'
Changed to
'Using the local covariance matrix and assuming a known plume altitude,
the errors for low SO2 amounts are as follows: 2 DU for plumes centred at 1.5 km, 1 DU for 3 km altitude, and these decrease further with altitude to 0.2 DU above 11km. Using the global covariance matrix the errors in SO2 amount are slightly higher. '

16 and onwards: The paragraph starting 'Moreover: : :is not clear at all, please recast.

Paragraph rewritten, in particular:

Note that when the plume is near the tropopause there can often exist two altitudes at the same temperature (either side of the tropopause) with very similar transmittances to the TOA. This results in a large error in retrieved plume altitude but does not significantly affect the retrieved SO2 column amount. In this situation the volcanic plume is well above the tropospheric boundary layer, which contains the majority of water vapour (that is the main cause of extinction in the ν_3 band), and so the SO2 signal does not varying between these layers with the same temperature. As a consequence, these layers are indistinguishable and the retrieved altitude depends strongly on the a priori and first guess. Still, the amount of retrieved SO2 is reliable.

11877 What are the justifications for the choices of optical depth and particle size for the aerosols in the synthetic spectra?

Values of optical depth have been extended, so they now cover the full range from optical thin to opaque.
Effective radius between 1 and 3 micron are the typical values that we retrieve for the Eyja eruption from AATSR and SEVIRI using the same optical properties that have been assumed in these simulations. (Siddans 2011, Grainger 2012).
These comments have been added to the text

1-4: not clear at all, please rewrite
7 and onwards: this section is particularly poor please rewrite this section through to line 23
27: presented 27 onwards: this sentence is also impenetrable

Section rewritten

11878
10: Add reference to Kearney and Watson, 2009

added

20: and onto the next page: I? not convinced that Zehner? categorisation of the eruption is widely accepted.

I think you should carefully read the JGR special issue, focusing on the papers by the volcanologists (esp. Thor Thordarson) for a more adroit analysis.

As far as we are aware, Thordarson is the person who wrote the description of the eruption in the report edited by Zehner.
Stevenson JGR et al 2012 (Thor Thordarson co-author) use similar classification for the description of the tephra dispersion:
phase1 : 14-18 April most powerful one, highest plume (12 km), discarge rate 5-10 10^5 kg/s
phase 2: height and discarge were much lower (4-10 km)
thirt phase 5-18 May
Peterson JGR 2012 divide in phase 1 (14-18 April), phase 18 April-3 May), phase e 3-17 May) and final phase 18-22 May.

15-16: where did these numbers come from?
References added (see comment on Referee1)

17-23: rewrite
24: Makes no sense

Paragraphs rewritten

28: Chronologise references: : :.
ok
11880 1: Chronologise references: : :.
ok
6-onwards: This section is also very weak.
Much of this has been approached in the literature, mostly in the JGR special issue. The section should be much better referenced to reflect this. It is also badly written: : :

Section have been changed and integrated with the addition of JGR papers (Petersen et al 2012, Stevenson et al 2012)

Some specifics: Meteorology is affecting the plume at all times, not just after the first 48 hrs. SO2 production during ?hase 1is NOT zero (see Fig 9). I? not convinced there? a ?ew injectionof SO2 most likely the eruption has run out of ice/water for scrubbing.

We agree with the referee about the ice/water interaction:
'The fact that there isn't any SO$_2$ detectable close to the volcano can be because the volcano itself is not emitting SO$_2$ or because, in this phreatomagmatic phase, the interaction with water deplete the SO$_2$ at the source.'

11881 1-9: The first paragraph is unreadable

Paragraph rewritten, in particular:
'Phase III starts during the morning of the 4th May with a strong
increase in the the \chem{SO_2} emitted. Between the 4th and the 8th
May the plume overpasses the western part of Europe, in particular the
UK, Ireland, France and Spain. Unfortunately a missing orbit in the
IASI dataset, towards the western side of the region shown, meant a

relatively large number of missing values had to be filled. Hence the relatively large amount of \chem{SO_2} seen on the morning of the 8th May in Figure 9 has to be treated cautiously. On the 9th May the \chem{SO_2} plume is located over the Atlantic (reaching the coast of Greenland). On the 10th, a filament of the plume passes over Southern Europe, starting over Spain and spreading to the east / north-east. On the morning of the 11th, a fresh plume passes over Ireland, moving towards mainland Europe. At the same time, an older and more dilute plume travels north-east (reaching Scandinavia on 12 May). A new \chem{SO_2} plume from the volcano is again transported over Europe on 14 May, 16 and 17 May.'

Additional refs:

The suggested references have been added

Carn, S.A., Krueger A.J., Bluth, G.J.S., Schaefer, S.J., Krotkov, N.A., Watson, I.M., Datta, S, 2003, Volcanic eruption detection by the Total Ozone Mapping Spectrometer (TOMS) instrument: a 22 year record of sulfur dioxide and ash emissions, Volcanic Degassing (eds Oppenheimer, Pyle and Barclay), Special Publication of the Royal Society, SP213, pp. 177- 203

Henney, L.A., Rodriguez, L.A., Watson, I.M., A comparison of sulphur dioxide retrieval techniques using mini UV spectrometers and ASTER Imagery at Lascar volcano, Bulletin of Volcanology, 2012, Volume 74, Number 2, Pages 589-594

Kearney C., Watson I.M., An ash correction for the 8.6 micron SO2 retrieval, Journal of Geophysical Research, doi:10.1029/2008JD011407

Realmuto VJ, The potential use of Earth Observing System data to monitor the passive emission of sulfur dioxide from volcanoes, Geophysical Monograph 116, 101-115, 2000.

Realmuto VJ, Sutton AJ, Elias T, Multispectral imaging of sulfur dioxide plumes from the East Rift Zone of Kilauea Volcano, Hawaii, J Geophys Res, 102, 15,057-15,072, 1997.

Realmuto VJ, Abrams MJ, and Buongiornio MF, and Pieri DC, The use of multispectral thermal infrared image data to estimate the sulfur dioxide flux from volcanoes: a case study from Mount Etna, Sicily, 29 July 1986, J Geophys Res, 99, 481-488, 1994.

Symonds, R.B., Rose, w.I., Bluth, G., and Gerlach, T.M., 1994, Volcanic gas studies: methods, results, and applications, in Carroll, M.R., and Holloway, J.R., eds., Volatiles in Magmas: Mineralogical Society of America Reviews in Mineralogy, v. 30, p. 1-66.