We would like to thank all four referees for their useful comments and corrections. The first three referees had mostly minor corrections. The fourth referee had more comments, (s)he questions in particular the advantages of the new retrieval algorithm and the fact that new retrieval results differ from previous/other retrieval results. Below we explain in detail why the new algorithm is inherently more accurate than previous retrievals. Part of the differences observed with previously published results for total mass estimates was due to a subtlety in the specific gridding technique we employed, and for this revision we reverted back to our old gridding technique.

We illustrate our retrieval algorithm on two well studied test cases, namely the Kasatochi and Sarychev eruption. While these examples are not meant as validation, they illustrate the many advantages of the new method. We have expanded the discussion of these example retrievals (including the addition of an extra figure on the total mass time series of the Kasatochi plume), and discussed possible differences with previous published results.

We believe we have adequately addressed all comments and corrections in the included revision. Please find below a detailed reply to each them. Our reply is in blue, referee comments in black.

Referee #1

This is an important paper as the SO2 product described here is used by the VAACs to advise the aviation industry. The paper gives the science behind this product. In general the paper is well written and I only have a few minor comments given below.

(i) Page 7244 last parag. As the author states the optimal estimation retrieval is more expensive than the method proposed here but nevertheless with modern supercomputers it is still feasible to run this in real time as is already done for ash retrievals. One point in favour of not going to the expense of an OE retrieval is in this case the retrieval only weakly depends on the a-priori. If one was to try and retrieve SO2 profiles however then OE would be more advantageous as the background water vapour profile would be important. This point could be made here. Indeed retrieving the height of the SO2 plume would be of great interest.

We have now made this point explicitly, adding the following sentence: “These are time consuming, but have the advantage of fully exploiting the spectral resolution by simultaneously retrieving competing species (e.g. H_2O) and potentially extracting plume altitude information.”

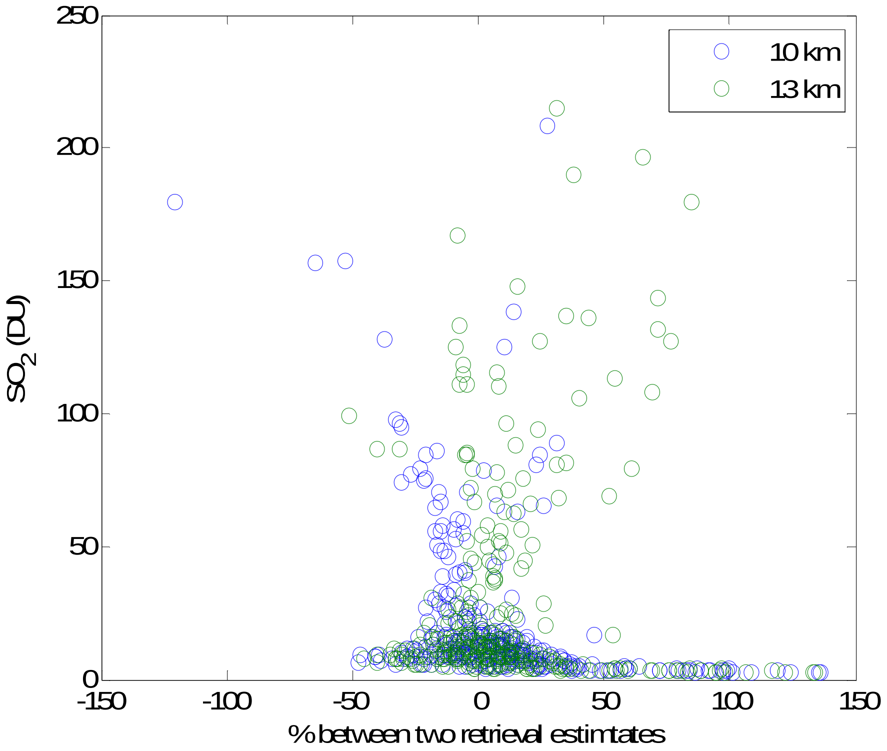
(ii) Page 7246 line 6 it is not usual to refer to IASI channels as microchannels. I suggest using the former.

Corrected

(iii) Page 7248 Last parag. I think a bit more detail on this dual estimate retrieval would be helpful with an example to show the differences between them. Note some users may find both solutions useful so they can decide in their own analysis systems.

In the plot below we have illustrated the differences on retrievals from 15 June 2009 (Sarychev eruption). In the paper we have now added a more general description, explaining the typical differences which can be expected:

“Also note that we find two estimates u_1 and u_2 for u, for each set of absorption and background channels. Theoretically, these two estimates should only agree when the assumed altitude corresponds to the real altitude (because the corresponding brightness temperature differences have a different pressure and temperature dependence). From looking at a few test cases, the two estimates generally agree well between 25 DU and 75 DU (with a standard deviation of around 10%). On either side of this range, differences increase, with the u_1 estimate superior for lower total column amounts and the u_2 estimate by construction superior for large column total amounts. When u_1 or u_2 exceed 100 DU, we used the u_2 estimate, otherwise u_1 was used. “



(iv) Page 7250 line 18. I found Figure 7 and the text here a bit confusing. What does "retrieved total masses (as a percentage of the maximum)" mean? Please clarify.

The idea is to illustrate the sensitivity of the total mass estimate to the assumed altitude. For that reason, we have normalized the total masses with the maximum measured mass (the one at altitude 5 or 7 km). The formulation wasn’t very clear, so we have now changed “(as a percentage of the maximum)" to “(as a percentage of the maximum measured total mass for a given altitude)”

(v) Page 7254 line 22 should read "...erupted, with all three releasing large amounts..."

Corrected, we simplified it to “..erupted, each releasing large amounts..”

(vi) Page 7271 Fig 8 The total column water vapour is presumably very different for these two cases also which explains some of the differences.

Yes, this is definitely the case. We have added this in the caption.

(vii) Page 7272 Fig 9 The units should be added to the legend.

These have been added.

(viii) Page 7275 Fig 12 I am not sure this will show up very well in a published paper.

We have remade this figure, added dates, changed the colorbar and made the geographical area a bit smaller, so that the plumes are better visible.

Referee #2

In this work a novel algorithm for the volcanic SO_2 retrieval above 500hPa using IASI measurements is described. The most important and original results of this paper is the possibility to retrieve total columnar abundances ranges over 4 orders of magnitude (from 0.5 to 5000 DU) with an extremely low theoretical uncertainty (<5 %) and a near real time applicability. A sensitivity analysis has been also carried out to estimate the retrieval errors due to the uncertainties of measurements errors, volcanic cloud altitude and plume aerosols (ash and ice).

The analysis seems methodologically correct and the results clearly presented.

I recommend the publication after minor revisions and corrections outlined below:

Page 7250, line 18: what does it mean "as a percentage of maximum"?

The idea is to illustrate the sensitivity of the total mass estimate to the assumed altitude. For that reason, we have normalized the total masses with the maximum measured mass (the one at altitude 5 or 7 km). The formulation wasn't very clear, so we have now changed "(as a percentage of the maximum)" to "(as a percentage of the maximum measured total mass for a given altitude)"

Page 7253, line 18: why the 5 km retrieval has not been cited?

The total mass at 5 km wasn't cited here because having all the mass at that altitude is unrealistic for such a large eruption (and the corresponding total mass of 7 Tg is therefore unrealistic as well).

Page 7253, lines 21-22: clarify the sentence ": : :we find that the values further increase
after the 9th to about 1.7 Tg on the 12th".

We have now added the whole Kasatochi timeseries, which shows that the retrieved mass reaches its peak on the 11th August; rather than on the 8th as one would expect.

Page 7268, Figure 5: - avoid the overlapping between the upper left plates;
- check the multiplication factor for the y-axis of all plates (10ˆ2);
- use the same limits (max and min) for the relative errors plates color ramp.

Corrected

Page 7272, Figure 9: add the units on color ramp.

Added

Page 7275, Figure 12: add date and time for the different images.

Added

Referee #3

This paper describes an SO2 retrieval scheme for space-based infrared sounding instruments such as IASI on MetOp-A, designed for near real-time volcanic cloud measurements. It is a useful contribution that describes the theoretical basis of the algorithm, the applicable range of SO2 columns, the associated errors, and the sensitivity of the retrieval to ash and ice in the volcanic cloud. I can recommend publication after attention to the following mostly minor issues:

P7242, L7: 500 hPa is a pressure level not an altitude; convert to approximate altitude?

We have changed this now to "~ 5km".

P7242, L21-2: list references in chronological order (unless AMT policy dictates otherwise).

Corrected

P7243, L2: change 'space' to 'satellite', also on line 4.

Corrected

P7243, L4: can use 'IR' instead of infrared hereafter.

We now use IR for infrared and TIR for thermal infrared after these are introduced.

P7243, L13: the wording here suggests that TOMS is still making measurements, but the final TOMS mission ended in 2005.

The wording has been altered and now reads …TOMS and subsequent follow-up ozone monitoring instrument…

P7243, L21: change to 'For an overview of satellite instruments capable of: : :'.

Corrected

P7243, L25, 29: for the benefit of non-spectroscopist readers, it might be useful to give wavelengths for the SO2 absorption bands in addition to wavenumbers.

These have now been added in figure 1 (which has all the other information on the absorption bands)

P7244, L7: although it is pointed out later on, it would be worth also stressing here that all IR measurements require thermal contrast between the SO2 plume and the underlying source of radiation.

This has now been added at the end of the paragraph. The sentence reads: “Note that all TIR measurements require thermal contrast between the SO_2 plume and the underlying source of radiation.”

P7244, L21: ‘: : :using high spectral resolution instruments: : :’

Corrected

P7244, L23: I’m not sure that the time constraints are that significant these days – except when retrievals are required in ‘near real-time’ for hazard mitigation.

It is true that a full optimal estimation retrieval is within reach of current technology, but doing so for e.g. 4 years of IASI data or 10 years of AIRS data represents a very large number of computing hours, while it is a trivial task with the presented algorithm.

P7245, L21: need to explicitly state here that Tc is cloud temperature, and also that Ts is measured brightness temperature.

These have been added.

P7249, L20: there is a very minor discrepancy between the 0.15K error for channel set 1 given here and the 0.14K given in Table 1 as the standard deviation.

The larger of the two, 0.15 K has now been used everywhere.

P7251, L8: remove parentheses and use ‘quiescent’ or ‘passive’ instead of ‘quiescence’.

Corrected

P7251, L14: I think ‘uniformly’ should be ‘uniform’ – meaning no spectral dependence in ash absorption across the v3 band? The authors could also comment on the effect of ash composition here.

We changed ‘uniformly’ to ‘uniform’ and added a sentence on the ash composition: “Note that the specific extinction depends on the total ash loading but also on the particle size distribution and the mineral composition.”

P7251, L19-20: should be ‘optically thick’. ‘Lower lying thin to medium optically thick’ is a bit of a mouthful – perhaps replace with ‘Low-altitude aerosol layers of low-to-medium optical thickness...’?

Corrected

P7251, L21: by ‘close’ I presume you mean just below the SO2 cloud?

Yes, we have clarified this now.

P7251, L28: switch to wavelength here is inconsistent with wavenumber used elsewhere.

Corrected

P7252, L18-19: instead of ‘atmosphere’ I would use ‘UTLS’. There have been some large effusive eruptions that emitted large quantities of SO2 into the lower troposphere.

Corrected

P7253, L2-4: Full sensor names should be given, if not given earlier, and whether they operate in the UV or IR.

These have been added

P7253, L10: I think the Bobrowski et al. (2010) reference refers to GOME-2, not OMI SO2 columns.

This was indeed referring to GOME2 and has been corrected

P7253, L15: ‘injection altitude’.

Corrected

P7253, L24: ‘shear’.

Corrected

P7254, L17: ‘gridding’.

Corrected

P7254, L22: use ‘2011’ instead of ‘this year’.

Corrected

P7254, L22: change to ‘: : :each releasing large amounts of SO2.’

Corrected

Fig. 2: please also give equivalent altitudes for the pressure levels, for the benefit of volcanologists.

These have been added in the caption.

Fig. 5: the parts of this figure overlap a bit and need some adjustment. Also, I think the ‘_10ˆ4’ on the pressure axis should be ‘_10ˆ2’

Corrected

Fig. 9: label the color bars (I presume it is SO2 column in DU). It is also not clear what altitude is assumed for the displayed SO2 columns?

The color bars have been labeled and the caption now indicates which altitude was assumed for the display (10 km).

Fig. 11: surely this image shows more than just the ‘maximum observed SO2 columns’ for the 20 May – 30 June 2011 period, as stated in the caption. It seems to be a composite of all IASI SO2 retrievals in this period.

The figure is a composite, in which the value for each grid cell equals the maximum observed SO_2 columns in that grid cell for the given time period. We have now clarified this in the caption.

Fig. 12: could dates be provided for each panel in this figure?

Dates have been added.

Referee #4

General Comments

I do not think that this manuscript is ready for publication, and recommend that the authors revise and resubmit the manuscript for another review. My concerns and comments are detailed in the following Specific Comments.

The virtues of the new retrieval procedure are not obvious. The authors claim that the new procedure is faster to execute, but present no comparisons of running time. In the cases were the authors compare results from the new and original procedures we see that the results are significantly different. Since the authors do not discuss these differences (in any detail), we can’t decide if the trade-off between faster processing and accuracy favors the new procedure. The authors need to spend more time on the validation of the new procedure or restrict this manuscript to a discussion of the theory behind the new procedure.

The presented procedure is both faster and more accurate than previous procedures. Optimal estimation methods are time consuming (easily 30 seconds per retrieved column), while the computational cost for the presented LUT approach is negligible (30 seconds for a complete day of 1.3 million IASI measurements). The procedure that was applied in e.g. Haywood et al (2010) uses a hybrid method, where the first stage uses optimal estimation, and is therefore inherently slower.

We believe that the presented method is more accurate precisely because it does not rely on optimal estimation. Such methods have the disadvantage of relying on an a priori and have their own sources of errors, such as the difficulty of retrieving concentrations from observations rich in aerosols or clouds. In the two step approach of Haywood et al (2010), the first step relies on the selection of a representative set of measurements to determine the average absorption coefficients c. The choice of a representative set of measurements is partly subjective. Also, such an approach makes the assumption that one given altitude always correspond to the same P,T pair, which is not necessarily the case for plumes that are spread out over a large part of the Northern Hemisphere. In the new approach such an assumption does not need to be made. For these three reasons the new approach is inherently more accurate.

The examples were not meant as validation, but were merely meant as illustrations. The differences between new and original procedures are not significant on a single measurement basis. One of the main reasons for the differences presented in the Sarychev time series was due to the use of a new gridding routine, which we thought dealt better with the calculation of masses from overlapping orbits. On investigation, this new gridding routine did introduce a general overestimation of masses, especially for long elongated aged filaments. In this revision we have therefore gone back to our original gridding procedure for the calculation of total masses. The timeseries of Sarychev new and original are now in much closer agreement (10-20%). Of course the use of a new gridding routine was not discussed in the manuscript and so there was no way the referee could have known this, but we hope that this addresses his/her main concern.

For Kasatochi, it is very hard to compare with the original published IASI retrievals since these used the weak v1+v3 absorption band (which has very different dependencies on altitude). One key advantage of the present algorithm is precisely the fact that we do not need to use this band (which is particulary noisy for IASI and can only be used in daytime) anymore to get reasonably total mass estimates even for extremely saturated plumes. Also given the scatter in the total mass estimates from other sounders and the fact that we deal with a multilayered sheared plume, we believe that the presented total mass estimates for Kasatochi are reasonable, and demonstrate the versatility of the new algorithm. In order for the readers to judge better whether the new algorithm is superior to the old one, we have now also added a figure of the total mass timeseries for Kasatochi, which shows the original retrievals from Karagulian et al, 2010, the new retrievals and the retrievals from Krotkov et al 2010. The new retrievals clearly fits the expected exponential decay better than the original retrievals and also overall compare better with the OMI timeseries.

Specific Comments

(1) 7242/2: Delete “and popularity”

Corrected

(2) 7242/4 – 5: Replace “primarily designed for weather forecasting…” with “” (including the definitions of the acronyms). Also you can delete “therefore often”

Corrected

(3) 7242/16 - 19: Here you cite studies showing that anthropogenic emissions of SO2 are 5 to 6X larger than volcanic emissions. However, you state in the Abstract (7242/6) that “most of the observed SO2 is found in volcanic plumes.” Are you saying that most of the anthropogenic emissions are not observed by satellites? Do you have any explanations why the anthro emissions are not observed?

“most of the observed SO2 is found in volcanic plumes” from the abstract refers to infrared sounders which have limited sensitivity to boundary layer SO2. A second reason why not all anthropogenic emissions are observed (even with UV instruments) is the fact that anthropogenic emissions are spread out in time and space and the ambient columns are often too low to be observed.

(4) 7243/5 – 12: This set of citations does not need summaries of the research topics. For example, the citation for Clarisse et al. (2011b) does not need the summary “found on average 8 major events of … Asia.” Remove the summaries for each citation to improve the readability of this section.
In addition, replace “it can be done in near real time…” (Line 9) with “when data are available in near real time…”

Corrected

(5) 7243/21: Here you mention that the "typical" IR footprint, is "smaller." What instruments are you comparing? TOMS? OMI? Also, you might want to say "higher spatial resolution" rather than "smaller footprint."

This is valid for the UV instruments most commonly used for SO2 sounding (TOMS, OMI, GOME2). We have changed "smaller footprint" to "higher spatial resolution".

(6) 7244/1 – 2: This statement that water vapor absorption is not important in the 800 – 1200 cm-1 region is not correct. While there are no strong water lines in this region, continuum absorption can be significant (10 – 30%).

The sentence read "…water vapour is not *that* important here…", refers to retrievals using the v3 band (sentence before). We have now made this clearer by replacing "that" into "as", it now reads: "…water vapour is not *as* important here.."

(7) 7245/6 – 8: This description of IASI is too brief. I suggest using the description from Clarisse et al., 2008 (the Jebel at Tair paper). In particular, the distinction between the spectral and apodized resolution and NEDT in the v1 and v 3 bands of SO2 are important topics for this paper (see below).

We agree, we have now added this information.

(8) 7245/18: At what altitude is the absorption of "other species" negligible? I assume that you are talking about water vapor, but you used the plural "species." What species (other than H2O and SO2) are incorporated in your model?

In our forward model we have included the standard (US 1976 atmosphere) of the most important trace (O3, CH4, CO2, …). It is true that water vapor is the most significant competing absorber in the spectral range we use, but formula (1) is a general formula which is valid under the stated conditions (regardless of spectral range).

(9) 7246/4: Replace "convoluted" with "convolved"

Corrected

(10) 7246/5 – 24: This discussion of pressure effects, together with Figure 2, is confusing. Based on your original definition of the absorption coefficient (Eq. 2), are you saying that Beer's Law is not valid at (very) low pressures? Similarly, you state that the (Beer's) absorption coefficient is a function of pressure, temperature, and column abundance (Line 1). Why then do you have problems fitting the (simulated) IASI brightness temperatures?

The main point is that Beer's Law is not valid on the level of IASI channels due to effects of instrumental line shape and apodisation as stated in lines 18-21. This effect is most pronounced for low pressures where the pressure broadening is not so important..

Regarding Figure 2, why are the brightness temperatures (BT) at 0 DU different for the high and low temperature plumes? Did you use the same atmospheric profiles (i.e. pressure, temperature, and water vapor) to simulate the IASI BT for the low- and high-pressure cases?

These two cases were taken at random from our set of forward simulations, and use a different atmosphere.

Rather than saying that the centers of spectral lines saturate more rapidly than spectral wings (Lines 14 and 15), I suggest that you say that the centers saturate at lower concentrations than do the wings (or something to this effect).

Corrected. The sentence now reads: “At very low pressure, spectral lines saturate at a lower concentration at their line centers than their wings.”

To address the effects of low pressure, you introduce an “explicit column dependence” for the absorption coefficient (Lines 21 - 23), but how is this definition different from the original definition (Line 1)? In Figure 2 the absorption coefficient (“c”) is defined in reciprocal Dobson Units (DU-1), suggesting that this “c” is the explicit column dependence rather than the absorption coefficient.

The absorption coefficient in the text and in Figure 2 are one and the same (they are both labeled c). Figure 2 shows the necessity of making c depend on the column u and not just P and T.

(11) 7247/5-16: The concept of “channel sets” needs to be clarified. As shown in Table 1, there are two sets of channels with each set consisting of two “absorption” channels in the □3 band of SO2 and two “background” channels outside of the □3 band. Fig. 3 is supposed to show plots of the BT for the two sets of absorption channels which should, by definition, include 4 channels. However, only two channels are shown in Figure 3. So what is the distinction between channels and channel sets? Table 1 contains the BT differences between the □3 and background channels for the two channel sets. There is only one BT difference per channel set, suggesting that two IASI channels are required to describe an absorption or background channel..

Figure 3 uses in fact both absorption channels, but for simplicity, and because for the two sets the wavenumbers are close together, the caption only mentioned one. We have added a tilde ~ in front of the wavenumbers in the legend of figure 3 and the caption to make this clear. The text in the caption makes clear that what is plotted is the estimation of the absorption using both channels.

For each channel set the absorption channel pairs are separated by 0.25 cm-1 (Table 1). Given that the IASI spectra are apodized with a filter width of 0.5 cm-1, how distinct are the pairs from one another? The background channel pairs are separated by 0.5 cm-1 and this seems to be the minimum separation for distinct channels.

The algorithm uses two channels to estimate Ts (instead of just one) and two channels to estimate Tucb. This doubling of channels was done to reduce instrumental noise and to approach the required Ts=Tucb in the absence of SO2 as good as possible. Using only one channel significantly increases the standard deviation between these two. This was not mentioned explicitly in the manuscript and we have added this now. Note that two adjacent IASI channels can be significantly different, even considering the spectral resolution is 0.5 cm-1.

Finally, the mean BT differences for channel sets 1 and 2 are -0.05 and 0.05 K, respectively (Table 1). Such high temperature resolution would not be possible if the NEDT of IASI was 0.2 K, as stated in this paper. As noted in Clarisse et al. (2008), however, the NEDT in the vicinity of the □3 absorption band is 0.05 K. This superior NEDT needs to be made explicit in the current paper.

Yes this is correct, following comment (7) the NEDT at the nu3 band has now been added.

(12) 7247/17 – 20: For the sake of completeness, your model is based on the assumption that there is no absorption or emission between the top of the plume and the sensor. Similarly, your assumption that the water vapor above a plume is colder than the plume is valid below the tropopause, but what about plumes in the lower stratosphere? There is not likely to be much water vapor at such altitudes, but you should at least consider stratospheric plumes when stating this assumption.

We have now added one sentence clarifying this: "..so disregarding significant water vapor above stratospheric plumes.."

(13) 7248/11: Replace "too much" with "overly"

Corrected

(14) 7248/14 – 22: There is no reason to list these 4 PT pairs, as you have not plotted them in Figure 5 (or so I gather from your text). Regarding Figure 5, it is difficult to discriminate the colors of the dots. Either make the dots a lot bigger or use contours. The contour approach would result in continuous data fields, but the relative changes in the absorption coefficients and errors would be much easier to see.

We have removed the 4 PT pairs. While preparing the original manuscript we have tried the contour approach, but this didn't give a satisfactory representation. Enlarging the dots is not really an alternative as they would start to overlap for low pressures. We believe that the current presentation allows to see clearly the range of errors involved, even though this means that small differences cannot be distinguished. Following the comments of the other referees we have remade the plot slightly, avoiding overlap between the different panels and colorbars.

The relationship between absorption and pressure at 10 DU is the inverse of that at 750 DU. For 10 DU the absorption decreases with a decrease in pressure while at 750 DU the absorption increases with decreasing pressure. Is this behavior typical for absorption coefficients or unique for your "column-weighted" absorption coefficient?

This effect is solely due to the choice of channel sets (the two diagrams at 100 DU show the same dependence).

Finally, errors of 3 – 5% are rather large for simulated observations in model atmospheres. You will certainly compound these errors when real observations are used.

First note that 3 – 5 % are the upper end of the errors, especially for the first channel set the error is often not larger than 1 %. Secondly, we think these errors are quite low given the fact that the absorption coefficients are averages from potentially very different atmospheres (tropical vs polar); and a given PT pair can correspond to very different altitudes at different locations.

How will these errors propagate into your retrievals? Why did you not discuss these modeling errors in Section 3?

These errors do not propagate as they are made in the final step (the step of the conversion). The other errors will propagate, and increase by 3-5% following that error. Since this was discussed in this section, they were not discussed again in Section 3 (they were explicitly mentioned though).

(15) 7249/9 – 10: Clarify the second category of error. Are you saying that the cloud temperature (Tc) can be measured directly?

Optimal estimation retrievals can estimate plume altitudes as for instance demonstrated in the paper on the Jebel at Tair eruption (Clarisse et al. 2008).

(16) 7249/19 – 20: Regarding the estimates of measurement error, estimated (Table 1) to be 0.15 and 0.25 K for the channel sets, we once again need to consider the issue of NEDT (comment #11). The estimated errors are less than or equal to the quoted NEDT of 0.20 K and therefore would be negligible. However, based on the revised NEDT of 0.05 K in the vicinity of the □3 absorption band, the estimated errors are significant.

They are larger than the NEDT in this spectral range because the standard deviation of the differences also includes differences due to the influence of other atmospheric parameters (e.g. water vapor profile).

(17) 7250/5: In addition to the random error discussed here, the mean differences from Table 1 indicate that there will be a negative bias (-0.05 K) in channel set 1 and a positive bias (0.05 K) in channel set 2. These offsets are small (~ NEDT), but could lead to over-estimates of SO2 for channel set 1 and under-estimates in channel set 2.

This bias is removed in the calculation of brightness temperature differences. This was omitted from the manuscript but has now been added in the discussion following equation (3).

(18) 7250/15: I think that you want to say "… the assumed water vapour above the plume …"

Corrected

(19) 7250/20 – 28: This section, together with Figure 7, needs some work. Figure 7 is very difficult to read as the plot lines are too thin and it is hard to discriminate the colors. In particular, the colors used to represent Merapi and Okmok appear to be the same color. You could illustrate the relationship between cloud altitude and apparent SO2 loadings with the results from a single eruption as the differences between the eruptions are due to differences in the local conditions that are secondary to this relationship.

We have remade figure 7 using thicker lines and different colors. We have chosen to keep the different eruptions as it is nice to illustrate the 'secondary' dependence.

I think that your discussion of the relationship between cloud altitude and apparent SO2 would be clearer if described in terms of the temperature contrast between the cloud and background (Tucb – Tc). Since the main source of upwelling radiance (in the vicinity of the □3 feature) is water vapor we can assume that Tucb corresponds to an altitude of 5 – 7 km. At cloud altitudes between 5 and 7 km the temperature contrast is low (Tc ~ Tucb) and the maximum amount of SO2 is required to produce the observed absorption. For clouds at the tropopause the temperature contrast is highest (Tc << Tucb) and the minimum amount of SO2 is required to fit the absorption. In the stratosphere the SO2 retrievals increase as Tc approaches Tucb, with the rate of increase controlled by the stratospheric temperature gradient.

We agree, and have now replaced that paragraph with a paragraph which follows very close your wording.

(20) 7251/1 – 6: Explain how c(T,P,u) and Tc "cancel" each other.

This sentence was confusing and not always true, so we have removed it.

The retrievals for low altitude clouds are more sensitive to cloud height due to the steep temperature gradient in the troposphere.

We have added this.

Does Figure 7 illustrate the estimation of cloud altitude from Ts (I don't think so)? If you do not use this sort of estimation in the work presented in this manuscript then I suggest deleting this sentence.

We have removed this sentence.

(21) 7251/8: I suggest deleting the first part of this sentence. A large short-lived eruption could release as much, if not more, SO2 as passive degassing over a longer period of time. Rather than debate over the meaning of "majority," just start with "Large eruption plumes contain…"

Corrected

(22) 7251/12 - 20: Figure 8 is very difficult to read. Do any of these plots represent actual observations (as implied in the text) or model spectra? Given that the ash/ice loadings are given in terms of extinction and "saturation," rather than abundance, I'm guessing that these are model spectra. I strongly suggest that you replace this figure with plots of brightness temperature differences (BTD), so that you can isolate the spectral features of interest for the benefit of your readers.

These spectra are observed spectra. To avoid confusing this has now been stated explicitly in the caption, so that a plot in BTD is not possible.

Define "saturation." Note that saturation has very specific definition in atmospheric chemistry. Are you talking about the abundance of ash or ice at which the cloud becomes completely opaque? Note also that you have labeled two spectra as "near saturation" in the Ice panel (Fig. 8).

Yes it refers to abundance, where the layer behaves like a totally absorbing black body. For the ash example we almost have a perfect black body at ~220 K with very little trace gas absorption (the ozone feature around 1000 cm-1 is present due to ozone absorption above the plume).

Finally, your contention that ash layers beneath SO2 clouds do not affect the SO2 retrievals implies that the impact of ash on the absorption and background channels (Table 1) is identical. Can you demonstrate that the impact is identical?

We have shown this using forward simulations as stated in on line 1 of page 7252 of the original manuscript.

(23) 7253/7-8: The wide range in reported SO2 loadings may also be due to the fact that most retrievals were based on a single cloud altitude, when we know that Kasatochi clouds were at two or more altitudes

We have added this note.

(24) 7253/15 – 20: It is not clear how your algorithm deals "more efficiently" with saturated bands. If the v3 bands are saturated then the cloud is opaque you are not getting any information on Tucb. For opaque clouds the optical paths you are sampling do not pass through the cloud, and any retrievals would be under-estimates. As noted on Pg. 7252 your algorithm will skip over opaque pixels since the

Tucb measured in a background channel will be approximately equal to the Ts measured in an absorption channel.

The comment on opaque pixels on page 7252 was referring to completely opaque ash layers. For SO_2, the fact that we use absorption channels at the edge of the SO2 (set 2) prevents exactly saturation problems (which is why we can theoretically measure loadings up to 5000 DU).

On a related note, your new estimates of total mass are up to 2X larger than the previous IASI-based estimate (Karagulian et al., 2010). Given that the previous estimates were based on a more rigorous optimal estimation technique, can you account for these differences (remember that you are likely to be under-estimating or ignoring pixels with the highest SO2 abundance)?

We disagree. The original estimate was 1.7 Tg. We measured maximum loadings of about 1.6 Tg and are therefore in good agreement. Like we mentioned in the reply to your main comment, they are several strong reasons to suggest that the new retrievals are superior (see argumentation in the introduction).

(25) 7253/20 - 25: How valid are the retrievals at 10 km? You know that there is SO2 at altitudes above 10 km - in fact you have found SO2 at altitudes up to 25 km.

The mean value of 10 km was taken from Krotkov et al 2010. As far as we recall we do not state that SO_2 was found as high as 25 km.

One of the principal assumptions for your forward model is that there are no absorbing species between the cloud and sensor. The 10-km case violates this assumption so you need to defend your choice of this altitude.

The absorption above 10 km for the selected channels is below the IASI noise level as can be demonstrated with forward simulations. As stated in the description of the algorithm, residual water vapour absorption is accounted for.

(26) 7254/12 – 15: Your contention that the differences between the new Sarychev time series (generated with the new retrieval procedure) and the original time series (Haywood et al., 2010) are minimal is not correct. On Day 25, for example, the new estimate for total mass is over 60% larger than the original estimate (Fig. 10). Once again you show large differences between the new and original estimates (Comment #24) with no explanation for the discrepancy.

As explained above, we have gone back to our old gridding routine for the calculation of total masses and the updated time series is now in very good agreement with the original time series. The total mass is estimated somewhat lower (~0.9Tg), but not incompatible with the estimate from Haywood et al, 2010. Also note that the new retrievals are much more stable for the first week after the eruption. The original retrievals had differences up to 100% for consecutive overpasses, and were therefore much more noisy. This is another indication of the robustness of the new algorithm.

Figure 10 indicates that there are 60 days in June! I think that you want to change the axis title to “Days Since Eruption.”

We have changed the caption to “Days since 1 June 2009”.

(27) 7256/13 – 15: You have presented an implicit comparison of the new and original retrieval algorithms by including results from the original algorithm in this manuscript. The new results are significantly different from the original results for Kasatochi and Sarychev, calling attention to the need for validation. If you intend this manuscript to be a purely theoretical exercise, then stick to model/synthetic results and remove the applications to actual IASI observations. As you note, a thorough validation of the new procedure will require a new, dedicated paper.

By reverting to the old gridding routine the results for Sarychev are in close agreement with the original results (especially for 20 June – 10 July). For Kasatochi, the total mass estimate is in good agreement with earlier published results, while the timeseries is now in much closer agreement with independent results from OMI. In response to your comments we have now expanded the discussion of differences between original and current retrievals of both the Sarychev and Kasatochi eruption. For Kasatochi, we thought it would be useful to show the full timeseries for IASI (which is not shown in Karagulian et al. 2010) and also to compare it with OMI measurements. While both examples are meant as an illustration, rather than a validation, they should convince the reader that the new retrieval algorithm is in all aspects superior to the previous approaches.