Final Response to Referee #1 and Referee #2:

We would like to thank both referees for their detailed review of our paper. Please see below our point-by-point answers to their comments.

Additionally, you can see a revised version of our paper attached as a supplement. The abstract and introduction were rewritten to clarify the main objectives and the implications of this work. The uncertainties were reevaluated and clarified in the text when needed. Also, we renamed the distribution of particles deposited on the coarse filters to "mixed" to be more consistent with the characteristics of that distribution. All changes necessary to address their questions and comments are highlighted in bold in the new version of the paper for easier identification.

Sincerely,
The authors.

Response to Referee #1:

Interactive comment on "Optical, microphysical and compositional properties of the Eyjafjallajökull volcanic ash" by A. Rocha-Lima et al.

In this paper the authors discuss the analysis of physical, optical and chemical properties of volcanic ash samples collected from the ground at 35 km from the Eyjafjallajökull volcano. The analysis and the results are definitely interesting and in my opinion deserve publication. However, there are a few aspects of the analysis and discussion that are not fully developed, explained or clear to me and in my opinion they require some significant revision/addition. Following are some general and some more specific comments.

General comments:
It is not clear if the sample so collected is representative of what was long-range transported over Europe. As this is one of the main motivations of the paper (flight safety), it seems advisable that the authors discuss a bit more on how the sampling location was chosen and why, and especially how representative should be the ash collected at such location with respect to ash transported over Europe. The discussion about why the assumption that light extinction is solely (or mostly) due to absorption is interesting, but I am not 100% convinced by the explanation and I think the paper would benefit from some additional discussion and information supporting this assumption or attempting to estimate the bias that the assumption might introduce. It could be useful to the community to report the values of the index of refraction and uncertainties (as a minimum) also in tabular format so that future investigators could use the data directly; if the authors have good confidence in these results. This might increase the impact
and usefulness of the paper. Also it could be useful to discuss how general these values might be with respect to other volcanic ashes. The paper motivates the study with the need to improve the retrieval and quantification of volcanic ash emitted by eruptions; however, there is not much discussion in the end of the paper or in the abstract on how the findings of this study really contribute to improve the retrievals. This also links directly to my

previous comments on the general applicability of the results found here. A discussion of the implications of this work might increase the impact of the paper. The added figure 13 is interesting. The use of the measured aspect ratio to evaluate the sensitivity of the obtained refractive index is a good addition. However, as mentioned later in the specific comments, the use of 2D imaging can substantially underestimate (at least for some type of particles) the aspect ratio due to orientation of the particles on the substrate and the unknown third dimension, so the assertion that the aspect ratio of 3 is an extreme and conservative value, might actually not be accurate (see the recent paper by Veghte and Freedman, Aerosol Science and Technology, 48:715–724, 2014 as an example of several studies on this issue). This does not mean that the analysis reported here is not interesting, on the contrary; however, the caveats of 2D top-view only analysis should be explicitly discussed. Manufacturer and model of each instrument should be explicitly reported, and the methods used for the analysis should in some cases be more explicitly explained. The meaning of the uncertainties should also be explicitly discussed (do the error bars represent 1 standard error, or 1 standard deviations, or something else?).

We appreciate all the comments the referee raised in this "General Comments" section. We agree that all of them are important questions to be addressed. Since all the questions raised here are also mentioned in the "Specific comments" below with more details, we decided to reply to each of the questions directly in the specific comments.

The abstract is a bit succinct, it briefly reports what was done and how, but there is no summary of what the significance of the results are. Some more elaboration (e.g. a sentence or two on the significance of the work and the implications) might be beneficial.

As suggested, we rewrote the abstract and the introduction to cover the significance and implications of this work.

Page 13272, Line 17: Please report explicitly the year of the last eruption.

The year was added to the text.

Page 13272, Line 26: As currently put, the motivation in terms of global effects due to large volcanoes seems a bit irrelevant here, as the authors mentioned just a few lines earlier that the Eyjafjallajokull was a small to moderate eruption (line 19).

The introduction was rewritten and this part was removed.

Page 13274, Line 2: What do the authors refer to by “correction models”?

The introduction was rewritten and this part was removed.

Page 13274, Lines 21-24: As mentioned in the “general comments” section, this part of the paper needs to have a little bit more detail with some justification of the chosen site and some consideration on how well (or not) the sample collected at a location so close to the volcano (35 km) might be representative for particles transported downwind over long distances (for example larger or denser particles might fall off, mixing, oxidation and agglomeration might happen/change etc.). Also the samples were collected 4 weeks after the eruption, this is of course reasonable considering the proximity to the volcano, but would this result in significant aging (for example morphological changes), weathering of the ash? I am not suggesting these samples are not useful, on the contrary, but I also think that the authors should address these issues.

There are some reasons that make us consider that this sample is representative for particles transported downwind. First, larger particles were removed from our sample during the sieving process, so our analyzed samples contained only the smaller particles that are the ones that have higher probability to be carried downwind over long distances. In relation to the weathering or morphological changes, particles fallen down close to the volcano or particles that were carried over long distances, might have been subjected to weathering effects. Particles collected on the ground might be less affected by weather effects because layers of ash deposited on the top can in some circumstances protect the layers underneath but specific studies on this issue would be needed to accurately answer this question. Finally, the direct intercomparison of composition and optical properties between samples collected on sites close to the eruption and on sites located at long distances is necessary and might be the most accurate way to evaluate this problem.

On this direction, N. Bukowiecki et al ("Ground-based and airborne in-situ measurements of the Eyjafjallajokull volcanic aerosol plume in Switzerland in spring 2010", Atmos. Chem. Phys., 11, 10011–10030, 2011) reported that the average chemical composition of volcanic ash particles that reached Switzerland is very similar to some material collected nearby the volcano. Also, Beeston, M. et al. ("Chemical and morphological characterization of aerosol particles at Mt. Krvavec, Slovenia, during the Eyjafjallajökull Icelandic volcanic eruption." Environmental Science and Pollution Research 19.1 (2012) 235-243) found evidence that the ash from the Eyjafjallajökull's eruption reached Slovenia (more than 2500km away from the volcano) based on the analyses of chemical composition of the aerosol, which has shown similar properties as those from the Eyjafjallajökull eruption. We added this information in the Sect. 3 of the manuscript.

Page 13274, Lines 26: could the “sieving” affect the particle morphology (e.g.; through fragmentation or abrasion)? It might be good to briefly comment.

Our sieving process cannot affect the particle morphology. It is performed using a stainless steel sieve with mesh of 45 um. This sieving is done by gently shaking the sieve. Sieved particles pass through the mesh naturally without any pressure applied on them. Ash from Eyjafjallajökull volcano was found to have a high hardness (S. R. Gislason et al, Characterization of Eyjafjallajökull volcanic ash particles and a protocol for rapid risk assessment, PNAS 2011 108 (18) 7307-7312 doi:10.1073/pnas.1015053108), which makes any fragmentation and abrasion of the volcanic ash unlikely in this process.

Page 13275, Line 9: The authors refer to the particles being “disaggregated”, what does that imply with respect to the original shape and size of the particle? Does this imply possible fragmentation of the original particles? If so, how do these fragmented particles represent airborne particles?

We refer here to cases in which there are agglomerates of particles. Samples of particles deposited on the ground show commonly small particles statically attached to large particles. For this case, the disaggregation/separation of these agglomerates is important because it will produce a more realistic particle size distribution. In addition, the single light-particle interaction theories (Mie and T-matrix) that were applied in our analyses assume this condition. We added a line in Sect. 2.1 clarifying this.

Page 13275, Line 13: "efficiency close to 100%" shouldn't that depend on the particle size? So to what size do the authors refer to? >0.4um?

The collection efficiency of particles definitely depends on the particle size. However, for a 0.4um pore filter, the collection efficiency is very high (above 90%) for all particles sizes (Cahill et al, 1979). This information was clarified in the text and this additional reference was included.

Page 13275, Line 14: Judging from figure 4, it seems like the 5um pore substrates might have a very broad size cut as several submicron particles still seem to be retained by the filter, do the authors still consider all of these as coarse? Does the retention of the top filter affect the measured vs actual distribution of the fine particles?

The collection efficiency for a 5um pore filter is around 50% for particles smaller then 1-1.5um, which means that there is a fraction of fine particles that will inevitably be collect on the first filter. The collection efficiency of the small particles decreases abruptly for particles lower than 1.5um for this filter. We agree that the name coarse is not quite accurate for this distribution, so we decided to rename it as mixed mode.

Page 13275, Line 16: Provide the model of the microbalance used and uncertainties.

Mettler Toledo UMX2 with resolution of 0.1ug. This information was added to the text (Section 2.1).

Page 13275, Line 21: SEM, please provide model and some basic specifications

JEOL 5600 with tungsten filament electron source and detectors for secondary and backscattered electron imaging. Its maximum resolution is 5 nm. This information was added to the text (Sect. 2.2)

Page 13275, Line 17: What procedure? Discuss how the procedure works, with at least some minimal detail.

The description of the weighing procedure was added to the text (Sect. 2.1).

Page 13275, Line 24: The area equivalent diameter is a useful parameter, but did the authors explore the issue of deposition orientation? In other terms, what is the third dimension of the ash? How tall are they? 2D analysis is very useful, but it also has limitations and it would be good to admit the limitation due to the unknown third dimension, to warn the reader about this caveat.

We are aware of the orientation issue. To evaluate any possible deposition orientation of the ash sample on the filters, we examined a set of particles with a top view SEM image and also with a SEM image taken from the same particles by tilting the sample in the microscope by 45 degrees. We estimated the ash particle's volume as V=A.r, where A is the top view cross section area of the particle and r is the radius extracted from this image. We added a paragraph in the text to address this question (Section 2.2).

In relation to the question raised about the aspect ratio in the "General Section" above, we found from the aspect ratio distribution (Fig 13a) that the probability of finding particles with aspect ratio higher than 3.0 is 0.6% for our sample. By looking at the particles with the tilted sample in the microscope, we could confirm that the aspect ratio of the particles in our sample does not change significantly with the deposition orientation, in contrast with some cases analyzed by Veghte and Freedman, Aerosol Science and Technology, 48:715–724, 2014.

Page 13276, Line 12: Is the pycnometer a custom made instrument? Describe briefly. Also what are the uncertainties associated with these volume measurements?

The pycnometer is a custom made instrument. We have included a brief description about its characteristics in the text (Section 2.3). Our system consists of a vessel of adjustable volume with resolution of 0.1ml and a barometer with resolution of 0.5psi. The final uncertainty of the density presented in the text 0.13g/cm3 was obtained by simple propagation of the errors of the volume, mass and pressure.

Page 13276, Lines 18-19: Discuss potential reasons for these discrepancies.

Values of the particles' grain density found in the literature are usually based on assumptions of composition of the main material constituent. These values do not come from a specific measurement of the material, which is a possible reason for these discrepancies. Since our methodology to retrieve the imaginary part of the complex refractive index is sensitive to the ash's density, an accurate evaluation of this parameter as well as its uncertainty was essential. A brief discussion about these discrepancies was added to the text (Sect. 2.3).

Page 13276, Line 25: provide some specification, manufacturer and model of the broad band light source.

High power UV-VIS light source - Hamamatsu - Model: L10290 and Reflectance Lamp – ASD Inc. This information was added to the text (Sect. 2.4).

Page 13277, Lines 7-14: It would be good to have some reference supporting these conclusions.

We have added a reference (Reid, J. S., P. V. Hobbs, C. Liousse, J. V. Martins, R. E. Weiss, and T. F. Eck: Comparisons of techniques for measuring shortwave absorption and black carbon content of aerosols from biomass burning in Brazil, J. Geophys. Res., 103, 32,031 – 32, 040 - 1998) to support our assumptions on the measurement of absorption from optical reflectance measurements. This work reports a comparative study of several techniques to measure the absorption of aerosol on filters. They show a validation of the technique we used by comparing it with a standard extinction cell and a nephelometer. According to their analysis, the reflectance results were in agreement with the extinction cell measurements for a large range of aerosol loading. This information was added to the text (Sect. 2.4).

Page 13278, Line 10: What is the “range considered”?

We refer to UV-VIS wavelength range. We have rewritten the sentence to make it clear.

Page 13278, Lines 22-23: There is also a dependence on shape.

Information added.

Page 13279, Lines 1-2: “with the assumption of spheroids”. . . how? Using a fitted ellipsoid? Explain. Also what was used as third axis of the ellipsoids? Are these prolate or oblate?

Ellipsoids created by the T-matrix code were completely defined by the "equal-volume sphere radius" distribution of the particles and their axial ratio, according to Mishchenko, M.I. et al. 1998. In the case of the T-matrix, the spheroids are symmetrical under rotation about their axis. We consider the particles as oblate spheroids. This information was added in Sect. 2.5.

Page 13279, Line 8-9: “. . .within a trusted interval”, specify what interval was used and how it was “trusted”

We performed the minimization to obtain Im(m) and Re(m) simultaneously. Since the sensitivity to obtain Re(m) is small, we used the average value obtained for Re(m) = 1.68 and we added a range of +/- 0.1 to study the influence of Re(m) on the minimization procedure. This range produces a maximum error in the value of Im(m) corresponding to +/-0.00025, which drives the error bar in Fig. 9. We added this explanation in Sect. 2.5.

Figure 2: In the caption, or better in the text, provide the manufacturer and the model of all the components, in particular the CPC and its size range.

These specifications were added to the text in Sect. 2.1.

Figure 3: Consider using higher resolution image format as the length scales are hardly readable.

We updated these images. They have better resolution now.

Figure 5: Here or in the text, it might be wise to put the explicit formula for Vash as a function of all the known parameters. Also was this a custom made instrument? What are the performances in terms of precision and accuracy?

This is a custom made instrument and more details about its characteristics and uncertainties were added to the text in Sect. 2.3.

Figure 6: The authors mention in the text that reflectances were measured at different angles. What angle is the reflectances in the figure measured at? Also what is the uncertainties on these curves? How do these uncertainties propagate; in other words, how do the index of refraction retrievals change if one would consider noise and other source of uncertainties?

The reflectance curves shown in this figure were performed at zenith angle of 10 degrees while the light source illuminated the filters at zenith angle of 45 degrees, in the same azimuth. Each curve is an average of 25 measurements of reflectance, reducing noise levels to less than 0.5%. For these measurements the filter should be as flat as possible; waves in the surface of the filter will increase the variability of the reflectance and increase uncertainties. The total uncertainty in these measurements is mainly driven by the smoothness of the filter and homogeneity of the particles collected on the filter. These were estimated to be of maximum 2%. This information was added to the text in Sect 2.4.

Figure 7: What do the error bars represent? Please discuss also how the uncertainties were estimated. Also is there any uncertainty associated with the choice of b? In the caption add to "power law fitting. . ." also what is reported on both axes, so that the figure can be interpreted as standalone. Is there any uncertainty on the loaded mass?

The error bars were obtained combining the uncertainties of the reflectance measurements and the loaded mass. Both axes were clarified and uncertainties explained in the caption of the figure. The uncertainty in the b parameter was not taken into account in this study.

Figure 8: What do the error bars represent? Please discuss also how the uncertainties were estimated.

The uncertainties in Fig. 8 were estimated from the error obtained for the coefficient "a" on the power law fitting. We added this information in Sect. 2.4 of the text.

Figure 9: What do the error bars represent? Please discuss also how the uncertainties were estimated.

Error bars represent one standard deviation interval of probability and were estimated based on a sensitive study considering the main sources of errors: the real part of the refractive index, mass absorption efficiency, particles' volume and density.

Figure 10: What do the error bars represent? Please discuss also how the uncertainties were estimated. Why there are uncertainties associated with the Mie but not with the T-matrix calculations?

The main sources of error in the T-Matrix method are the same as the Mie method and they are driven by the uncertainties in the real part of the refractive index, mass absorption efficiency, particles' cross sectional area, volume and density. For this reason, we estimated that the uncertainties on the imaginary part of the refractive index for the T-Matrix calculation have the same magnitude as the uncertainties estimated for Mie Theory. Uncertainties for T-Matrix were not explicitly shown with the curve to make it easier to read this figure. We added this information to the text in Sect. 2.5.

Figure 11: What do the error bars represent? Please discuss also how the uncertainties were estimated.

Error bars represent one standard deviation of the concentration obtained from the measurements of twelve samples of fine filters and four samples of mixed filters. This information was added to text.

Figure 12: What do the error bars represent? Please discuss also how the uncertainties were estimated.

Error bars were estimated from direct error propagation of the measurements of concentration. This information was added to the text and to the figure's caption.

Technical corrections

Page 13273, Line 3: Consider removing "in the form"

Corrected.

Page 13273, Line 23: Consider to add "to be" in front of "known"

Corrected.

Page 13275, Line 17: Probably a "t" is missing in "weighed"

We double check this word and we believe "weighed" is the correct form in that sentence.

Page 13276, Line 9: Consider adding "with the distribution from " after "overlap"

Corrected.

Page 13277, Line 24: Consider removing “fraction” after “reflectance”

Corrected.

Page 13278 (and 12381), Line 11 (and line 26): Change Moosmuller into Moosmüller

Corrected.

Page 13279, Line 14: Consider changing “bellow” to “below”.

Corrected.

Page 13280, Lines 13: “modified gamma function” modified how? Also do the authors mean “modified gamma distribution”. . .

The modified gamma distribution follows the definition of the T-matrix code. We added a reference that contains the explicit form of this distribution.

Page 13281, Line 20: Consider changing “submitted” to “subjected”

Corrected.

Figure 4: There seems to be a gap at bin 9 um. Is that real or is something missing?

This is an effect of the sampling process. Obtaining a bin with no counts is statistically possible in this region of particle’s size due to the low frequency.

In the caption, consider changing “bellow” into “below”. What does the sentence “as presented by Seinfeld and Pandis” really mean (or refers to) here?

Corrected. The sentence referred to the form used to plot the distributions, normalized by the width of the bins. We changed the text to clarify this meaning.

Figure 13: The x-axis in plot A is labeled “axial ratio”, for consistency with the text the authors should consider labeling the axis as “aspect ratio”.

Corrected.

Response to Referee #2

Response to Interactive comment on "Optical, microphysical and compositional properties of the Eyjafjallajökull volcanic ash" by A. Rocha-Lima et al.

The manuscript "Optical, microphysical and compositional properties of the Eyjafjallajokull volcanic ash" by Rocha-Lima et al., describes the physical, chemical and optical characterization of a sample of ash taken from the vicinity of the Eyjafjallajokull volcano after it erupted in 2010. I think the data the authors are presenting is of interest; however, the manuscript is lacking a thorough discussion of their results, and a thorough error analysis. I do not recommend publication in ACP in the present form. Following are my recommendations for the manuscript to be suitable for publication.
One of my main concerns is the retrieval of the imaginary part of the RI. First, from the calculation of the mass absorption efficiency (_abs) using Eq.1, the authors assume that the attenuation is only due to absorption. It is not clear to me that this assumption is correct and "Light scattered forward by the particles will most likely hit the white surface of the filter underneath and scatter backward on its path back to the spectrometer" is not a strong justification and the authors are completely ignoring multiple scattering in the filter. The authors should include either a supplementary section to support their assumption, or add it to the main text.

To address this question we have added a reference (Reid, J. S., P. V. Hobbs, C. Liousse, J. V. Martins, R. E. Weiss, and T. F. Eck: Comparisons of techniques for measuring shortwave absorption and black carbon content of aerosols from biomass burning in Brazil, J. Geophys. Res., 103, 32,031 – 32, 040 - 1998) to support our assumptions on the measurement of absorption from optical reflectance measurements. This work reports a comparative study of several techniques to measure the light absorption of aerosol on filters. They show a validation of the technique we used by comparing it with a standard extinction cell and a nephelometer. According to their analysis, the absorption coefficient derived from optical reflectance was in agreement with the extinction cell measurements for a large range of aerosol loading.

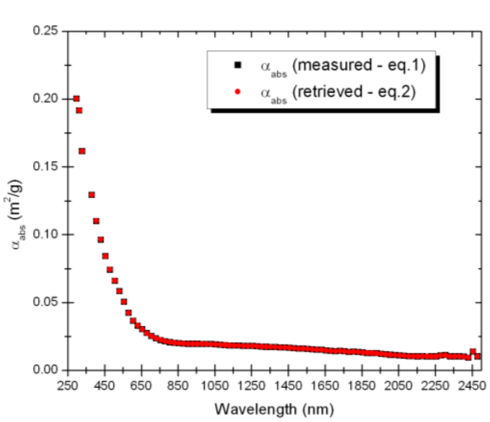
In contrast to fiber filters such as Teflon, quartz or paper, Nuclepore filters have the important characteristic of collecting particles on the surface of the filter in opposition to having them trapped inside the fibers of the filter. Artificial enhanced absorption due to multiple scattering is indeed observed when particles are inside the fibers of the filters. Multiple scattering effects are minimized in the Nuclepore filters due to particles being collected on the surface of the filters (Martins et al., 2009), for this reason these effects were neglected in this work. We clarified this discussion in Sect. 2.4 of the text.

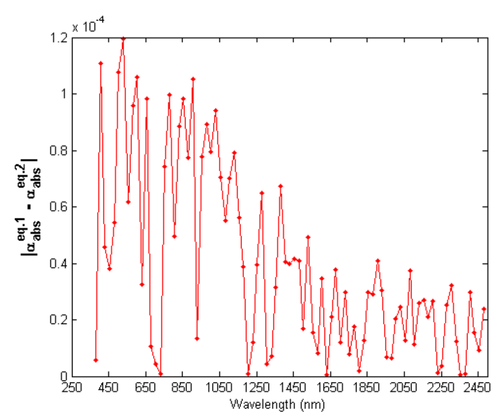
Second, about the size distribution analysis, it is not clear how the authors take into account the clear over-sizing and under-counting by their ImageJ software and how errors in the determination of the number, area, and volume distributions will translate into the retrieval of the imaginary part of the RI. An error analysis is also needed here.

In addition to the *ImageJ* software, Photoimpact 3 was used to take into account cases that are not easily resolved. For cases where there are superposition of particles and particles that do not contrast well with the background, the Photoimpact 3 tool called "lace" was used to manually define the contour of the particles. We added in Fig. 3 an additional image showing the intermediate step done by the Photoimpact 3. Errors related to the cross section area A(r) and volume v(r) of the particles are estimated to be around 10% . This error was added quadratically to the errors from the main sources of uncertainties: real part of the refractive index, mass absorption efficiency, and cross-section, volume and density of the particles. The final uncertainty is shown as error bars in the imaginary refractive index Fig 9.

Finally, the authors calculate _abs using Eq. 2 by changing the m to get a value for Qabs to calculate the mass absorption efficiency and minimize the difference with the mass absorption efficiency calculated from Eq. 1 (taken from the reflectance measurements). When they reach a minimum in their minimization procedure, they take the inputted m as the derived value. The authors should plot _abs from Eq. 2 (from the spherical assumption and from the T-matrix calculations) in Fig. 8. A minimum difference can always be achieved but showing that the _abs using Eq. 2 is equal (or within error of the _abs calculated with Eq. 1) will show the robustness and reliability of their retrieval. Also, what is their convergence criterion?

The first figure below shows alpha_abs from the measurements of reflectance (as described by Eq. 1) overlapped with alpha_abs derived from the minimization process (as described by Eq. 2). The second figure shows the difference between alpha_abs (Eq. 1) and alpha_abs (Eq. 2). The difference between alpha_abs (Eq. 1) and alpha_abs (Eq. 2) is lower than 0.2% in the full wavelength range. These figures illustrate the robustness of our minimization procedure. These results are for fine mode only, considering Mie theory. The differences of alpha_abs for the cases of mixed mode and for T-matrix calculation were found to have the same order of magnitude as the curves shown below. We added a paragraph in Sect. 2.5 summarizing this information.





The authors mention on Section 2.6 that energy dispersive X-ray fluorescence analysis (EDXRF) of the ïnˇA¸ne and coarse particles was used to investigate dependence of the refractive indices with chemical composition, but they do not elaborate, there is no mention of what might be (or is) the dependence. They just conclude the manuscript stating that further studies will be needed to explain the differences in optical properties observed between fine and coarse particles. This section should be elaborated.

The compositional analyses were performed to characterize the studied material. The differences of composition found between fine and mixed particles can only support the existence of optical differences but it is out of the scope of this study to try to obtain direct correlations between refractive index and composition. We rewrote the sentence to make clear these objectives.

From their introduction, it is also not clear what is the main purpose of the paper. The authors mention aviation in their introduction, but they don't state or elaborate on how will these results help aviation?

The imaginary refractive index is a key parameter needed in any aerosol atmospheric model. The main purpose of the paper is to help to fill the gap between observations and atmospheric models by providing optical and microphysical properties of the Eyjafjallajökull volcanic ash. We rewrote the abstract and the introduction to clarify the purpose of this paper.

They also mention modeling and remote sensing. How will their results improve modeling and/or remote sensing? What are the implications of their results in comparison with previous publications? Where do the differences come from? etc.

One important input parameter in studies with remote sensing and modeling is the complex refractive index of the different types of aerosol. There is a significant lack of information available for this property in the literature. This often leads people to use the spectral imaginary part of the refractive index as a constant, not depending on the wavelength. Some works with remote sensing and modeling adopt the complex refractive index of ash equals to dust due to the lack of data available. In this work we report our measurements of the mass absorption efficiency and the retrieved imaginary part of the complex refractive index in a broad spectral range for the Eyjafjallajökull volcanic ash, and also the methodology we used to extract this quantity. We rewrote the introduction to clarify this importance in the text.

Other comments: The abstract should be re-written. It states the different methods used to do the measurements, but it does not give a concise summary of the results that the authors are presenting.

As suggested, we rewrote the abstract to better summarize our results and emphasize the importance of our work.

Why is a value of the aspect ratio of f=3.0 is 'certainly overestimating' the effects of shape? From the filter is clear there are some extreme shapes.

We agree with your affirmation that there are extreme shapes and we can see that in Fig. 13. However, from the aspect ratio distribution, the probability of finding particles with aspect ratio higher than 3.0 is 0.6%. In the T-matrix calculation performed in this study, we adopted a representative value of aspect ratio for our collection of particles equal to the median aspect ratio. We would certainly be overestimating the representative aspect ratio of our collection of particles if we assume an extreme case, e.g. f=3.0. We changed our text in Sect. 3 to clarify this information.

Can the authors expand on why they think their results are different that the previous published values of the RI?

Our results of Im(m) are in agreement with values found in the literature for some specific wavelengths reported by these other studies. To the best of our knowledge, there is no data available for the Eyjafjallajökull volcanic ash in the full range of wavelengths we measured. This discussion can be found in Sect. 2.5 of the text.

What is the error in the density measurements? Is the value in parenthesis (13) the error? If yes, add a ±. Why is the density value lower than previous reported values?

The uncertainty in the density is 0.13g/cm3. We show this value in the suggested form. Values of the particles' grain density found in the literature are usually based on assumptions of composition of the main material constituent. These values do not come from a specific measurement of the material, which is a possible explanation for the discrepancies. Since our methodology to retrieve the imaginary part of the complex refractive index is sensitive to the ash's density, an accurate evaluation of this parameter as well as its uncertainty was essential. A brief discussion about this difference was added to Sect. 2.3 of the text.

In the discussion section the authors began by stating that the α_abs values are 'different' for the fine and coarse modes. However, it is clear from Fig. 8 that the difference only arises below around 600 nm, and 0.1 m2/g is not a big difference. Then the authors state that the imaginary part between the modes is only 'slightly different', but the imaginary part of the RI is proportional to α_abs. The authors should be consistent in their statements.

We changed the sentences in the first and second paragraphs in Sect. 3 to make them more consistent.

It is incorrect to say imaginary refractive index; this should be changed throughout the text to imaginary part of the complex refractive index

Corrected.

Page 13273, lines 3-4: what does 'respectively' refer to? The phrase is not clear.

The word “respectively” should not be there. We removed it.

Page 13273, lines 8: ‘environment’ should be ‘environmental ‘

We rewrote the introduction and this part of the text was removed.

Page 13275, line 3: add ‘the’ after (B)

Added.

Page 13278, line 6: For the equation _abs = G/(2a), the ‘a’ should be ‘_’, as it was defined in equation 1.

We rewrote the sentence to explain that “a” is the fitted parameter from the power law fit.

Page 13279, line 13: High imaginary indices. . . High with respect to what?

Corrected. We rewrote this sentence to make it clear.