Review of the paper "Variability of the infrared complex refractive index of African mineral dust: experimental estimation and implication for radiative transfer and satellite remote sensing" by Di Biagio et al.

Submitted for publication on ACPD

Comments in black, answers in red

At first, we would like to thank the reviewer for having carefully read the paper and provided valuable comments which helped to improve the quality of the manuscript. We have taken into consideration all the questions raised by the reviewer, and changed the paper accordingly. The details of our changes are highlighted in the text. The point by point answers are provided in the following.

Referee 1

Di Biaggio et al. present a very interesting study on the infrared refractive index of desert dust. The manuscript is well structured and the work is precisely described. As the presented work might be of very high value for many researchers I strongly recommend publication of the manuscript after some minor revisions.

I have only one major comment which the authors might wish to think about. I would strongly encourage the authors to provide the refractive index data of their study, either as table or in the form of harmonic oscillator parameters. Potentially the data can be provided online as supplementary material along with the online version of the manuscript, if the authors wish to not add an additional table. As they correctly describe, many applications rely on at best inappropriate descriptions of the optical constants of mineral dust. So providing the new set of dust refractive indices would potentially be extremely benefitial for a whole range of applications.

We thank the reviewer for its suggestion. Refractive index data are already available as supplementary material to this paper.

Minor comments:

l. 94: Chlorite is not really a clay mineral.

Clay minerals are divided into four main groups: Kaolinite, Illite, Smectite, and Chlorite groups. Chlorite is sometimes considered as a separate group, however formally it should be included among clay species.

l. 95: I know that iron oxides locally strongly contribute to mineral dust. What would be typical fractions of titanium oxides? Are they important on large scales?

Recent works performed near source areas suggest the contribution of titanium oxides to be of the order of 1-2% in mass, so a little lower than iron oxides which are estimated at about 4-5% of the dust mass (e.g., Formenti et al., 2014). At the large/global scale they are thus

expected to contribute for a little fraction of the dust mass. However, there are several works highlighting the importance of these components in several chemical reactions in which dust particles take part (e.g., Gustaffsonn et al., 2006).

l. 223: Ryder et al. (2013) found particles up to 100_m during the Fennec campaign. Given the OPC measurements, how likely is it that the large / giant particle fraction is underestimated in the size distributions?
As discussed in Ryder et al. (2013), the occurrence of very large/giant particles depends on the distance from the source and the aerosol load. In their study, they sampled very large particles in correspondence of heavy dust loads (aerosol optical depth > 2) in proximity of fresh dust sources. In our study, dust events were characterized by a lower optical depth (<1.30) and corresponded to low/medium range transported plumes (with the exception of the SOP1-8 case for which the highest content of coarse particles is observed). This may explain why we did not observe significant fractions of giant particles in our data.

l. 291: I understood that the particles have diameters smaller than 20 µm, which would be about the Mie-scattering particle size. How realistic then is the assumption, that scattering effects can be neglected? It would be worth to comment a bit more on this assumption.
As discussed in the paper, since dust constitute only the 0.1% of the mass of the pellet, its scattering signal is negligible compared to that of KBr, which instead constitutes the 99.9% of the pellet mass. This is corroborated by observations, since the measured dust spectra (Figure 1) do not show a detectable scattering signal but only absorption. The reviewer is right and this is undoubtedly an important point, since the fact of not measuring dust scattering with the pellet approach strongly limits the applicability of the results. A discussion on that point has been added in the Introduction (lines 145-151).

Section 5.3: OPAC (and other older databases) have a quite coarse spectral resolution in the thermal infrared window. It would potentially be worth to discuss this point, especially in the light that a couple of spectral features are not represented in the OPAC optical properties.
We thank the reviewer for this very useful comment. We rewrote the following sentence in Section 4.3 (587-590) as: "*The spectral signatures from the different clay species appear smoothed in the curves taken from literature, mainly because of the rather coarse spectral resolution of these datasets. Only a major single peak between 9 and 10 µm is observed, compared to our data where multiple clay peaks are detected in the 8-12 µm spectral interval*.".

l. 660: Which algorithm for calculating the Mie-scattering functions has been used? Especially for the small (x<1) particles described here, how stable is this algorithm when approaching the Rayleigh limit? For the Rayleigh limit itself, has there any other approximation been used, which is not original Mie theory? If so, which one?
Concerning the Rayleigh limit, we did not perform specific approximations, but we used the original Mie theory. Calculations have been performed by using the mie_single.pro IDL routine (http://www.atm.ox.ac.uk/code/mie/mie_single.html), which does not show any instability issues when approaching the Rayleigh limit.

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Referee 2

This paper is describing an experimental method to estimate the dust infrared refractive index from five samples collected during the AMMA campaign at Banizoumbou (Niger) and Tamanrasset (Algeria). Along with transmission measurements, size distribution and complete mineralogical measurements were performed. The results were then compared to previous studies and highlight a relatively good agreement.

The introduction is well written but the following sections need to be restructured. Indeed the manuscript is long and probably too long. As the introduction stated it, you should use your collected data to retrieve the refractive indexes (RI) and compare it to the databases to highlight discrepancies. However, the last parts (Implications for satellite remote sensing, Implications for dust radiative forcing) are superficial and do not add anything to the paper. Indeed, this manuscript is long (more than 30 pages) and probably needs to be shortened.

The paper needs major revisions before submission to ACP.

Following the reviewer suggestion, we tried to restructure the text in order to make it more linear and concise. In particular, we reorganized the discussion in the Introduction and in Section 3.1 (the new Section 2.1). To reduce the paper length we decided to move Section 2 "Selection of dust events and identification of their source region" at the end of the paper, in Appendix A. The information contained in this Section are not strictly necessary to the comprehension of the paper (while, however, they provide a full description of the case studies which may be of interest for several aspects), and its displacement also significantly contribute to simplify the discussion. We also rewrote some sentences along the paper and eliminated some redundant bibliography with the aim of shortening the manuscript. However, despite these modifications, due to the changes in the Introduction and in Section 2.1, the full length of the paper has not significantly changed compared to its starting version.

Concerning your comment about Section 6.2 and 6.3 ("Implications for satellite remote sensing", "Implications for dust radiative forcing", now Sections 5.2 and 5.3) we do not agree with the reviewer in stating that they do not add anything to the paper. The main aim of these

Sections is to give some elements of discussion about the possible implications associated to the variability of the dust refractive index and intensive optical properties. To this, some simple, but in our opinion also effective, examples are given to highlight the different consequences in terms of both satellite remote sensing and radiative forcing calculations. For instance, for what concerns the satellite remote sensing part, we discuss some possible uncertainties both in dust detection and SST retrieval associated to the variability of dust extinction and its spectral behaviour. Then, in terms of radiative forcing we try to give a quantitative estimate of the forcing sensitivity to the extinction of the particles based on both our observations and published results. We find that these two Sections are of relevance and increase the significance of the paper, also because they contribute to underline the necessity for a better investigation of dust properties to reduce several uncertainties in climate studies.
Finally, as also evidenced by the reviewer in one of its following comments, it is out of the scope of the paper to go more in deep in the implications here by providing specific analyses, because such a more detailed study would require a paper by itself.
To clarify the scope of the whole Section, the title has been changed in: "*Infrared intensive optical properties of mineral dust: variability and implications*".

General comments:

First the authors need to proof read the paper. There are several careless mistakes that need to be corrected within the paper as well as some grammatically incorrect sentences. I highlighted some but there are more that I probably missed.

We carefully checked the paper and tried to remove all mistakes. We thank the reviewer for having highlighted some of them.

From my knowledge, this technique is used since the 70s. What are the benefits of your technique compare to earlier studies? Please add descriptions of how the earlier studies retrieved the RI, otherwise the readers can't understand (i) The benefits of your study and (ii) where the differences highlighted in figure 7 are coming from. Moreover, what are the main limitations of this technique? To create your pellet you have to modify the dust properties such as the size (random selection of dust from the sample) and probably the chemical composition during the storage in an oven (100_C). How could you be sure that the size distribution within the pellet is the same than the one on the filter and thus the same than measured by the OPC? If there is any difference how would it change your results? Could you do a sensitivity study to estimate the errors? I would like to see a discussion on those issues in the paper.

We thank the reviewer for this comment, since it has helped us to realize that one crucial point of the paper was not clear. As now discussed more clearly in the Introduction and in Section 3.1 (the new Section 2.1), the technique used in our study is almost the same as used in past literature works. This choice was made to help the comparison between past and new data, so to highlight the variability of the complex refractive index as a function of particle physico-chemical properties. In both cases (past literature and our study) the pellet spectroscopy approach is used, and also the data analysis to retrieve the refractive index is mostly based on the same assumptions, i.e. dust scattering is negligible and the shape effect

not taken into account. Then, we additionally assume that the particles verify the Rayleigh limit, which is coherent with our dust size field measurements. We do not consider necessary to give in the text more details on the experimental procedure and retrieval approach used in past studies, since the necessary information to understand and interpret results are now more extensively provided. Also, further explanations necessary to understand some differences between our results and literature data are provided at the end of Section 5.3 (new Section 4.3).

Concerning your comment about the limitation of the pellet approach and the possible modifications induced on the dust sample during laboratory manipulation/pellet production, we have added a discussion on this topic in the Introduction and in Section 3.1 (new Section 2.1). Also in this case we thank the reviewer for its helpful comment, since this aspect is very important to discuss in order to completely understand both the state of the art of dust infrared studies and the impact of our results.

Some more specific answers:

1. In terms of size distribution, we have highlighted in the text the possible modifications, especially in the pellet pressing phase; we exclude significant effects on the size due to an incomplete transfer from the filter to the pellet, as the extraction procedure is performed in a very careful way by repeating the ultrasonic shaking several times, and by ensuring that all the dust on the filter is collected;
2. Regarding the fact the size distribution within the pellet is or not the same that the one measured by the OPC, we have added some comments in Section 2.1. This aspect is already specified this point in Section 4.1: "*The fact of observing differences in the absorption spectra which are coherent with the variability of the size distribution measured for the different samples indicates that the main features of dust size have little changed during sample manipulations and pellet production*."
3. Concerning the possibility of estimating the error induced by the sample modifications on the refractive index estimates, it is clear that this is very difficult to do since we are not able to quantify how much the size, composition, and shape have changed. Thus, we need to leave this as an "undefined" uncertainty. This point has been however clearly specified in the main text (lines 244-250).

It has been proven (Schuster et al., 2013 and several others) that the refractive index of dust particle in the visible depends on the mineralogical composition of dust. To calculate dust refractive indexes you are using the same assumption (n=1.53) for each sample on the refractive index of those dusts in the visible (i) to resolve the equation 5 and (ii) to correct the dust size distributions. You need then to explain the impact of this assumption on your results? Is there any modification of the dust RI in the visible as a function of the type/mineralogical composition of dust particles?

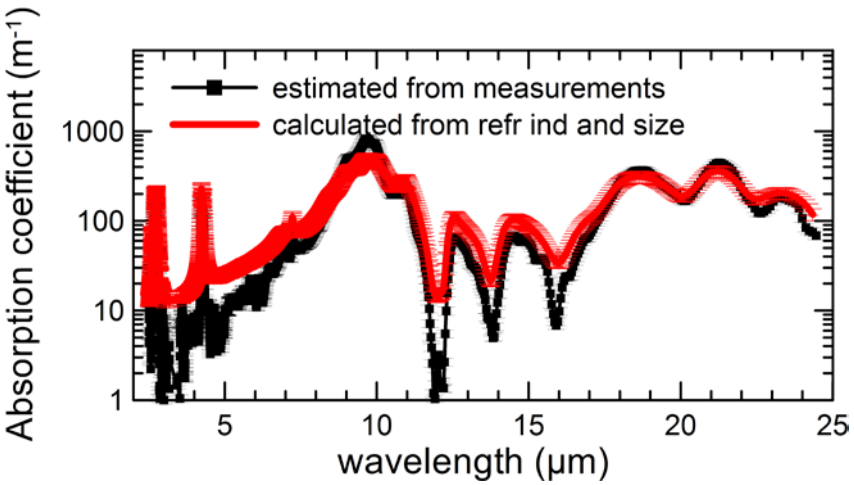
Yes, as correctly stated by the reviewer, the refractive index of dust in the visible depends on the mineralogical composition of particles and for instance it may vary as a function of the dust source region. For instance, several field studies report values ranging in the 1.42-1.56 interval (Osborne et al., 2008; Petzold et al., 2009; McConnell et al., 2010; Klaver et al., 2011, among others). In this study, we do not have an independent estimation of the dust refractive index in the visible (for instance from measurements of the dust optical properties

at visible wavelengths and inversion calculations based on the size data), so we need to fix it based on literature values or ancillary calculations. As stated in the text, we decided to fix the value of the refractive index at 1.53, which corresponds approximately at the mean of the values reported in the literature for African dust. As shown in Figure 7, n_{vis}=1.53 also agrees with the values assumed or retrieved in the limit of visible wavelengths by most of past literature studies on the dust refractive index. Thus, also to maintain retrieval conditions similar to that of past studies, we decided to assume the value of 1.53 for n_{vis}.
In order to better justify our choice, we have added this sentence in Section 3.1 (new Section 2.1, lines 314-315): "*Our choice of 1.53 is also in agreement with the values reported by most of past literature studies on the dust infrared refractive index (see data shown in Fig. 7)*". Moreover we have added the references of McConnell et al. (2010) and Klaver et al. (2011) in the text.
Despite all the previous considerations, which support our choice of n_{vis}=1.53 in the refractive index retrieval algorithm, we undoubtedly recognize the importance of this assumption. In fact, the value chosen for n_{vis} determines the "offset level" for the retrieved n(λ) and, by that, also the absolute value of k(λ) throughout the whole infrared range. Therefore, an independent estimate of n_{vis} should be preferred in these calculations to reduce the uncertainty on the obtained results. We consider such a detailed discussion not necessary, however we have added a sentence in Section 5.2 (new Section 4.2, lines 529-531) better specifying that some uncertainties come also from the choice of n_{vis}: "*Another source of uncertainty for n and k, which we do not quantify in this study, is also associated to the choice of the* $\left(n_{0,m}\right)_{vis}$ *value in Eqs.(5)-(7), here set at 1.53 in agreement with past literature studies (Volz, 1973; Carlson and Benjamin, 1980; Longtin et al., 1988).*".

Concerning the effect of the choice of n_{vis}=1.53 on the GRIMM particle size correction, I have conducted some sensitivity studies to evaluate the impact on this assumption. I have calculated the GRIMM optical to geometrical diameter conversion by varying n and k in the range n=1.53-1.56 and k=0.001-0.003, which are the most used n and k data intervals assumed for African dusts (e.g., McConnell et al., 2008; Formenti et al., 2011). The results of the sensitivity study suggest that the choice of the refractive index to perform the correction has a limited impact on the shape of the retrieved GRIMM particle size distribution for our measurements. For instance, the uncertainty associated to the variability of n between 1.53 and 1.56 is less than 10% in the whole GRIMM diameter range. So we may conclude that our choice of 1.53-0.002 for the dust refractive index to estimate the GRIMM diameter correction to do not significantly affect our analysis.
It should also be pointed out, moreover, that in this paper the size distribution is not used in a quantitative way, and so possible artefacts associated to this choice have a little impact on the dust infrared refractive index estimations.

You state that in average 94 and 98% of the particles in number concentration satisfy the a/lambda < 0.1. Could you estimate the errors due to large particles made on your calculations?

The reviewer is right, and this is undoubtedly an important point. Actually, it is not so evident to provide with a quantitative estimation of the error induced by the assumption a/lambda < 0.1. A possible approach is to use Mie theory to try to reproduce the dust absorption spectrum (which can is also directly measured with spectroscopy) based on the retrieved refractive index and the measured size distribution (not used in the retrieval algorithm). The difference between the "measured" and "calculated" extinctions may give an indication of the accuracy of our refractive index retrieval, and possibly an indication of the size effect you mentioned in other comments. We have tried this approach, for instance, for sample SOP1-17. In this case (see Fig. below) we are able to well reproduce by calculations the different features of the absorption spectrum in the entire wavelength range. The red curve in the figure is the calculated spectrum, where the uncertainty of this spectrum is obtained by performing Mie calculations considering the refractive index +/- its uncertainty. Some differences in terms of intensity are observed between the measured and calculated absorption at different spectral bands, especially between 12 and 16 µm and below 8 µm, so where dust absorption is very weak (absorption spectrum in our Fig 2 close to zero). These differences may in part be associated to the effect of the size to which the reviewer referred. It has to be pointed out, however, that it is complicated to assess the fraction of this discrepancy which is associated to the size effect, as also other factors may play a role in determining the goodness of the comparison. Among these, for instance, the dust homogeneity within the pellet or the choice of the optical theory, as also highlighted by the reviewer. In conclusion, based on the different observations of this study we can assume that the retrieval algorithm for the refractive index provides with consistent results, also if some degrees of uncertainty still remain due to the different assumptions which necessarily have to be made in the retrieval approach.



Finally, Dubovik et al. (2006) described a new code for spheroids that would be much more suitable than Mie code to retrieve optical properties for dust. Have you tried it?

No, we have not tried it. We do not know if the reviewer refers to the refractive index retrieval described in Section 2.1 or to the calculations in Section 5.1. For what concerns Section 2.1, has discussed in the paper, we have preferred to use a retrieval approach the most possible similar to that of previous studies in order to facilitate a quantitative comparison

with past data. So, since no shape effects were considered in past studies, we decided to make the same choice (see lines 301-303). For what concerns Section 5.1, in order to maintain the same approach, and also because we were mostly interested at the absolute variability of the optical properties, we continued performing calculations with Mie theory. However, the effect of the shape on the retrieval algorithm is an important aspect which will deserve future investigations. In addition to the Dubovik code for spheroids, there are other approaches, such as the Rayleigh theory for specific particles shapes (such as disk, needles, etc.), which have been shown to work quite well in reproducing dust infrared spectra (Hudson et al., 2008; Laskina et al., 2012, among others).

Specific comments :

P10598, L18-20 Need to rephrase 'We also found that the real and the imaginary parts of the refractive index from part of literature studies do not verify Kramers–Kronig relations, thus resulting theoretically incorrect.'
We have rephrased in: "*Furthermore, the real and the imaginary parts of the refractive index from part of literature studies are found not to verify the Kramers-Kronig relations, thus resulting theoretically incorrect.*"

P10598, L20-24 : You should split this sentence.
The sentence has been split in two: "*The comparison between our results, from Western Africa, and literature data, from different locations in Europe, Africa, and the Caribbean, nonetheless, confirms the expected large variability of the dust infrared refractive index. This highlights the necessity for an extended systematic investigation of dust properties at infrared wavelengths.*"

P10600, L 26 : You have to be more specific cause some optical properties may be measured in the IR like the absorption, extinction and scattering coefficients. . .
The reviewer is right since there are several remote sensing instruments, i.e. AERI or other spectroradiometers such as those on board of satellites like IASI or AIRS, which allow measuring the aerosol extinction/absorption/scattering signal in the infrared. However, the retrieval of the aerosol optical properties, i.e. extinction coefficient or optical depth, from these measurements is quite difficult, due for instance to the superimposing effect of other atmospheric constituents, such as CO2 or water vapour. To better specify this point the text has been rewritten as (lines 89-96): "*Dust infrared optical properties, in fact, cannot be directly measured by in situ instruments, and also their estimation based on ground-based or satellite remote sensing observations is difficult, due to the fact that the aerosol signature is partly covered by that of main atmospheric constituents (water vapour, CO_2) and that a priori knowledge of the investigated optical properties is often required by inversion algorithms. So, dust infrared optical properties are generally estimated through an alternative approach, which consists in calculating them based on the knowledge of the physico-chemical properties of particles , i.e. composition, size distribution, and shape (Levin and Lindberg, 1979; Highwood et al., 2003).*"

P10601, L26 : the composition of dust may BE additionally modifIED due to the. . .
The correction has been performed.

P10602, L4-21 : This paragraph is really confusing. First, you describe your set up and your methodology and then you said that methods that have been used are far from perfect. Maybe here you should start with the limitations from previous studies and then highlight the improvements in your method and therefore state that the 'natural variability of the dust infrared refractive index remains not represented'.
This part of the manuscript has been rewritten (see also the answers to general comments).

P10602, L 22-28 : You need to integrate that paragraph in the previous one.
This part of the manuscript has been rewritten (see also the answers the general comments); as it is in the present form we consider that this paragraph should remain separated from the previous one.

P10603, L 9-10 : Need to rephrase that sentence : 'of dust at most after 1–2 days of their atmospheric transport.'
We have rephrased as "*As here we want to focus on the refractive index variability near source regions, the five cases have been chosen to be representative of local emission episodes or of dust at most after 1-2 days of atmospheric transport*".

P10604, L 1 : Wintertime 'corresponding to the' dry season
We have rephrased in "*one of the dust samples was taken during winter in correspondence of the dry season*".

P10604, L5-9 : Long sentence. . . You need to rephrase it in order to facilitate the reader's understanding. 'The SOP0–47 case sampled during the dry season occurred on February 9th 2006. This event was associated to a medium-range transport event originated from the Central-Western part of Niger, between the Algeria-Niger and the Mali-Niger frontier regions.' Have you looked to the satellite measurements to make sure the desert areas close to the Air massif were included in the source area?
We have rephrased following the reviewer suggestions. As described in Klaver et al. (2012), the origin of each dust event was determined by combining Hysplit back-trajectories and satellite Infrared Difference Dust Index (IDDI) images (Legrand et al., 2001). For the SOP0-47 event, backtrajectories+satellite data indicate a possible contribution from the desert areas close to the Aïr Massif. However, since this level of detail is not necessary in this context, we decided to eliminate the sentence "*possibly including desert areas close to the Aïr massif*" from the main text.

P10604 : UTC is usually the most common unit for time. You should consider replacing UT by UTC.
We have replaced UT with UTC throughout the main text and Table 1.

P10604, L18-22 : ‘SOP1–17 was instead a post-erosion event characterized by the advection of dust which was locally emitted at Banizoumbou _ 3 h before the sampling started.’ So it’s a local event ? How long the dust erosion event over Banizoumbou last for?
Yes, it was a local event. The erosion occurred 3 hours before the SOP1-17 sampling started and lasted for about 15 minutes

P 10605, L 2-6 : Need to rephrase this sentence.
The sentence has been rephrased as: *“The event considered here in correspondence of the Monsoon season over the Sahel occurred between the 21st and the 27th of July 2006. The dust event originated at the Algeria-Niger frontier and was caused by the succession of three cold pools outflows generated by Mesoscale Convective Systems (MCS). The ITD (Inter-Tropical Discontinuity) organized the three cold pools in a dust front, which propagated northwards to Tamanrasset (see Cuesta et al., 2009a). The N32 sample was taken only in the first part of the episode, between the 23rd and the 24th of July.”.*

P 10605, L18-26 : It seems like this very general paragraph has been added after the others and do not belong here. Also part of it has been already said in the introduction. And :‘among the largest sources for dust’.
The scope of the paragraph was to underline the importance of the dust sources considered in this study also with respect to medium and large scale transport, so to highlight the impact of the obtained results. Now that Section 2 is in Appendix to the paper this general consideration results unnecessary. So, also following the reviewer suggestion, this paragraph has been eliminated.

P10606, L3-5 : Which criteria did you use to collect dust ? The total concentration had to be larger than a threshold or the concentration of particles larger than a fixed diameter was larger than another threshold? Could you be more specific?
Sampling has been performed by using different instruments in parallel (different filter lines, each dedicated to a specific sample analysis, and a TEOM microbalance). The dust sampling interval during the dust events was set considering the mass requirements of the different specific analysis techniques. For instance: 1. A maximum of 500 µg could be sampled on filters dedicated to the analysis of the dust iron content, which used the CBD method and XRF measurements; 2. A minimum of 800 µg had to be sampled to perform the analysis of the particle mineralogical composition which uses the XRD technique. The aerosol mass concentration was continuously monitored during the dust events by means of the TEOM instrument. Then, based on these co-located data, the sampling time was adjusted to respect the mass limits of the different analysis techniques. Depending on the type of event, the dust sampling time could be shorter or longer. For instance, erosion events are typically very rapid and characterized by an elevated dust concentration, so the dust sampling could be of ~1 to few hours; transport events, on the contrary, last generally for a longer time and are characterized by a lower dust concentration, so the filter sampling may be quite long in these cases.

In this part of the manuscript, the text has been changed in (lines 194-196): "*The sampling time for the different cases (see Table 1) varied between a few hours to 1-2 days, depending on the aerosol concentration and the duration of the event.*".

P 10606, L 15 : the wavelength range 2.5–25 μm (400–4000 cm−1). You need to add sigma in the parenthesis.
The sentence has been rewritten as: "in the wavelength range 2.5–25 μm (4000-400 cm^{-1} wavenumber)", to specify that in the parenthesis we report the wavenumbers. Despite the symbol generally used to indicate wavenumbers is σ, we preferred not to use it in the paper to do not confuse with the σ_{exp} uncertainty, reported in the caption of Figure 5.

P1060, L23 : 'a delicate operation' . Yes it is, so could you estimate the errors due to an incomplete transfer from the filter to the pellet ?
See the answers to the general comments.

Section 3.1 : You shortly described the pellet technique earlier (P10602) Where it was probably not adapted.
In the revised version of the paper we have extended the description of the pellet technique in the Introduction and discussed limits and uncertainties of this technique; this part is important to understand the motivation and the objective of the paper. The technical details of the pellet technique are instead provided in Section 3.1 (the new Section 2.1).

P10607, L3 : Left not leaved.
The correction has been performed.

P10607, L 17 : 'Passing the samples in the oven does not modify the dust mineralogical composition, as at these temperatures the only effect is water evaporation'. Could it remove the volatile components that may be present on dust's surfaces? Could it rearrange the mineralogical structure of dust ? This is an important hypothesis and I believe that it deserves at least one reference.
As stated in the manuscript, passing the dust samples in the oven does not modify their mineralogical composition, as at these temperatures the main effect is the evaporation of water. In addition, as stated by the reviewer, also volatile compounds, which might have contaminated the dust sample during laboratory manipulation, are expected to evaporate at 100 °C.
Apart from the water vapour or the possible volatile compounds evaporation, there are no reasons why the mineralogical structure of the dust should change at these temperatures, which are relatively low to induce some effects on the crystallographic structure of the particles. Concerning this point, also refer to the different phases of the calcination process (see http://fr.wikipedia.org/wiki/Calcination; I apologise but the most complete website reference I have found is in French). The organic component of dust, which represents a small fraction of the particles, evaporates at very high temperatures (>500 °C). Also, as described in several old studies (e.g., Bradley and Grim, 1951), considerably high temperatures (from 500 to 1300 °C) are required to induce some modifications of the clay

structure. We also want to point out, finally, that the procedure we have used here follows the standard sample preparation procedure for infrared spectroscopy measurements (see J. D. Russel, chapter 4, 133-173, "Infrared methods" in "*A handbook of determinative methods in clay mineralogy*", Edited by M.J. Wilson, 1987), normally used for mineral studies. See also, as a reference for sample preparation for infrared spectroscopy, the paper by Bertaux et al. (1998). Given all these considerations, we have decided to rewrite the sentence as it was in its original form but just replacing "*as at these temperatures the only effect is water evaporation*", with "*as at these temperatures the main effect is water evaporation*", to include also the possible evaporation of volatile compounds. However, we do not have a specific reference to add here.

P10607, L 22 : Does the 10 tons (AND this is not a pressure ! this is a weight.) change the size, the morphology or the mineralogical structure of the dust particles ? If yes what are the consequences for your study if not this statement deserves also a reference.
The reviewer is right since some modifications are expected to occur during pellet production; however, it is very difficult to estimate them correctly as well as to evaluate the effects of them on the measured spectra. Regarding this point, we have added in the text this small discussion (lines 244-250): "*We should expect, as already mentioned, that some of the dust properties, such as the size distribution, aggregation state, or also the morphology of the particles, may partly modify during the pellet production, thus affecting the representativeness of the analysed sample compared to airborne conditions. It should be pointed out, however, that the estimation of the effects of these modifications on the measured spectra is very difficult, and a certain degree of uncertainty on this aspect remains not quantified.*".
Then, 10 Tons has been replaced with 10 Tons cm^{-2}, which is a pressure.

P10607 L23-24: Then all the pellets are put in the oven at 100 _C until they are used for transmission spectroscopy measurements. How long could that period last for ? So what are the consequences for dust particles (see comment just above) ?
As already stated in the main text, the effect of passing the pellets in the oven is to avoid as much as possible water vapour to be absorbed by the highly hygroscopic KBr, so altering the spectroscopic measurements. No other relevant effects are expected. Pellets remain in the oven for a period of about 1-2 hours; this point has been specified in the text (line 242).

P10608, L25: You need a reference for the dust density. Is it the same density you used to plot the volume size distributions of dust particles (Fig 1b)? Besides, you used the same density for each type of dust? Could you at least note it within the manuscript, estimates the errors (compare to the dust smallest and largest density ever measured?) and refer to previous studies.
The value of 2.5 g cm^{-3} was selected as representative of the mean particle density (ρ) for the different analysed cases. This value was chosen approximately at the mean of the range of desert dust aerosol densities as reported in several literature studies, i.e. 2.1-2.75 g cm^{-3} (e.g., Maring et al. 2000; Winfield, 2000; Haywood et al., 2001; Iwasaka et al., 2003; Reid et al., 2003 and 2008; Shen et al., 2005; Fratini et al., 2007; Kaaden et al., 2009). A value of 2.5 (or

lower) is frequently assumed, especially for transported dust (Maring et al., 2000; Reid et al., 2003; Wagner et al., 2009) for which the particle density is also supposed to decrease due to sedimentation and ageing processes. This may be the case for our samples which correspond, with the only exception of the SOP1-8 and SOP1-17 cases, to transport events. It should be mentioned, however, that in several recent studies higher dust densities, ~2.6-2.7 g cm^{-3} (e.g., McConnell et al., 2008; Schladitz et al., 2009), are frequently assumed.
In this study, the chosen value of 2.5 g cm^{-3} also agrees with the density calculated from particle mineralogical composition (weighted average of the dust density of each composing mineral). When the main composition of dust is considered (clays+quartz) we obtain indeed a density of 2.52-2.55 g cm^{-3} for the different cases.
Following the reviewer suggestion, we have specified in the text that the density is the same for all the samples, and also added a sentence reporting the range of values from literature, and past references (lines 279-282).

Concerning the comment about the volume size distribution shown in Fig. 1b, the density is not used in the calculations. The volume size distribution dV/dlogD is estimated from the number size distribution as:

$$\frac{dV}{d\log D} = \frac{\pi}{6} D^3 \frac{dN}{d\log D}.$$

P10609, L 2 : Dust particles are known to be a particle that scattering the solar light more than absorbing in the visible range. So you need to be specified the wavelength range you are talking about.
We have rewritten as: *"In case extinction is dominated by absorption, Q_{ext}~Q_{abs}, as it can be assumed at infrared wavelengths for dust in pellets,"*.

P10609, L 8 : 'Following Mie Theory for Rayleigh spherical particles'
The correction has been performed.

P10610, L11-13 : This sentence is not clear and I don't have a clue on what you are doing with these iterative non linear fitting.
The sentence has been rewritten as: *"A non-linear fit procedure is then applied to the experimental $Q_{abs}(\omega)/a$ spectrum to determine the 3N values of the oscillator parameters (ω_j, γ_j, F_j) in Eq. (6)."* We changed accordingly also the text in Section 4.2.

P10611, L9 : This discussion should occur earlier in the manuscript. What would be the influence of the shape of the aerosol? Could you evaluate the associated errors? The code to take into account the non-sphericity already exists and was developed by a French team. Did you ever contact them to run the scheme for you? Another hypothesis is that the real part of the dust refractive index has been set to 1.53. Do you think that this number is valid for all type of dust from all the sources in the world or even in West-Africa?
The discussion about the different hypothesis performed in our retrieval procedure has been moved along the main text of Section 3.1 (new Section 2.1). Concerning the assumption on the dust shape, the new text is (lines 299-303): *"Moreover, in our formulation (Eq. 4) we*

assume dust particles to have a spherical shape, and thus to be described by the Mie theory. Taking into account particles non-sphericity would require a much more complex retrieval scheme. We have decided to neglect this effect at this stage, thus keeping ourselves in retrieval conditions which are similar to those of previous literature studies.". In terms of uncertainties, we are aware about the importance of the shape on the calculation of the dust refractive index and optical properties, however we consider out of the scope of the paper this analysis.
Concerning your comments about the refractive index, as discussed in the answer to the general comment, we agree with the reviewer about the variability of the dust refractive index as a function of particle composition and origin. However, due to the absence of direct estimates of the visible refractive index for our samples, we preferred to fix it to a mean value valid for all cases.

P10613, L 7: If I understood well you corrected the measured size distributions with a mean refractive index given in the literature to retrieve the refractive index of sampled dust. Could you evaluate the errors that this hypothesis could cause to your retrievals?
The size distribution is not used in the refractive index retrieval algorithm, so the uncertainty associated to the OPC optical-to-geometrical diameter conversion does not have any influence on the refractive index results.

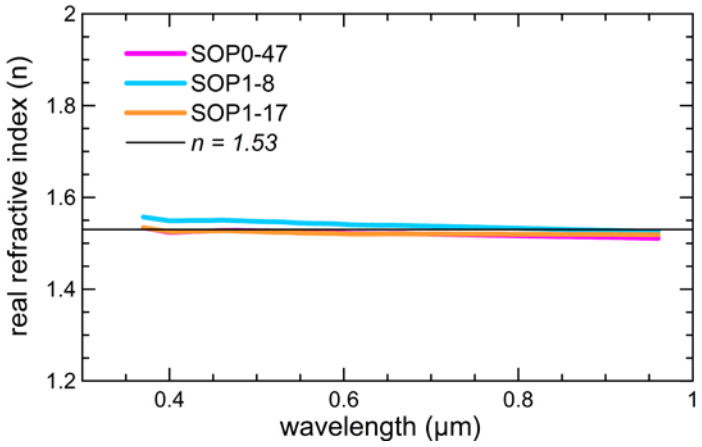
P10613, L11-12 : 'Whereas (remove for) Tamanrasset measurements '
The correction has been performed.

P10613, L18 : You are using a 5 mode fit. Is it usual ?
The number of modes necessary to fit the size distribution depends on the specific features observed in the measured number size data. Generally 3 to 5 modes are used to fit dust size distributions (e.g., Kandler et al., 2009; Weinzierl et al., 2009; Formenti et al., 2011; Ryder et al., 2013). However, in this study, in order to represent as closely as possible the measured dust number size distributions we decided to use up to 7 modes. This choice may lead to a certain uncertainty due to the high number of free parameters of the lognormal functions, i.e. 21. Nonetheless, since the main dust modes were clearly evident from size data, the modelled modes resulted very well constrained by measurements.

P10613, L20 : You meant that the left side of the tail is not well defined. You should clarify that point.
We have rewritten as: "*Because of the lower size cut of the Grimm OPC at 0.3 µm, the lower tail of the first mode at diameter <0.6 µm is not very well defined.*".

P10614 L12-13 : Does the clays partitioning differences may change the real part of refractive index ?
The real part of the refractive index at visible wavelengths is very similar for kaolinite and illite and this leads to small differences associated to their different clays partitioning. To test this statement, we calculated for the three Banizoumbou cases (which have different clays partitioning, see Table 1) the real dust refractive index in the visible from mineral data by

assuming an internal mixing hypothesis (the refractive index is calculated as the volume-weighted average of the refractive index of each composing mineral). Results are shown in the following Figure, and indicate that the different clays partitioning determines differences less than 2% in the visible. By comparison, also the assumed value of 1.53 has been shown in the plot.



P10614, L 21-22 : 'SOP1–8, emitted in correspondence of a strong Sahelian local erosion event' What does that mean ?

We have rewritten as: "*SOP1-8, sampled in correspondence of a strong Sahelian local erosion event*". Similarly, we have also rewritten the sentence concerning SOP1-17, few lines above, as: "*even if sampled during a Sahelian erosion event, SOP1-17 presents a mineralogy very similar to that of SOP0-47 and N32. This may be explained considering that SOP1-17 particles has been collected more than 3 hours after the main erosion event had occurred*".

P10614, L21-22 : Studies using lidar measurements have shown strong differences in dust optical properties according to dust sources. You also found that the mineralogical composition is changing a lot. Why would you think that the real part of the refractive index would be the same?

See the answers to the general comments.

P10619, L5: The errors associated with the real part of the refractive index are extremely low compare to the imaginary part. How could those errors be that low knowing the pellet technique limitations?

The results for n resulted much less sensitive than k to the absolute value of the Q/a ratio in this retrieval scheme. However, as stated in the text, the reported uncertainty for n (<1.5%) is the average over the whole spectral range. Maximum values of the n uncertainty may reach up to 5-6% in the 8-12 µm window region and above 17 µm.

P106021 : For the comparison of your results with the literature, it would be valuable to add the error bars associated with your results as well as the error bars from previous studies if given.

The reviewer is right since error bars are very important in comparing results from different studies. We have decided, however, not to show the error bar in the plot (Figure 7) for two

reasons: 1. The first one is that uncertainties are not always provided in the considered studies; 2. The second one is that we find the plot already quite complex (a lot of plots, large variability of several curves, etc.), and we feel that adding error bars would largely compromise the readability of the plot.

P10622, L 27 : They result incorrect ??? As I said earlier the paper need to be proof read to remove all the grammatically incorrect sentence.
We have rewritten as: "*they are incorrect*".

P10624, L 11-13 : Why is that surprising? Size does matter for the refractive index calculations. Could you introduce the main goal of this paragraph ?
As stated in the first two paragraphs of Section 6.1 (now Section 5.1), the main goal of this part of the paper is to calculate the aerosol intensive optical properties relevant for radiative transfer (and so for climatic studies and remote sensing applications) and analyse their sensitivity to the refractive index and size distribution of dust particles. The effect of the shape is not considered here. So I do not evaluate necessary to reintroduce here again the main goal of the paragraph. Instead, we agree with the reviewer that the effect of the size seen here on the dust properties is not that surprising, since it is obvious that the use of different size distributions will increase the differences in particle calculated properties. For this reason we have rewritten the beginning of the paragraph as: "*As expected, the differences between the different samples, and also between our samples and OPAC, considerably increase when the own size distribution for each case is taken into account (Fig. 8b).*"

P10624, L 18 : 'Due to dominant mode of particle at 5um in its size distribution' That is not properly said. . .. Due to the presence of a particle mode centered on 5um' or something else but you can't leave it like this.
We have rewritten as: "*mainly due to the presence of a dominant particle mode centred at 5 μm*".

P10624, L 27 : Replace 'in situ' by 'in-situ' and 'particles size distribution' by 'particle size distributions'.
"In situ", which is a Latin expression, cannot be in any case written as "in-situ". It can be written as "in situ" or "*in situ*". Conversely, "particles size distribution" has been corrected throughout the text.

Again I don't think that the next sections should appear in this manuscript. It's really interesting but need more work and deserve a paper by itself.
See the answers to general comments.

P10626, L 3-6 : What is your solution on a global scale ?
In my opinion, the solution at the global scale is to provide a more accurate description of dust properties, i.e. a new dust model to replace OPAC, which will be able to take into account for the observed differences linked to particle mineralogy. To specify this point we have added the following sentences at the end of Section 5.2 (lines 738-741): "*All these*

considerations thus evidence the necessity for a new optical model, to use in place of OPAC, able to provide a better description of the spectral and regional variability of dust properties. The use of this new model would help increasing the accuracy of satellite inversions over regions affected by the presence of dust."

P10626, L 18-20 'For example, the integrated area of kext over the 11 and 12 μm MODIS bands is 0.089 and 0.087 for OPAC, compared to 0.123 and 0.110 for the estimated maxima values of kext at the bands, and 0.067 and 0.054 for the corresponding kext maxima' I'm not sure what you are talking about? The first numbers are coming from OPAC that is what you said and then where all thoses following numbers are coming from ? Estimated from which calculations ?

The 0.123 and 0.110 values comes from the integration of the N93-own size curve (green dashed line) in the plot, while the other two numbers come from the integration of the SOP1-8-own size curve (light blue dashed line) and the N32-own size curve (blue dashed line). We have also found an error in line 21 were it was written "maxima" while instead we referred to "minima" values. The text has been rewritten as (lines 730-734): "*For example, the integrated area of k_{ext} over the 11 and 12 μm MODIS bands is 0.089 and 0.087 for OPAC, compared to 0.123 and 0.110 for the maxima values of k_{ext} at the two bands (sample N93, green dashed line), and 0.067 and 0.054 for the corresponding k_{ext} minima (samples SOP1-8, light-blue dashed line, and N32, blue dashed line).*".

Fig. 1 : SOP1-8 shows a mode at _20um similarly to N92 and N32. Then, it is hard to believe that the size distribution is playing a major role between these samples. These samples may be the worst to work with according to the a/lambda conditions. Could you comment on that ?

As stated in the text, we do not assume that the size distribution plays a major role, since main differences between the samples are basically associated to the mineralogical composition of the particles. However, we find in our data some features which may be linked to the differences in the size distributions, which we consider appropriate to discuss in the text. However the reviewer is right and we probably emphasised too much the role of the size distribution in the text. So, we decide to eliminate the comment on the size distribution from the Abstract ("*Size distribution, and the coarse fraction in particular, plays also a role in determining the refractive index*"). In the Conclusions we rewrote (lines 803-805): "*The variability of n and k is linked to the variability of particle mineralogy, mainly clay amount and speciation, and to a lesser extent, size distribution, in particular the coarse fraction.*"

Concerning the comment about the fact that these samples may be the worst to work based on the a/lambda condition, we agree with the reviewer with the fact that the larger the size the worst may be the accuracy of the assumption. Nonetheless, these samples verify the condition at least for the 94% of the particles, which we assume to be sufficient to verify our assumption. We also invite the reviewer to address to the answer to the general comment for a further discussion on that topic.

Figure 7 : 'The real and the imaginary parts of the Longtin et al. (1988) refractive index are plotted against the right side y-axis for both plots. ' Do you mean that the Longtin RI axis are

located on the right side while all the other studies are associated to the left side ? If yes it needs to be rephrased and probably to appear later in the caption. Also in the caption you refered to curve 4. What are they ?

The reviewer is right and the sentence referring to Longtin et al. (1988) was not clear; we corrected it and included it at the end of the caption. With curve 4 we referred to the plot number 4 in the Figure, so that by Carlson and Benjamin (1980); this point has been specified in the text. The caption has been rewritten as:" *Figure 7. Comparison of our results with other direct and indirect estimates of the dust refractive index as reported in the literature. The plot reports the real and the imaginary parts of the refractive index for: 1. Volz (1972) (indicated as VO72 in the plot): rainout dust aerosols collected in Germany and composed of a mixture of soil particles, fly ashes, and pollen (spectroscopy method; also used as the "dust-like" model in Shettle and Fenn, 1979); 2. Volz (1973) (indicated as VO73): Saharan dust collected at Barbados, West Indies (spectroscopy method); 3. Fouquart et al. (1987) (indicated as FO87): Saharan sand collected at Niamey, Niger (spectroscopy method); 4. Carlson and Benjamin, (1980), Sokolik et al. (1993) (indicated as CA80): mineral dust model, as used in the OPAC and GADS databases (based on a synthesis of measurements on Saharan dust or generally on desert aerosols); 5. Longtin et al. (1988) (indicated as LO88): dust sand, i.e. modelled as an internal mixture of hematite (10% by volume) and quartz. In the internal mixing hypothesis the dust refractive index is calculated as the volume average of the refractive indices of individual minerals; 6. OPAC desert model: modelled as an internal mixture of a water soluble component at 80% relative humidity (5% by volume), and mineral dust in nucleation (3% by volume), accumulation (72% by volume), and coarse (20% by volume) modes (Hess et al., 1998); the dust refractive index for the three modes is that reported by CA80. The region in blue in the plot indicates the range of variability of the results of this study. The legend in bottom panel identifies the line styles used in the plot for literature data. Refractive index y-axes are located on the left side of the plots, with the only exception of LO88 for which the y-axis is on the right side.*".

Figure 8a : I still don't see the point of that figure. Of course the size distribution is playing a major role in the RI calculations. Using the same aerosol size distribution for all the samples is gonna smooth all your rg, w, kext calculations.

As stated in the manuscript, and also discussed in the previous answers, the objective of this Figure is to show the absolute value and variability of the dust intensive optical properties as a function of the refractive index and particle size. These quantities are generally used in climate and remote sensing applications, so their calculation may be of relevance for people working in these fields.

Figure 9. : This figure deserves its own legend. The readers don't have to go to Figure 8 (and not Figure 9 as you wrote in the caption) to know what colors correspond to what sample . . ..

The caption has been rewritten as: "*Figure 9. Mass extinction efficiency (k_{ext}, m^2 g^{-1}) calculated for the five dust cases in the 7.5-12.5 μm spectral range. According to Fig. 8, continuous and dashed lines corresponds to k_{ext} calculations performed by considering respectively the same size distribution for all dust cases (continuous lines) and the own size distribution measured in correspondence of each sample (dashed lines). Vertical lines and*

the two shaded areas refers to the following different satellite remote sensing channels: (grey dashed lines) six AIRS channels for dust retrieval in the thermal infrared (8.10, 8.15, 9.31, 10.36, 11.48, 11.86); (black dashed line) IASI channel for dust optical depth retrieval at 10 µm; (grey shaded areas) the two MODIS broadband channels (10.78-11.28 and 11.77-12.27 µm) used for Sea Surface Temperature (SST) estimation.".

Appendix A : Again, about the AOD/PM relationship, in your case the aerosol mass is a linear function of the AOD due to homogeneous aerosol vertical distribution. Unfortunately, the aerosol hygroscopicity could impact the relation between column integrated measurements and aerosol mass within the BL. Do you have any RH profiles, could you use the RH profiles from CALIPSO ?

The reviewer is correct as the RH may affect aerosol optical properties and thus column integrated measurements. However, the effect of RH is not so relevant for dust particles, which are mostly hydrophobic, especially close to their source region (Kaaden et al., 2009). Thus we may assume water vapour to have a negligible effect on our retrieval procedure for dust observations at Tamanrasset.

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