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.

Specific Comments

(1) 7242/2: Delete "and popularity"

(2) 7242/4 – 5: Replace "primarily designed for weather forecasting…" with "such as AIRS, TES, and IASI" (including the definitions of the acronyms). Also you can delete "therefore often"

(3) 7242/16 - 19: Here you cite studies showing that anthropogenic emissions of SO_2 are 5 to 6X larger than volcanic emissions. However, you state in the Abstract (7242/6) that "most of the observed SO_2 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?

(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…"

(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.”

(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%).

(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 ν1 and ν3 bands of SO_2 are important topics for this paper (see below).

(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 H_2O and SO_2) are incorporated in your model?

(9) 7246/4: Replace “convoluted” with “convolved”

(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?

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?

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).

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.

(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 ν_3 band of SO_2 and two "background" channels outside of the ν_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 ν_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.

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.

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 ν_3 absorption band is 0.05 K. This superior NEDT needs to be made explicit in the current paper.

(12) 7247/17 – 20: For the sake of completeness, your model is based on the assumption that there is no absorption <u>or emission</u> 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.

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

(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.

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?

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

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

(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 ν_3 absorption band, the estimated errors are significant.

(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 SO_2 for channel set 1 and under-estimates in channel set 2.

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

(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 SO_2 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.

I think that your discussion of the relationship between cloud altitude and apparent SO_2 would be clearer if described in terms of the temperature contrast between the cloud and background ($T_{ucb} – T_c$). Since the main source of upwelling radiance (in the vicinity of the ν_3 feature) is water vapor we can assume that T_{ucb} corresponds to an altitude of 5 – 7 km. At cloud altitudes between 5 and 7 km the temperature contrast is low ($T_c \sim T_{ucb}$) and the maximum amount of SO_2 is required to produce the observed absorption. For clouds at the tropopause the temperature contrast is highest ($T_c << T_{ucb}$) and the minimum amount of SO_2 is required to fit the absorption. In the stratosphere the SO_2 retrievals increase as T_c approaches T_{ucb}, with the rate of increase controlled by the stratospheric temperature gradient.

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

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

Does Figure 7 illustrate the estimation of cloud altitude from T_s (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.

(21) 7251/8: I suggest deleting the first part of this sentence. A large short-lived eruption could release as much, if not more, SO_2 as passive degassing over a longer period of time. Rather than debate over the meaning of "majority," just start with "Large eruption plumes contain…"

(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.

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).

Finally, your contention that ash layers beneath SO_2 clouds do not affect the SO_2 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?

(23) 7253/7-8: The wide range in reported SO_2 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

(24) 7253/15 – 20: It is not clear how your algorithm deals "more efficiently" with saturated bands. If the ν_3 bands are saturated then the cloud is opaque you are not getting any information on T_{ucb}. 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 T_{ucb} measured in a background channel will be approximately equal to the T_s measured in an absorption channel.

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 SO_2 abundance)?

(25) 7253/20 - 25: How valid are the retrievals at 10 km? You know that there is SO_2 at altitudes above 10 km - in fact you have found SO_2 at altitudes up to 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.

(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.

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

(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.