Dear AMT Editor, dear Anonymous Referees,

please find in the following our point-by-point reply to the 3 Anonymous Referees. In **bold** are the Referee's comments and in normal font our replies. We cross-refer to the comments with the following abbreviations: MC = major comment, SC = specific comment, MiC = minor comment. We refer to page X and lines from Y1 to Y2 using the following code: PX LY1-Y2. The page and line numbering are referred to the revised manuscript. The cited text from the manuscript is in "*italics*".

In general, we would like to thank the Referees for their constructive criticism. Their comments contributed to the production of a revised manuscript that, in our opinion, is far more complete and conclusive than the AMTD version.

The main general points are the following:

- 1) A new Sect. 6 is added, with title: "Information content analysis: high spectral resolution and broad-band spectral features approaches", to reply to Referee #1 MC1, SC2 and SC3; Referee #2 MC1 and SC8; Referee #3 Mic16. The new results of Sect. 6 complete the discussion in Sect. 3.4.1 "Broad-band spectral features" (ex Sect. 3.4). They justify and explain the reasons of definition of broad-band spectral features and show possible applications, in contrast to high spectral resolution approaches. They allows the quantification of the information content of both approaches (high spectral resolution and broad-band features). Two tables have been added (Table 3 and 4). The abstract and the conclusions are modified to include these new results and discussions.
- 2) A new Sect. 3.4.2 is added, with title: "Size distribution sensitivity at fixed mass", to reply to Referee #1 SC5; Referee #3 MC3. The new results allow the disentanglement of the extinction coefficient sensitivity to the size distribution parameters and to the sulphate aerosols mass (which indeed varies with the size distribution parameters). A figure has been added (Fig. 5). The conclusions are modified to include these new results and discussions.
- 3) Several references are added and discussed with respect to our results, following the suggestions of the *Referees #1* and *#3*.
- 4) To avoid confusion about the instrumental configurations studied in the new Sect. 6, the title has been changed to "Sensitivity of thermal infrared nadir instruments to the chemical and micro-physical properties of UTLS secondary sulphate aerosols"

Referee #1

MC1) This paper presents a sensitivity study of TIR sounders to concentration and size distribution of stratospheric sulphate aerosols. It is well written; what has been done is carefully explained and discussed. However, the paper brings very little to the current literature on sulphate sounding. As a dedicated sensitivity study, it does not go nearly as deep as

one would hope for. So while there is nothing wrong with the paper, it is lacking depth and innovation - so that I would recommend either a major expansion, or resubmission to another journal.

We have already replied to this major concern of Referee #1 in a "Quick Comment" [AMTD 8, C2917–C2919, 2015]. Please refer to [AMTD 8, C2917–C2919, 2015] for a detailed description of our point of view about the innovation brought by the manuscript in the AMTD version. Nevertheless, we have carefully considered the **SC**s of **Referee #1** (as well as the other **Referees**) during the review phase and we think that the revised manuscript is a major expansion of the AMTD version (it has two new sections, one new figure, two new tables and several new references). Please find in the following a detailed point-by-point description of how we tackled these specific comments to expand and clarify the paper.

Comments (and some suggestions for expansion and improvement):

SC1) while numerous references are cited, the results from this paper should be compared and discussed with what has been found in earlier studies. The introduction of this paper majorly understates the contributions of earlier work. For instance properties such as size distribution and concentration have been retrieved before e.g. Bauman et al (2003), Doeringer et al. 2012, Echle et al (1998), Yu and Rose (2000) (Yu, T. & Rose, W. I. Retrieval of sulfate and silicate ash masses in young (1 to 4 days old) eruption clouds using multiband infrared HIRS/2 data Remote Sensing of Active Volcanism, 2000, 87-100). The sensitivity study by Steele et al (Steele, H.; Eldering, A. & Lumpe, J. Simulations of the accuracy in retrieving stratospheric aerosol from effective radius. composition. and loading infrared spectral transmission measurements Appl. Opt., 2006, 45, 2014-202) has been completely ignored.

All papers cited by Referee #1 have been considered during the discussion of the results (including the new results, see **Referee #1 SC2**, **SC3**, **SC5**). In particular, a detailed discussion and comparison with the results of Steele et al., 2006 is provided at the end of the new Sect. 6.

SC2) The analysis focuses on a few wavenumbers. To make it relevant for IASI, a more comprehensive study could for instance exploit information theory (Rodgers et al, Inversion methods for atmospheric sounding, Section 2.4 and 2.5) in which after calculation of appropriate Jacobians it becomes straightforward to discuss what and how much information can be extracted on sulfate aerosol from IASI like spectra (i.e. considering all spectral channels at once). Doing this would yield far more useful and conclusive results.

A new Section (Sect. 6: "*Information content analysis: high spectral resolution and broad-band spectral features approaches*") has been added to reply to this comment. In this Section, the information content (degrees of freedom and retrieval uncertainties for the three aerosol parameters [Ne, re, c]) has been quantitatively

estimated for different instrumental configurations:

- (1) IASI HR IASI with full spectral resolution and all channels at once
- (2) IASI BB IASI with only a few channels, to construct the three broad-band spectral features identified in Sect. 3.4.1
- (3) MODIS BB MODIS channels 32, 31 and 29, which are adapted to construct the mentioned broad-band spectral features
- (4) SEVIRI BB SEVIRI channels 10, 9 and 7, which are adapted to construct the mentioned broad-band spectral features

We are confident that these new analyses and the new Sect. 6 replies to the two concerns: why we introduce broad-band features, and how much information can be extracted by both high-resolution and broad-band features approaches.

SC3) The analysis is in my opinion not nearly quantitative enough (this links to the previous point). For instance the reader is left in the dark on lower detection limits, estimated retrieval uncertainties etc.. An example of a more quantitative study is the Steele et al. paper cited above.

As mentioned in our reply to **Referee #1 SC2**, we have quantified and discussed the independence of the information and the retrieval uncertainties for the 4 highresolution and broad-band spectral features configurations in the new Sect. 6 and in particular in the Tables 3 and 4. The main results are discussed at P28 L10-17 (background conditions): "One important result is that high spectral resolution is particularly critical at background conditions. At these conditions, broad-band features provide strongly dependent information on [Ne, re, c], with all instruments (DOFs for IASI, MODIS and SEVIRI BB are 0.24, 0.22 and 0.01, and total error higher than 100%, except for c retrievals with IASI BB). The DOF for IASI HR for aerosols at background conditions is 1.34, so more than one independent piece of information is retrievable. Nevertheless, even with IASI HR, background Ne has a total error higher than 100%, while retrieval uncertainties on re and c are around 35%." and L17-23 (volcanically-enhanced conditions): "For volcanically-enhanced conditions, the added-value of the high spectral resolution, with respect to broad-band features, is smaller than at background conditions. Broad-band features are reasonably well adapted to characterize chemical and micro-physical properties of sulphate aerosols in volcanic conditions. In the best case (IASI HR, volcanic conditions) we have found a DOF of 2.7 and retrieval uncertainties of about 30, 15 and 10% for Ne, re and mr. This indicates 3 guasi-independent pieces of information with relatively small retrieval uncertainties." These new results complete and finalize the discussion of Sect. 3.4.1 and are mentioned in the Abstract and Conclusions.

SC4) A lot of the analysis is based on the extinction coefficient. In reality, because of the radiative transfer (thermal emission and multiple scattering), it is better to work with actual simulated spectra (as only done in the second part of the paper)

As thoroughly discussed in the "Quick Comment" [AMTD 8, C2917–C2919, 2015], one of the main aims of our paper is to better identify the spectroscopic origin of the sulphate aerosol signatures in satellite spectral observations. Therefore, the very basic optical properties of sulphate layers are first analysed in terms of the

extinction coefficient rather than as pseudo-observations. In addition, this approach allows the generalisation of our results to different nadir instruments (and the results of Sect. 3 are also applicable to limb observations). Nevertheless, we think that the add of the new Sect. 6 provided a better balance between the "first" (analysis of the optical properties of sulphate aerosols, Sect. 3) and the "second" part of the paper (analysis of the BT spectra pseudo-observation, Sect. 4, 5 and 6).

SC5) In several places it is said that the radius is the most important parameter determining the extinction. However this conclusion is based on keeping the number concentration constant for varying radius. This is deceiving as the mass concentration is proportional to the cube of the radius, so increasing the radius by a factor two will increase the sulfate mass by a factor of 8! Better would be to keep the sulfate mass constant whilst adjusting the radius.

We added a new Sect. 3.4.2 "Size distribution sensitivity at fixed mass" to discuss this aspect. The aim of this section is to disentangle the sensitivity of the extinction coefficient to the size distribution parameters Ne and re, and to the consequent change of sulphate aerosols mass (increasing mass for both increasing radius and number concentration, see Eq. 7 – indeed the mass increases with the cube of the radius, as mentioned by the Referee #1). To do so, we have calculated the extinction coefficient with varying Ne and re, with fixed mass. The new Fig. 5 shows the maximum extinction (@ 1170 cm-1, i.e., ME) at fixed mass as a function of Ne and re, for typical background and volcanic masses. The results of this analysis is summarized at P19 L20-29: "The sulphate extinction is two orders of magnitude stronger for a volcanically-typical mass with respect to background. By keeping Me fixed, the sulphate extinction varies by more than 20% from smaller to bigger re, in both conditions. The dependence on Ne, on the contrary, is very limited. These further simulations allow to attribute most of the variability due to the size distribution parameters, as described in Sect. 3.3, to the increment of sulphate mass when increasing Ne or/and re. In particular, the dominant role of re in determining significant extinction signatures is mostly due to an increase of the sulphate mass in presence of bigger particles. Nevertheless, even at fixed Me, there is a significant additional variability of the extinction with varying re. This evidence confirms that re is the dominant factor determining the sensitivity of the sulphate aerosol extinction spectra."

Minor comments:

MiC1) The English is not bad, but could be improved in places

We have tried to improve the level of English text during the review process. A number of specific correction have been suggested by Referees #2 and #3 (specific *MiC*s).

MiC2) I would remove table 1 and 2, as these are trivial

We agree that Ne and re can be easily derived from N0 and rm. Nevertheless,

giving the systematic use of Ne and re in the text and figures, we think that Tables 1 and 2 should be kept to have a more readily available reference to N0 and rm. If the tables are removed, the used values of Ne and re have to be listed in the text, which seem a little bit impractical. For this reason, we prefer to keep these tables. If Referee #1 still thinks that it is useless, we will finally remove them.

MiC3) Page 8455, the discussion of the "angle of gradients" is unclear and it would be good to write down the exact formula that was used.

We have added explicitly the formula of the angle between gradients.

Referee #2

General comments

MC1) In the manuscript the authors present a work based on simulations of brightness temperature spectra for a nadir spectrometer (like IASI) to show which parameters affect the BT spectra and the associate spectral shape and magnitude, for background and volcanic conditions. The simulations are carefully done with the state of the art knowledge for H2SO4.

Results are no unexpected, but they can be useful to understand the influence of H2SO4 on nadir measurements and to identify the spectral range that can be used for retrieving H2SO4 in thermal infrared spectrometer (and eventually an ad hoc radiometer).

They show that the dependence, of the H2SO4 signal, on temperature is smaller compare to dependence on other parameters as: mixing ratio, concentration and radius of the particles. And these last 3 parameters all affect significantly the radiance in the considered spectral range. Seen the big amount of simulations they have already done, it will be nice, if possible, to end the paper checking in which condition these 3 parameters can all be retrieved and in which condition they have to be constrained with other data; e.g. compute the information content/degree of freedom in the measurements.

We thank **Referee #2** for the kind words. As suggested, we have added an entire new Section (Sect. 6) to quantify the information content of different instrumental configurations, in terms of the degrees of freedom and retrieval uncertainties of the retrieved vector [Ne, re, c]. Please refer to **Referee #1 SC2** and **SC3** for more details. We think that this addition completes the paper in the direction indicated by **Referee #2**.

Specific Comments

SC1) page 8444 - line 5 'were compared to IR high spectral resolution observations': which instrument?

We have added more information about the validation of 4A/OP at P6 L12-14: "...were compared to IR high spectral resolution observations (aircraft observations with the High Resolution Interferometer Sounder and the Air-borne Research Interferometer Evaluation System) and found capable..."

SC2) p 8845 I I4-5-5 'phase function), which can be represented by the integral asymmetry parameter (van de Hulst, 1957).' Seen that this paper is on 'simulation only' this is fine, but it could be mention that for a proper retrieval in condition where they said the scattering is not negligible (as for volcanic conditions), a better approximation for the phase function will be to consider the Legendre moments.

We agree and, correspondingly, we added the following line at P7 L15-17: "...(van de Hulst, 1957). It is worth mentioning that using Legendre moments would be a better approximation of the phase function, when scattering processes are more important."

SC3) p 8446 l 2-3 'we fixed ?r to 1.86 (a typical value, see, e.g. SPARC, 2006)'. Any idea on how much the simulated spectra change if this parameter is different?

Yes, changing σm from 1.86 to 1.50 or 2.20 (about 20% decrease/increase), the extinction coefficient varies of just a few percent (background, both σms) to about 20% (volcanic conditions, $\sigma m=2.20$). The stronger increase for volcanic conditions and wider distributions is probably due to the higher single scattering albedo and then the scattering contribution for bigger particles (more present for volcanic conditions).

SC4) p 8449 I 'This result suggests that the scattering component of the extinction, even if relatively small with respect to the absorption, cannot be neglected'. This is true for volcanic condition (SSA 0.2) but for SSA of 0.01 (as fig 1 show for background conditions) maybe scattering can be neglected.

Yes, of course. We corrected the sentence (P11 L20-22): "This result suggests that the scattering component of the extinction, even if relatively small with respect to the absorption, cannot be neglected in volcanically-perturbed conditions."

SC5) p 8450 line 16-17 'This suggests a strong sensitivity of the sulphate aerosols extinction to the size distribution'. This suggest that bigger particle (rm=0.3 micron) affect the TIR radiance more than the smaller particle (rm=0.06 micron), as expected from Mie theory.

We agree.

SC6) p 8452 I 4 'in general, the extinction does not vary much with temperature' please give some numbers as 'don't vary more than XXX'

We have modified the sentence to (P14 L9-12): "In general, the extinction does not

vary much with temperature, except for a few cases, e.g., a 20% variation of the absolute extinction, from 183 to 293K, with a mixing ratio of 57% in both background and volcanic conditions. For other mixing ratios, the variation with temperature is less than 10%"

SC7) p 854 I 16 missing the '(1)' I think.

Yes, thank you.

SC8) p 8456 I 13-17. 'All these considerations suggest that the three aerosol parameters are retrievable as independent quantities only for limited conditions, when using broad-band sulphates extinction spectral features and constraints should be given to at least one parameter (e.g., the number concentration).' In which limited condition we can retrieve the 3 parameters independently? Under volcanic condition with high concentration and bigger radius? It will be nice to check if the information content in the measurements can be used to retrieve the 3 parameters in both a spectrometer 'IASI like' and a radiometer as 'MODIS like'. For example showing the averaging kernel or computing the degree of freedom for IASI and MODIS. Otherwise we don't know if you need to constrain one parameter, maybe you need to constrain 2 or maybe with few channels you can have 3 degree of freedom. The sentence is reasonable but it can be computed and presented.

This is discussed in the new Sect. 6. Our new results suggest that the 3 parameters can be retrieved independently and with reasonably small errors only with high spectral resolution spectrometers (IASI-like) and volcanically-enhanced conditions. Broad-band radiometers (MODIS and SEVIRI-like) can still provide partially independent information (2.11 and 1.48 DOF) in volcanically-enhanced conditions but cannot at background conditions (0.22 and 0.01 DOF, with total uncertainty > 100% for each aerosol parameter). Please refer to **Referee #2 MC1** and **Referee #1 SC2** and **SC3** for a more detailed reply.

SC9) I 17-21 A nice addition will be to overplot the spectral range covered by MODIS and SEVIRI channels to an H2SO4 spectral signature. (for example on top of figure 8?).

We have modified Fig. 8 (now Fig. 9 due to the addition of a new figure) to include this information.

SC10) p 8457 I 26 'Impossible' is a big word, Maybe it is possible to use some information from this bands if the retrieval is done together with the O3 retrieval.

We have changed "impossible" to "very hard" (it looks indeed very hard to use ionic bands and we do not focus further on this aspect in our paper)

SC11) p 8459 I 13 'specral' -> spectral

Done

SC12) p 8464 I 11-13 Also CO2, SO2 and H20 have different spectral BT signature/shape from H2SO4 and can have similar magnitude. Parameters that affect BT spectra of comparable magnitude should be retrieved simultaneously with H2SO4 parameters or constrained from independent data.

Yes, that's how we plan to do at the operational level (retrieve simultaneously or constrain with external data). To account for this, we corrected the sentence (P30 L29 - P31 L1) to: "At the operational level, these interfering parameters should be retrieved simultaneously with sulphate aerosols or constrained with independent data."

SC13) I 25-27. see my comment above on 3 parameters.

Please refer to *Referee #2 SC8* and other related comments.

SC14) Fig 2 Maybe a legend with color line used in the plot and concentration will be better than a colorbar? where are the red and dark blue lines in the second rows plots?

We have used a continuous colorbar because the available mixing ratios are not exactly identical for different temperatures (depending on the laboratory measurements of Biermann et al. (2000)). Extreme H2SO4 mixing ratios are not present for laboratory measurements of the refractive index at 213 K and then are not shown in the plots of the second row. In general not all concentration/temperature combinations are available, as mentioned in the text.

SC15) Fig 3 Legend with line color and numbers instead of colorbar.

Done

SC16) fig 6. Seen that the 'y' steps are not linear, you should plot the y values for every plotted row. Otherwise, for example in the first plot, the reader don't know which is the value of the concentration in the lower row and if the concentration in the top row is 10 or 9

Done

Referee #3

The article 'Sensitivity of thermal infrared sounders to the chemical and micro-physical properties of UTLS secondary sulphate aerosols' by P. Sellito and B. Legras uses simulations to investigate the potential of IR nadir sounders to retrieve microphysical properties of sulphate aerosol. Generally

the manuscript is very detailed and well structured. However, I have some major and minor concerns that should be addressed before publication.

Thank you for the kind words

Major comments:

MC1) The introduction motivates why it is important to observe UTLS sulphate aerosol. Please explain here, how well nadir observations can give an altitude information. Do you assume that sulphate aerosol is mainly located in the UTLS region? Can nadir measurements only observe UTLS aerosol but not aerosol in the lower and middle troposphere?

In this work, we focus on UTLS sulphate aerosols. Sulphate aerosols can be found also in the lower and middle troposphere. We fixed the UTLS vertical region with two main assumptions: 1) the altitude of the aerosol layer in the generation of the IASI pseudo-observation with 4A/OP forward radiative transfer model, 2) the modelled composition of sulphate aerosols (droplets of H2SO4/H2O solution, with varying H2SO4 mixing ratio), i.e., their refractive indices. For this latter point, please note that sulphate aerosols at lower altitudes than UTLS are potentially more complex in terms of composition, e.g., ammonium sulphates, sulphate/nitrates ternary systems or other. For this reason, we don't think that merely changing the altitude of the layer (e.g., testing the sensitivity at lower altitudes, with the same H2SO4/H2O assumption on composition) would be useful – it is not realistic. Nevertheless, simulating more complex compositions is outside the scopes of the present paper. Of course we plan to study different compositions of sulphate-containing aerosols in the future, to test the sensitivity of lower/middle tropospheric sulphate aerosols, e.g., from anthropogenic pollution.

MC2) If you focus on UTLS sulphate aerosol, I would recommend to constrain the temperature and mixing ratio range to UTLS conditions. At first reading it was not clear to me if this should be a general sensitivity study or a study focussing on the UTLS.

The temperature/mixing ratio combinations are constrained to UTLS typical conditions in Sect. 3.1 and from Sect. 3.3 on, i.e., when discussing the variability of the sulphate extinction and of the synthetic observations (i.e., using these optical properties in forward radiative transfer modelling). A wider variability of temperature/mixing ratio combinations is only investigated in Sect. 3.1 (so, it appears only in Fig. 2). This larger range was necessary to better discuss the temperature and mixing ratio dependence of the extinction properties of sulphate aerosols. We prefer to show the complete variability of the extinction properties, with respect to these two variables, to have a more general context for the subsequent discussion. We think that this is necessary, especially when dealing with the temperature dependence. The temperature can significantly vary also if the UTLS region is fixed. We have added the following lines (P12 L5-7) to explain this aspect: "The whole set of temperature/mixing ratio combinations, i.e., not constrained to UTLS conditions, is analysed in the present section. This has been

done to have a more complete view on this dependency."

MC3) Section 3.3 and Figure 3 are very critical to this study. When keeping the radius constant and increasing the concentration (or keeping the number concentration constant and increasing the radius) the volume and hence the extinction increases. To investigate the effect of different radii, the volume should be kept constant. Also I suggest to normalize the spectra in Figure 3 to make the differences/similarities in the spectral slope more visible.

To clarify this aspect, we have added a new section (Sect. 3.4.2 "*Size distribution sensitivity at fixed mass*"). Please refer to **Referee #1 SC5** for more details. As for the proposed modification of Fig. 3, we think that both depth (absolute value at a fixed wavenumber) and slope are important to interpret the differences in the spectral extinction coefficients. In addition, the slope is investigated in Sect. 3.4.1 (RE1 and RE2 are linked to slopes calculated between different spectral ranges). For these reasons, we prefer to leave Fig. 3 in the present shape.

MC4) When discussing the retrievability of sulfate aerosol properties I highly recommend to also discuss previous work on this topic with IR measurements, e.g. Baran et al., 1993, GRL Clarisse et al., 2010, Applied Optics Karagulian et al., 2010, JGR Lambert et al., 1997, JGR

Yes, we agree with Referee #3 and we added these works in the inherent discussion (Clarisse et al., 2010 was already cited and discussed in the AMTD version).

Minor

MiC1) p.8442 I. 27 What do you mean with 'different sulphates'? also ammonium sulphate or different concentrations?

We have clarified this aspect at P5 L6-8: "...the different sulphur-containing species (e.g., undissociated sulphuric acid molecules, sulphate and bisulphate ions) contained..."

MiC2) p.8443 I.6 What is the source of the size distributions used?

These size distributions are typical for background and volcanic conditions and can be found in [SPARC, 2006], as indicated in the manuscript.

MiC3) The mixing ratio of sulphate aerosol depends on temperature and water vapour concentration. Can you please discuss how much variability of sulphate can be expected in the UTLS?

The mixing ratio of sulphate aerosols depends in a complex (and still largely unknown) manner on temperature and humidity, due to the nucleation process. There exist parametrizations to link H2SO4 mixing ratio to these conditions but we think that discussing this is not in the scope of our work. In any case, in our

manuscript (except Sect. 3.2, where the mixing ratio is allowed to vary from 0 to 80%) we have fixed the available mixing ratios at 213-215 K to 60-75%, which is indicated as typical in the UTLS by [SPARC, 2006].

MiC4) p.8446 I.22-24 in the IR and for particles smaller than about 15 microns you should be careful with the effective radius, because scattering is not necessarily linear with the effective radius (when varying the median radius and the distribution width), but with the scattering radius.

Yes, that is true but: 1) the scattering component is very small in our case, and 2) we are not searching explicitly for a linear relationship between extinction and a size parameter.

MiC5) Section 3 titles: 'dependence on' instead of 'dependence from'

Done

MiC6) p.8449 I.5-7 Griessbach et al. consider scattering for sulphate aerosol

Yes, that is true and we have substituted Griessbach et al. with other more pertinent references in this context.

MiC7) p.8450 I.9 please quantify the size of 'bigger particles'

We have corrected the sentence to "...with bigger particles (volcanic conditions, see Fig. 1 and inherent text)."

MiC8) p.8451 I.11-15 Isn't the mixing ratio dependent on temperature?

We think that Referee #3 refers to the dependence during the nucleation phase of the gas-to-particle conversion leading to secondary sulphate aerosols. Yes, in this case, the mixing ratio depends on temperature (see also **Referee # 3 MiC3**). Nevertheless, in this paragraph we rather discuss the absorption spectra as a function of the temperature, in terms of dissociation equilibrium and then relative absorption of ions and undissociated H2SO4.

MiC9) p.8452 I.14-15 If the altitude cannot be retrieved, how do you know that the sulphate aerosol is in the UTLS region? (e.g. longer life time in UTLS that in lower troposphere?)

We affirm that the altitude cannot be retrieved using the temperature dependence of the extinction (which is very small). At the operational level, other criteria should be considered to locate the sulphate aerosols in the UTLS, e.g., prior knowledge (retrievals in regions affected by volcanic eruptions or pollutants transported by deep convection)

MiC10) p.8453 I.7 What do you mean with 'severe volcanic conditions'? After Pinatubo extinctions?

Yes, and we indicate that in the text

MiC11) p.8454 I.13 Please give some examples for broad band radiometers you have in mind.

These are MODIS and SEVIRI, and are indicated at the end of the section. Please note also that these instrumental configurations are more specifically studied in the new Sect. 6.

MiC12) p.8460 I.4 How many levels did you use? What was the vertical step?

The TIGR profiles are sampled at compatible pressure levels as for 4A/OP standard atmospheric profiles (variable thickness, from about 0.5 km in the lower troposphere to 1.5-2 km in the upper troposphere and a few km in the stratosphere).

MiC13) p.8461 I.3 Does 'smaller spectral resolution' mean broader or finer resolution?

It means "finer". We have corrected that sentence to clarify it.

MiC14) p.8464 I.11-13 Isn't the ash effect on the radiances stronger that the effect of sulfate aerosol? Please give a reference for this statement.

The reference, (Corradini et al., 2009), is already mentioned at the end Sect. 5.2, where this aspect is discussed: "The spectra are expressed in radiance units as a function of the wavelength to more readily compare the impact of the sulphate absorption to the impact of ash, as reported in Fig. 1 of (Corradini et al., 2009). The reduction of the TOA radiance due to ash is typically 10 to 25 % at its maximum absorption spectral range (about 10–11 μ m, 1000–910 cm-1), depending on the ash optical depth. Sulphates can impact for a few percent, for small and diluted droplets (green lines in Fig. 9), to up to about 25 % for volcanically-enhanced conditions (red lines in Fig. 9) at its maximum extinction spectral range (about 8.5–9.0 μ m, 1175–1110 cm-1)." We don't think that re-citing this reference in the Conclusions would be useful.

MiC16) Can you show how much better results would be for high resolution instruments in conrast to broad band instruments?

Please refer at the new Sect. 6 and the detailed replies to **Referee #1** (**SC2** and **SC3**) and **Referee #2** (**MC1** and **SC8**).

MiC17) Figure 8: the colors are very pale (close to invisible)

We have corrected that, i.e., the colours of O3 and CO2 absorption bands are now more visible.

MiC18) Finally, I would recommenend to carefully read and correct the paper after reworking. There are several typos and mistakes, e.g. p.8442 I.2 '...,due to the their extended...' ... p.8446 I.14 '.. of a few tents particles...' ... p.8463 I.2 '...where the the sulphate...' p.8463 I.8 '...bynary...' p.8464 I.26 '...contraints...' (these are just a few examples)

We have corrected a number of typos; thank you for indicating some of them.

Sincerely, Pasquale Sellitto and Bernard Legras