Response to the reviewer

We are very grateful to the reviewer for the series of peer reviews in this article. The answers to the reviewer's comments are as follows.

The revised version of the manuscript is clearly improved and most of the referee comments were considered in the revision. Some changes were not applied to the whole manuscript (for example the words "optimal" or "best" were mostly removed, but remain in the abstract, in the conclusions and in some other places; also the update to the use of average RMS instead of sum of RMS is not done all along the manuscript) and some specific comments remain.

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In the revised manuscript, all "optimal" and "best" have been removed. We also tried to avoid similar expressions.

A major concern remains about the goal of this work and how exactly future scientists can rely on the conclusions done in the manuscript. The precise goal of this work is not clearly stated, which makes it difficult to make the link with the conclusions. If I understand it correctly, the goal of this work is to allow selecting a refractive index for an eruption, using IASI (or other TIR) observations. From that selection, one can then get information on the particle composition.

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Since the refractive index (RI) models used in this study are linked to ash chemical composition, we had to include consideration of the composition when discussing the results of individual volcanic plumes. However, it is not easy to estimate the composition of volcanic ash directly from observations, and the discussion in this paper is limited to a general consistency (mafic or felsic).

The goal of this work is described in some parts of the revised manuscript (Line 56-57, 70-72, 1095-1096). In particular, the primary purpose is to investigate whether the RI model estimated from the sample composition and/or the SiO2 content of the volcano can well explain the observed brightness temperature spectra (BTS). If the BTSs of the observations and simulations match within an acceptable error, it is expected that the same RI model can be used for the imager analysis to perform an accurate estimation for VAC parameters. This paper does not mention the imager analysis because it is not

performed, but the ultimate goal is to improve the accuracy of imager analysis.
We think there is a possibility of obtaining compositional information from observations in the future, but as the reviewer says, there are still many difficulties at present. In the revised manuscript, the description takes these into account.

However, I think that too many uncertainties remain on the selection of the "best", "optimal" refractive index (and indeed those words were mostly removed from the revised version) to allow that goal to be reached. Therefore, I see the manuscript more as a succession of case studies for a number of eruptions, with a nice analysis of a selection of IASI spectra and a vague conclusion about which refractive index can be selected (vague because the differences in the results for different RI are too small to be really conclusive, especially considering all uncertainties and biases that can play a role in the calculations).

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When applying the results of BTS analysis by the infrared sounder to the imager analysis, it is necessary to select one of the RI models. For that reason, the previous version proposed an "optimal" model as a RI model that explains the observations. However, as the reviewer says, the differences between RI models in RMS results are generally small, and it was inappropriate to determine a particular RI model from them. In the revised manuscript, the expression for the RI model was changed as a whole. The text of the abstract and conclusions has also been revised.

At the end the method is thus not strong enough to be considered a general method for obtaining ash composition from IASI. At least I am not convinced. But maybe this is not precisely the goal of the analysis? Some more thoughts are also in comment 5.

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Simulations using the conventional RI model of volcanic ash (Pollack andesite) did not always reproduce the measured BTS of the volcanic ash plumes. Our previous paper tried to estimate the values of RI from the observed BTS. Although the research in this paper is related to our past attempts, it has a different purpose from our previous paper. That is because a number of new RI models of volcanic ash have been provided corresponding to the ash composition. The goal of this study has changed to investigating whether a new RI model for volcanic ash composition reproduces the observed BTS. In this study, we are trying to evaluate the RI model only by the reproducibility of the BTS pattern without explicitly giving a priori information to the volcanic ash cloud (VAC) parameters. Without constraints on the VAC parameters, it was difficult to clearly determine a particular RI model, as the reviewer stated. In particular, it was found that similar BTS results can be produced regardless of the RI model if the particle size is freely selected. However, there are some parts of the observed BTS pattern that cannot be reproduced by adjusting the VAC parameters. Although it was a small difference in the RMS averaged over all channels, the ranking results for each volcanic event provided information on which RI model, mafic or felsic, was more consistent with the observed BTS. Otherwise, the plot in Fig. 8 should be a more random distribution. Such BTS analyses are likely to provide information for selecting RI models when performing ash analysis with a multi-channel imager, especially in the absence of ash composition information.

More specifically:

1. The abstract is a bit too “enthusiastic” about the conclusion that can be reached

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The abstract was corrected. It was summarized that infrared sounder measurements and simulation by accurate radiative transfer calculations for the VACs contribute to estimating the appropriate RI model in satellite VAC analysis.

2. The comparisons of MBCRM with lbl and RTTOV are not clearly detailed (more below).

3. Comparison with lbl: the authors should provide the reference to the selected line parameters (for example HITRAN version ..., or any other of course); also the information is not given about the simulations themselves: which spectral resolution was used, was an instrument line shape applied and which one, ... Those all can have a decisive impact on the comparisons. About the results, I find the RMS to be quite significant (especially for nadir only, usually differences increase with the viewing angle) although mostly lower than the IASI NEdT. Between 1200 and 1600 cm-1 the RMS of the differences is even higher than the IASI noise for a number of channels, so I don't agree with the sentence in lines 93-94, especially since that part of the spectrum is used in this work. If these RMS of differences are compared to, for example, a similar analysis with RTTOV, one can see that the RTTOV bias with respect to LBL is much lower:

https://nwp-saf.eumetsat.int/site/software/rttov/download/coefficients/comparison-with-lbl-simulations (and then in the list below you can select the IASI instrument). I do not mean that this RT code is not good, just that this bias / uncertainty (we don't know if the sign is constant, right?) is not negligible, and that combined to the IASI noise it leads to a significant uncertainty that reflects on the choice of RI. And this is not at all discussed.
4. Comparison with RTTOV: we can't see the difference good enough on the plot to assess it. I have a feeling that differences are up to 1K. This has to be taken in the discussion also; not that RTTOV is perfectly simulating aerosols, but that there's in addition to the IASI noise significant uncertainty in the aerosols radiative transfer that can amount up to 1K (if I see well, and again this is nadir only and will be larger for large viewing angles). In addition, again, the details of the RTTOV calculations are omitted. Which way are the aerosols simulated: Chou scaling (the default, very fast approximation with no explicit treatment of scattering and, to my personal knowledge, not appropriate for ash aerosols if looking for high accuracy simulations) or Discrete Ordinate method? This has a non-negligible impact on the simulations! How many streams if DOM is used? ...

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For the Line-by-Line calculation used in Fig.1, the detailed explanation was added (Line 96-99). The related sentences were also revised (Line 99-102).
For the comparison between MBCRM and RTTOV (Fig.2), there was a mistake in the setting of the sea surface emissivity in MBCRM. Without the reviewer's comments, we wouldn't have noticed this mistake. In MBCRM, the scattering properties of the ash particles for a specified effective radius are estimated by interpolation from the data table, which causes interpolation error. In the revised version of Fig.2, the interpolation error is corrected. Also, in the calculation of RTTOV, there was a mistake for the setting of ozone calculation flag. The revised RTTOV calculation in Fig. 2 used the output of 8 streams DOM (results were the same for more streams). We apologize for not being accustomed to RTTOV calculations. As a result, the consistency between MBCRM and RTTOV is improved compared to that of the previous version. Since the error of the sea surface emissivity affects all the results in this paper, the retrievals of ash cloud properties and brightness temperature simulations have been conducted again. Although the overall context was the same, there were some changes in the RMS ranking. Therefore, some selected RI models were changed in the revised manuscript.
It is understandable that error assessment is important, as the reviewer says. However, for example, it is difficult to incorporate the RI models used in this paper into RTTOV and perform same BTS simulations by the revision deadline. Also, we do not know how

much error will occur in the RTTOV calculation using the new ash RI models. A detailed understanding of multiple scattering calculations at infrared wavelengths by RTTOV is necessary. For the atmospheric profile and sea surface temperature, it is difficult to estimate the error for the values of the global analysis at that location because the true values at the observation time and location are unknown.

Instead of the required error analysis, we modified the content of the paper. In the revised manuscript, we mentioned that there are many potential error sources in the BTS simulations, such as the applied atmospheric environment and MBCRM itself, and stated that different methods may give different results (Line 446-451).

5. I really miss a discussion about all the uncertainties, biases and errors (IASI noise, RT bias/uncertainty, additional uncertainty linked to uncertainty on surface and atmospheric temperatures, ...) compared to the small differences between the simulations for different RI. Figure 9 (very useful) shows very clearly that many RI are within standard deviation of the "best", for many of the analysed eruptions, and this is without considering the uncertainties I just mentioned. This is the part that fails to convince me that the method allows to really gain useful information on the particle composition. At best with the current information in the manuscript, I would say that the method allows selecting a decent refractive index to use in simulations when trying to obtain other ash parameters such as optical depth of the plume, or particle size. A decent conclusion could also be that, after careful discussion and analysis of all uncertainties, it is actually very challenging to gain information on particle composition by selecting a refractive index based on comparisons between simulations and observations, and that all those uncertainties have to be reduced before one can actually get particle composition information from IASI.

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As the reviewer says, the RMSs between simulations and observations are large, which can be due to the various assumptions used in the radiative transfer calculations. In this study, we used IASI data that are considered to have small MC contamination, but it is considered that the effect of MC could not be completely eliminated. As mentioned in the above response, the main purpose of this study was to investigate whether the newly proposed RI models can reproduce the observed BTS. Since each RI model is linked to the volcanic ash composition, we discussed composition information for individual volcanic eruptions, but also state that the results were not always as expected. We also discussed that the choice of RI model can particularly affect the estimation of the effective radius. The results in Fig. 9 show that it is difficult to determine a particular RI model with this method. However, note that a group of RI models with relatively similar characteristics are selected in the top rankings (Fig.8). Although the differences in mean RMS are small due to the large number of averaging channels, it indicates that there was a difference in the reproducibility of the observed BTS depending on the rough difference in the RI models. If the RMS results are completely independent of the RI model, the ranking plot for the RI models should be more randomly distributed. Since the wavelengths are basically in the atmospheric window region, the wavelength dependence of the optical properties of volcanic ash particles can be considered to play a major role in explaining the observed BTS, and refractive index is the most upstream parameter that determines the optical properties of the particles. We don't think that errors in the atmospheric profile and sea surface emissivity are the decisive factors in changing the BTS pattern. To determine a specific RI model from the group of RI models with similar large values of RMS, a careful discussion and analysis of all uncertainties is necessary. We think the reviewer is correct. The basic motivation for selecting one of the RI models was to consider analysis by satellite imagers, but I regret that it was inappropriate. However, if the reviewer's opinion is that the RI model should not be discussed until all uncertainties have been analyzed, that is not our view. In the revised manuscript, we discussed some RI models selected in the analysis and removed strong expressions about the results.

Specific / technical:

- Carefully check for the use of “optimal” or “best”, and also “sum of RMS” or “total RMS” all along the paper, for consistency with changes done in this version

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The words “optimal” and “best” were completely removed. The expression for RMS has been changed to follow the reviewer’s comment.

- Lines 64-66: I think it needs rephrasing: now it looks like the particle size causes the large negative bias, but it is the ash plumes. I recommend splitting in 2 sentences.

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The sentence was split (Line 62-65)

- Lines 70-71: needs rephrasing. The IASI observations are not used to simulate BTS, but the simulated BTS are compared with the IASI observations if I understand the work correctly.

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The sentence was modified for better understanding (Line 73). But we use BTS data of IASI measurements for the RMS calculations in VAC and O3/SO2 parameter retrieval.

- Line 82: It is still written "we developed an original radiative transfer code", while the code comes from a previous publication by different authors. I strongly suggest to rephrase.

⇒

The word "original" was eliminated (Line 85). The numerical code is not provided by Buehler et al. (2010). Part of our code was developed by referring the text of Buehler et al. (2010).

- Lines 310-314: I am uncomfortable with the fact that the authors simply copied my explanation from the referee comment. It does not really belong here in this form and reads very weird in the context.

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We apologize for misunderstanding the reviewer's comment. The text was modified (Line 341-343).

- Line 385: the SO2 layer height should also be given

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The sentence was modified (Line 411). Estimated SO2 heights were provided in Table 3 and in Supplemental Material (S2). Some related sentences were also corrected.

- Lines 388-390: I am still a bit puzzled with this. I understand the authors explanation (in the answer to referee comments) why to remove this spectral range from the final RMS calculation, but I still do not understand why at step (a) this spectral part is not removed. It feels inconsistent. Also I think that at least a short explanation should be included in the manuscript, not just in referee answer

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A sentence was added (Line 420-422). "Although the range of $650\ \mathrm{cm}^{-1} \leq \nu < 750\ \mathrm{cm}^{-1}$ is important for estimating the VAC parameters, especially the top height of the ash plumes, we exclude this range to avoid RMS values related to error in the applied atmospheric profiles and to clarify the difference between the RI models in the atmospheric window region." We included the region of O3 and SO2 absorption bands in RMS calculations because we implemented O3 and SO2 retrievals.

- Figure 9: the text says it shows the mean RMS, the caption says the minimum RMS. I guess it is indeed the mean.

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The sentence was corrected (Line 604).

- Line 601-602: I don't agree with this sentence, see the major concerns above: "Furthermore, this result supports the feasibility of estimating ash RI from satellite infrared sounder measurements.

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The sentence was eliminated.

- Line 662: Even with the correction, I still do not understand what would be biased (bias is a difference with respect to a reference data set and I see no comparison to any reference here). <u>Or do you simply mean that no change of htop and tau was observed when changing the refractive index?</u>

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The first sentence for h_{top} and τ_c was deleted. We only mentioned about r_{eff} in the second sentence (Line 698-699).

- Line 733: Why here is the list of suitable RI limited to 3, while just 2 lines above there were 5? If there is a link with SO2, I do not understand it and maybe it could be explained? And then in the following lines only 2 RI are mentioned, this is not very consistent.

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Both PG070 and PG080 were written in the text (Line 772). The related sentences were also changed.

- Line 884: the report referenced is not in English, I am unsure how to deal with this

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We could not find out any English documents because it is relatively resent eruption. In the previous version of manuscript, we referred the Japanese document, thinking that we can see some plots for compositional analyses. In this manuscript, the web address at that place was removed (Line 923). (The same address is also cited in the caption of Fig.7.)

- Lines 1079-1081: this is a much too strong conclusion…

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As well as abstract, a number of corrections were made in the conclusion part to follow the reviewer.

- The conclusion section in its whole is not very clear to me, but I think this is in line with the fact that the goal of the work is not clearly stated in the introduction.

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The goal of this work was stated in the introduction (Line 56-57, 70-72).