ANONYMOUS REFEREE #2

The manuscript shows interesting results of measurements in an area little explored. Composition analysis was well done, combining different complementary methodologies. However, the authors made strong hypothesis without enough justification and thus the main conclusions are not as strong. Beside, manuscript english writing needs improving in some points.

Although the paper has potential interest for the ACP audience, I find it in need of major changes. My concerns are:

1) Calibration of SEM XEDS are not shown nor referenced from another paper. Reliability is only accessed by comparison with EDXRF, for which the calibration was not shown/discussed as well. If they used NIST standard for calibration, or a different one, should be clarified.

Scanning Electron Microscopy combined with X-ray energy dispersive (XEDS) microanalysis requires calibration of the electron column, for morphological measurements, and of the energy dispersive spectrometer, for XEDS microanalysis. Differently from other analytical instruments, like EDXRF, both procedures are not performed on daily, or anyway frequent, basis, since a periodical (about every 3 – 6 months) recalibration is sufficient to maintain the reproducibility of signals (mainly: secondary electrons, backscattered electrons and X rays emitted from sample), especially if samples from the same type of matrix are analysed, on routine. This is also the case of the Philips XL30 ESEM employed in this study, as by this instrument particulate matter samples collected on filter membranes are almost exclusively analysed, on laboratory routine. This is also the reason why neither calibration details are generally reported in the scientific literature on SEM XEDS microanalysis of environmental particulate matter, nor calibration curves are reported as well. Nevertheless, a short sentence has been added in the text of revised Manuscript (Section 2.2) indicating that the calibration procedures are in line with the US EPA Guidelines (2002) on the application of SEM XEDS microanalysis to particulate matter samplers. Moreover, it should be considered that quantification methods properly targeted on the SEM XEDS microanalysis of individual particles from environmental matrices does not exist, as discussed in the Manuscript (Section 2.3), and standard materials of environmental particulate matter properly dedicated to the quantification of the elemental composition of individual particles (relating to individual particles from environmental matrices) are not available. For this reason, the authors applied an internal standard approach to achieve the goal of quantification of particle elemental composition, which is an unavoidable step in the analytical structure of this study, and assessed the reliability of this approach (and of the procedure of particle allocation to mineral classes) by comparison with the quantitative results of elemental composition obtained by EDXRF on the bulk PM10 dust samples. Finally, given the above considerations, the term 'semi-quantification' (and related terms) is more appropriate than quantification, in the case of SEM XEDS individual particle microanalysis applied to environmental matrices.

2) PCA cannot be used for the proposed analysis because it allows negative mass/concentration. The reference method in case is PMF (positive matrix factorisation).

In this work the PCA is employed to discuss results of the elemental ratios obtained by SEM XEDS microanalysis of individual dust particles; no limitation exists, to the author's knowledge, in applying the PCA to this type of data. The term 'apportionment' in this study was used to indicate the assignation of each dust particle to the proper mineral group, and not referring to the field of the source apportionment (where Chemical Mass Balance, Multilinear Engine, and Positive Matrix Factorization models, are reference methods). As a matter of fact, indeed, mass apportionment is neither presented nor discussed all over this work. Mass data have been treated by a mass closure approach in Section 3.3, on the results of the assignation procedure, to the goal of assessing the reliability of this procedure versus the quantitative determination of the mineralogical composition by XRD. For sake of clarity, the term 'apportionment' has been replaced by the term 'assignation' (and related verb) in the revised Manuscript.

3) Discussion on size distribution is problematic because samples were produced in the lab. Authors did not mention, nor discussed if their method of resuspensions actually reproduce the same size distribution as would be measured in the atmosphere.

The focus of this study is the characterization of PM10 mineral dust at the dust source, and not in the atmosphere. This has been better clarified in the revised Manuscript. The approach of laboratory resuspension of dust by mechanical ventilation along an adequate time and by simultaneous sampling in the controlled environment of the chamber, is widely employed in the research field of the mineralogical and microphysical characterization of airborne crustal dust (e.g. Gill et al., 2006 and references therein; Feng et al., 2011; Aimar et al., 2012; Dobrzhinsky et al, 2012). By this approach, indeed, it is possible to reproduce with good approximation the conditions of the field sampling at a dust source, and the size distribution of the resuspended particles is negligibly affected by the laboratory procedure, with respect to the original distribution in the source material. This is extensively treated by Gill et al. (2006). Moreover, it should be taken into account that the PM10 samples of this work are obtained by a PM10 sampling head compliant with EN12341 standard (as reported in the paper by Pietrodangelo et al., 2013, cited in Section 2.1 of the Manuscript). Therefore particles in the samples of this work have aerodynamic diameter below 10µm and can be considered, with sufficient approximation, as if they were collected at the dust source. To better clarify this point, some comments have been added in Section 2.1 of the revised Manuscript. Under the above arguments, the mineralogy of the PM10 particles collected by chamber resuspension in this study can be considered representative of the mineralogy of the same particles in the geological source materials. The approximation by which this assumption is made depends strictly on the confidence on the ability of this approach, as reported in literature, of reproducing the conditions of field sampling at a dust source, as above discussed, at least with respect to the interference of the PM10 sampler on the dust source itself.

Considering that about 95% of mineral particle included in this study show physical size below, or equal to, 5 µm, our results are also in line with arguments reported by Mahowald et al. (2014): *"Accurate representation of the dust particle size distribution (PSD) in the atmosphere begins with a parameterization of the dust PSD at emission. Note that the different measurements of the size distributions at emission are all in rough agreement for dust aerosols smaller than 5 µm in diameter …. This is quite remarkable, considering that these measurements were taken over different soils, in different source regions, and using different techniques. For larger particles (> 5 µm), the size distributions do differ substantially, a possible cause of which is discussed in the next section. In order to parameterize the dust PSD at emission in models, the dependence on wind speed and soil properties, such as soil PSD, needs to be understood. A number of studies have reported measurements of the dust PSD at different values of the wind friction speed…. Most of these measurements show no dependence of the dust PSD on the wind speed at emission …….*
On balance, the measurements indicate that the dust PSD is independent of the wind speed at emission. This conclusion is supported by the observation of Reid et al. (2008) that the PSD of dust advected from individual source regions appeared invariant to the wind speed at emission."

4) All the discussion / conclusion on the RT calculations are simple direct implications of the ADHOC index of refractions chosen from the literature.

The choice of adopting refractive index (r.i.) data from literature was driven by the fact that the 6SV code requires as input the spectral trend of the real and imaginary parts of r.i., and these measurements were not available from our laboratory. Concerning the volcanics sample, it was not possible to build the real and imaginary parts of r.i. on the basis of the mineralogical composition determined, e.g. introducing a complex mixing model, due to the lack of numerical data, in literature, in the wavelength range required for simulations by the 6SV code. Indeed, the availability of the spectral trend of the imaginary part of r.i. is limited to 2500 nm for most minerals. Moreover, available spectral data of the r.i. account only for ab. 70% of the mineralogical composition of the volcanic sample; the uncertainty which would be introduced by not considering mineral phases, such as plagioclase and pyroxene, for which appropriate data are not available in literature, would be thus large. Therefore, the choice of assuming the r.i. spectral trend of the “water-insoluble” aerosol component reported in Kokhanovsky (2008), which is rich in silicate minerals similarly to the volcanics dust of this study, was considered more suitable. Concerning travertine, finally, the assumption of r.i. of calcite from literature is explicable on the basis of the travertine mineral composition (at least 95% calcite), as discussed in the Manuscript.

The authors have added results about radiative effect by introducing the radiative forcing efficiency (RFE) for the travertine and volcanic to define better the role of the radiative transfer calculation in this work. In fact, the retrieval of RFE requires models for aerosol-free fluxes calculated *in situ* only from RT runs. Furthermore, the fluxes simulations are normalized to the aerosol optical thickness (RFE) to evaluate the radiative forcing of the two components of local dust independently from the aerosol loading.

Some specific suggestion to the authors follows bellow.

a) Modify the abstract and introduction to better state what your work is about and why it is important.

The abstract and the introduction have been revised following the Reviewer’s suggestion.

b) Use Aeronet data for comparision. There are many years of data from Rome and from L'Aquila and you could select periods when dust concentration was expected to be high. From the inversion you will have not only the size distribution, but also the asymmetry parameter and single scattering albedo... and even the real and imaginary parts of the refractive index!

Considering the goals of this work, declared by the authors, the AERONET data are not useful for any comparison. The AERONET data (size distribution, refractive index, asymmetry parameter, single scattering albedo..) are referred to the mixed aerosol in the atmospheric column. This basic characteristic of the measurements returns column-integrated products not comparable with the results of this work where the simulation has been performed under conditions related to an atmosphere where the only aerosol component is the PM_{10} mineral dust (volcanics or travertine, alternatively), at dust source. Furthermore, Rome Tor Vergata AERONET station is not close enough to the identified dust source, for considering AERONET products representative of microphysical and optical properties of the local dust. L'Aquila station is farer than the Rome station, increasing the distance and discarding the chance to consider the samples of the presented work as the coarse component of the products obtained from AERONET radiative measurements.

c) Use transmission or reflectance methods in the lab to measure the resuspended material deposited on the filters. That will give you scattering and absorption directly.

The reviewer suggests methods for radiative measurements which could be applied if appropriate equipment were available in laboratory; this is not the case of this work. For these reasons, the authors have applied an approach which allows to meet data and tools actually available and which is suitable for the aerosol optical properties and radiative effects evaluation, as declared in the goals of this work.

ANONYMOUS REFEREE #2 – SUPPLEMENT

In the attached manuscript I tried to identify all the typos and points were attention is needed.

p. 13348 L. 1. As the first sentence of the abstract, this is a bit confusing.

"The first sentence has been rephrased in: *In this work, new information has been gained on the laboratory resuspended PM_{10} fraction from geological topsoil and outcropped rocks representative of Rome area, Latium. Mineral composition, size distribution, optical properties and the radiative efficiency of dust types representing the compositional end-members of this geological area have been addressed"*

p. 13348 L. 5. It is also unclear which techniques you applied to which type of aerosols.

*"A multi-disciplinary approach was used, based on individual-particle scanning electron microscopy with X-ray energy-dispersive microanalysis (SEM XEDS), X-ray diffraction (XRD) analysis of dust, size distribution of mineral particles, and radiative transfer modelling (RTM).The mineral composition of Rome lithogenic PM 10 varies between an end-member dominated by silicate minerals and one exclusively composed of calcite."*This sentence has been rephrased in*:* *"A multi-disciplinary approach was used, based on chamber resuspension of raw materials and PM10 sampling, to simulate field sampling at dust source, scanning electron microscopy / X-ray energy-dispersive microanalysis (SEM XEDS) of individual mineral particles, X-ray diffraction (XRD) analysis of bulk dust samples, number and volume size distribution (SD) building from microanalysis data of mineral particles and fitting to Log-normal curve, and radiative transfer modelling (RTM) to retrieve optical properties and radiative effects."*

p. 13348 L. 20. In the atmosphere? or did you resuspended in the lab some material collected in the field?

This point has been clarified in the revised Manuscript. Please refer also to reply to General comments #3.

p. 13348 L. 25. please define the **acronym**.

"BOA" has been defined *"Bottom Of Atmosphere"*.

p. 13348 L. 25. but have you actually measured particles with this composition in the atmosphere? how much in # and mass are their contribution?

"The downward component of the BOA solar irradiance simulated by RTM for a ***volcanics-rich or travertine-rich atmosphere*** *shows that volcanics contribution to the solar irradiance differs significantly from that of travertine in the NIR region, while similar contributions are modelled in the VIS."* The sentence has been re-written to better address that the simulation is performed assuming an atmosphere in which <u>the only aerosol component</u> is, alternatively, or volcanics PM10, or travertine PM10 dust. *"The downward component of the BOA solar irradiance simulated by RTM for* ***an atmosphere composed of pure volcanics and pure travertine*** *shows that volcanics contribution to the solar irradiance differs significantly from that of travertine in the NIR region, while similar contributions are modelled in the VIS."*Please refer also to reply to General comments #3.

p. 13349 L. 1. not true in general. think for instance over the ocean, or over tropical forests. if this is true for continental europe or italy, please cite a reference.

"Airborne geological dust from topsoil and surface rocks represents a ***critical contribution*** *to the total mass, composition, microphysical and optical properties of the atmospheric aerosol."* This sentence has been rephrased in*:* "*Airborne geological dust sourced from topsoil and surface rocks critically contribute to the total mass, composition, microphysical and optical properties of the atmospheric aerosol in continental regions, and largely impacts different Earth's compartments by transport and deposition (Scheuvens and Kandler, 2014)".*

p. 13349 L. 6. There are many other previous papers that showed complex organic molecules and mineral components in particulate matter. It is not a consequence of the occurrence of lithogenic dust.

This sentence has been deleted in the revised Manuscript.

p. 13349 L. 26. **indirect effect** refers to aerosol changes in the radiation balance through **cloud-aerosol interactions**.

*“Airborne lithogenic dust plays a role both in the direct mechanisms (light scattering and absorption) and in the indirect mechanisms (**warming or cooling of the atmosphere**) which tune the Earth’s radiative budget (Sokolik et al., 2001; Choobari et al., 2014).”* The sentence has been rewritten *“Airborne lithogenic dust plays a role both in the direct mechanisms (light scattering and absorption) and in the indirect mechanisms (**cloud-aerosol interactions**) which tune the Earth’s radiative budget (Sokolik et al., 2001; Choobari et al., 2014).”*

p. 13349 L. 28. If you are using "indirect effect" differently than current current scientific consensus (e.g. IPCC reports) then you should better properly define it.

Please refer to the reply to reply to previous comment.

p. 13350 L. 1. Cloud-Aerosol interaction can be affected IF heterogeneous chemistry happens on particle's surface, but it is not a necessary condition for it to happen.

“While indirect effects depend on the heterogeneous chemistry occurring at particles surface (Levin et al., 1996; Buseck and Pósfai, 1999; Sokolik et al., 2001; Krueger at al., 2004; Kandler et al., 2007), the light scattering and absorption are mostly controlled by the mineralogical composition, shape features and microphysical properties of geological particles (D’Almeida, 1987; Kalashnikova and Sokolik, 2002 and 2004; Kokhanovsky, 2008; Hansell et al., 2011).” The sentence has been re-written in: *“Considering direct effects, airborne lithogenic dust plays a key role in the light scattering and absorption, which are mostly controlled by the mineralogical composition, shape features and microphysical properties of geological particles (D’Almeida, 1987; Kalashnikova and Sokolik, 2002 and 2004; Kokhanovsky, 2008; Hansell et al., 2011).”*

p. 13350 L. 22. please define or maybe rephrase (rain aggressiveness). please define (FFAO index).

“Latium is also affected by high rain aggressiveness, within the scale of FFAO index, and is characterised by a large surface where poorly-developed soils and debris deposits are present, which are easily affected by massive erosion”. This sentence has been deleted in the revised Manuscript.

p. 13350 L 25. you can expect, but if you do not measure in the atmosphere you will never know.

“Considering also the high anthropic impact on the Latium territory, it has to be expected that the re-suspension of mineral dust from local lithological domains is non- negligible in this region.”

Following the Reviewer’s suggestion, this point has been further discussed in the revised Manuscript. Some comments, on the frequency and the influence on the mass concentration, of local crustal dust resuspension to the ambient PM_{10} in the Rome area have now been added in the Introduction, and two figures (Figures 2S and 3S) have been added to the Supplementary materials (Supplementary materials_revised), to support the discussion on this item. To summarize briefly, a long period has been analysed (2005 – 2011 and 2005 – 2015, depending on the site), for which data are available at two different background sites in Rome area (as showed in Figures 2S and 3S). The goal was to evaluate the number of days and the entity of the crustal contribution, on days of desert dust intrusion at-ground (DD-days) and on days showing a large crustal contribution (above 50% of total PM_{10} mass) without occurrence of desert dust at-ground, indicating a crustal contribution from local sources (LD-days). Interestingly, among the above described days, the mass concentration of the crustal matter on LD-days is in many cases comparable with that observed on DD-days.

p. 13351 L. 2. why not using **SFC (surface)** as it is more standard?

In literature, the radiative effects are referred as TOA for the Top Of Atmosphere and BOA for the Bottom Of Atmosphere, as reported in the NASA website for the AERONET inversion products: http://aeronet.gsfc.nasa.gov/new_web/Documents/Inversion_products_V2.pdf

p. 13351 L. 9. didn't you actually measured the size distribution of the particles in the atmosphere?

Size distributions have been obtained from the data set of SEM XEDS microanalysis of individual mineral particles of our samples, as discussed in the Manuscript. Please refer also to reply to

General comments #3. Furthermore, the sentence:
"To investigate relationships among these different aspects, a multi-faceted analysis was performed, on the basis of the following approaches: individual-particle scanning electron microscopy combined with X-ray energy-dispersive microanalysis (SEM XEDS), bulk mineralogical analysis by X-ray diffraction (XRD), parameterization of the size distribution to log-normal function, and radiative transfer modelling (RTM)." has been rephrased in: *"To investigate relationships among these different aspects, a multi-faceted analysis was performed, on the basis of the following approaches: chamber resuspension of raw materials and PM10 sampling, to simulate field sampling at dust source, scanning electron microscopy / X-ray energy-dispersive microanalysis (SEM XEDS) of individual mineral particles, X-ray diffraction (XRD) analysis of bulk dust samples, number and volume size distribution (SD) building from microanalysis data of mineral particles and fitting to Log-normal curve, and radiative transfer modelling (RTM) to retrieve optical properties and radiative effects.*

p. 13351 L. 17 This is not clear. Do you mean you collected 4km2 of samples? Or that all the sampling sites are located within 4km2? Or that each site is relatively uniform so that the sample is representative of at least 4km2 around the sampling position?

"Collection areas of about 4 km were selected on the basis of criteria established after geological analysis of the Latium region, within main local geodynamics domains, namely: the volcanic complexes, the marine (limestones, marlstones and sandstones) deposits, the siliciclastic series (mainly flysch) and the quaternary deposits (mainly travertines)." This sentence has been rephrased in: *"On the basis of criteria established after geological analysis of the Latium region, the following geodynamics domains were considered: the volcanic complexes, the marine (limestones, marlstones and sandstones) deposits, the siliciclastic series (mainly flysch) and the quaternary deposits (mainly travertines).Sampling areas of about 4 km2 were selected within each local geodynamics domain; a number of dust collection points was identified, within each area, to obtain sub-samples of raw material, from which the final samples were obtained. The number of sampling areas varies within each domain, depending on the geographical extension and the geological complexity of the domain".*

p. 13351 L. 23. this should be clearly stated in the abstract and introduction.

"PM 10 dust was laboratory re-suspended from the bulk rocks samples, and from road dust, by a re-suspension chamber, and collected by low-volume sampling on polycarbonate membranes for SEM XEDS microanalysis."

This aspect has been clearly stated in the title, abstract and introduction.

p. 13353 L. 3. Launching?

"launch" has been replaced with "launching".

p. 13353 L. 8. Or?

"and" has been replaced by the form "both on": *"....both on field areas and on individual particles, by using the EDAX control v. 3.3 package (EDAX Inc., 2000)".*

p. 13354 L. 9. Secondary target?

The term "target" has been added in this sentence: *The mineralogical characterization of dust samples has been carried out on the 50 μm sieved dust fraction, by an automatic diffractometer Scintag X1, equipped with a Si(Li) detector using a Cu Kα target, ..."*

p. 13354 L. 28. At some point you should show the calibration curves for your instrument, or reference the paper where that was done.

This point has been extensively discussed in the reply to General comments #1.

p. 13355 L. 20. This might not be clear enough for those who are not specialized on EDAX. We have an EDXRF in our lab., for instance, and it is not possible to get the matrix losses from the quantification routine itself.

The theory of microanalysis by SEM XEDS is widely treated in literature. The estimation of the Z (atomic number), A (absorption) and F (secondary fluorescence) factors to take into account the matrix effects is treated by the ZAF algorithm, which allow to improve quantification obtained by the application of the Castaing's first approximation. The Z, A and F factors are commonly estimated in the sample matrix by the quantification routines included in the SEM XEDS software packages, based on the net intensities measurement of the X-rays spectrometer and on the specific instrumental parameters of each scanning electron microscope.

p. 13356 L. 6. Please clarify…Do you mean total weight of the particle that could be identified < 50%?

The sentence related to this comment has been rephrased as*:"….total percent weight (%wt) of the particle that could be identified below 50%….".* Please note that, following suggestions from the Reviewer #1, the part of Section 2.3 concerning the internal standard approach to quantification of particle elemental composition, where the above sentence is placed, has been moved from the Manuscript to Appendix I (new) in the Supplementary materials.

p. 13357 L.3. But that is only representative of what you would observe in free atmosphere if your resuspension method precisely mimic nature. Do you have evidence that you method doesn't prefer, for instance, to lift large particles in detriment of small particles?

We have experimental evidence that about 95% of particles included in this study have physical size below or equal to 5 µm; this is in line with literature on this issue (e.g. Gill et al., 2006 and references therein; Feng et al., 2011; Aimar et al., 2012; Dobrzhinsky et al, 2012), as extensively discussed in the reply to General comments #3.

p. 13357, Log normal curve function. Why fitting the data instead of showing the measured sized distribution? Besides, why fig. 4 doesn't look like a fitted size distribution?

The 6SV code requires, among other inputs, the parameters (µ and σ) of the probability density function (PDF). Therefore, as widely explained in the Manuscript, the number size distribution obtained by the experimental data of SEM XEDS microanalysis has been fitted to Log normal curve, as commonly performed in the literature on this issue (e.g. as in Mahowald et al., 2014)., and the PDF parameters have been obtained. Figure 4 shows the volume size distributions of some minerals and of the different dust types, in the PM10 fraction, and are obtained directly from the experimental data of SEM XEDS microanalysis, as discussed in the Manuscript. Therefore size distributions in Figure 4 are not the result of a curve fitting.

p. 13357 L. 25. please rephrase

"An atmospheric radiative transfer code was employed, generally used in the remote sensing, to retrieve the optical and radiative dust properties." This sentence has been changed in *"An atmospheric radiative transfer code was employed to retrieve the optical and radiative dust properties."*

p. 13358 L. 6. Dust particles are definitely not spherical. How much wrong can your result be? Can you give an estimate?

In this work, the assumption of particle sphericity has been adopted, due to the requirements of the 6SV code for radiative transfer modelling (as discussed in the Manuscript: *"This code is able to retrieve optical properties of the aerosol and to model the atmospheric radiative field by using the aerosol microphysical properties, under the hypothesis of spherical and dry particles."*). This has been better clarified in the revised Manuscript, as follows:

"Physical size of particles was assumed as the diameter of the equivalent spherical cross sectional area (ESD) (Reid et al., 2003; Kandler et al., 2007; Choël et al., 2007) measured by SEM". This sentence has been changed in: *In this work, the assumption of particle sphericity has been adopted, due to the requirements of the 6SV code for radiative transfer modelling. Therefore, physical size of particles was assumed as the diameter of the equivalent spherical cross sectional area (ESD) (Reid et al., 2003; Kandler et al., 2007; Choël et al., 2007) measured by SEM*."

As explained in text, all parts of the study have been performed under the assumption of particle sphericity. As regards the simulation, an estimation of the accuracy can be performed in case of availability of measurements, or of ability of the model in simulating optical properties and radiative effect with non-spherical aerosol. This is not possible with the 6SV, as explained in [Kotchenova et al., 2008]: "*We also mention that all RT codes involved in this study used aerosol phase functions that were calculated on the basis of the Mie theory for homogeneous spheres. Such an assumption of sphericity is not valid for desert dust aerosols, which consist of mainly non-spherical particles with aspect ratios 1:5.*" The aspect ratio of the local dust used in the presented work is between 1:1 and 1:4 with a probability of 88% for a samples of 4800 particles. This value attests that the simulation of optical properties and the evaluation of RFE have been performed within the validity domain for the aerosol shape where the 6SV model meets the accuracy requirement of 1% for simulation studies [Kotchenova et al., 2008].

p. 1 13358 L. 21. Thus,

"By this way," has been changed with *"Thus,"*

p. 13358 L. 26. why not using radiosondes from Rome's airport? or even reanalysis over the region? Should give an estimate of how wrong the result can be by doing this crude approximation.

"Concerning meteorological parameters, the profiles of temperature, pressure and humidity were assumed by the 1976 U.S. Standard Atmosphere included in the 6SV code."

The radiosondes are useful if a comparison with radiative measurement is performed. In this work, the radiative effects have been simulated to evaluate the RFE of the two components of local dust. The variability of the meteorological parameters induces an error on RFE evaluation which is generally negligible with respect to the absolute values of RFE. As reported in Garcia et al., 2008 "*The flux calculations are performed for multi-layered atmosphere with US standard atmosphere model for gaseous distributions and single fixed aerosol vertical distribution (exponential with aerosol height of 1 km). The deviations of these assumptions from the reality are also potential source of errors, although, our tests did not show any significant sensitivity of flux estimates to these assumptions. Differences less than 1 W/m2 due to different vertical profiles were observed on the downward solar flux at the bottom of the atmosphere.*"

p. 13359 L. 5. higher than what? you did not mention other AOD value before.

*"In this study, however, an **higher** value of aerosol optical thickness, τ 550 = 0.7, was chosen...."*

The adjective has been corrected *"In this study, however, an **high** value of aerosol optical thickness, τ 550 = 0.7, was chosen..".*

p. 13359 L. 8. This is only true if other aerosol sources in the region always give contributions of AOD << 0.7. Please cite the previous studies how showed that. or use AERONET data from Rome or L'Aquila. In this case you could even get an inverted size distribution and evaluate if the strong dust episodes indeed happen or not in your region.

The authors have previously explained that the radiative simulation are referred to one component of the local dust, as yet discussed in the reply provided concerning p.2 L.25, and any comparison to radiative measurements or AERONET products are not useful for the purposes of this work.

p. 13359 L. 10. chosen from what? Where? since you got a refraction index form the literature and are running a Mie code you should calculate g and w

"Among optical properties, the single-scattering albedo and the asymmetry parameter were chosen, as they are crucial to perform analysis of the aerosol contribution on the radiative field (Dubovik et al., 2002; Kassianov et al., 2007)." The sentence has been rewritten *"Among simulated optical properties, the single-scattering albedo and the asymmetry parameter were presented, as they are crucial to perform analysis of the aerosol contribution on the radiative field (Dubovik et al., 2002; Kassianov et al., 2007)."* The Mie theory is implemented in 6SV model and its runs simulate the optical properties, including single-scattering albedo and asymmetry parameter, and the radiative quantities describing the radiative field in the Earth/Atmosphere coupled system.

p. 13360 L. 5. Legend of Table 1 should properly explain the units. What is % rsd? What is Δ+-prop.err.? Why the consistency is not shown for travertine?

The editorial rules of ACP indicate that extended legends should be avoided. Anyway the units in Table 1 have been described in the footnotes at bottom of Table 1. Consistency of the microanalysis on extended fields of the sample with results by EDXRF is not shown for travertine, as field acquisitions by SEM XEDS have been not performed on this sample, given the basically constant calcite concentration in the matrix of this sample.

p. 13360 L. 11. Why do you say XEDS is less reliable than EDXRF? To assess that you should compare both to the same PM standard from NIST.

Arguments concerning this issue have been extensively discussed in the reply to General comments #1.

p. 13361 L. 4. Did you use a NIST standard or not? Line 8, last page = you say "dust sample".

Arguments concerning this issue have been extensively discussed in the reply to General comments #1. Particularly, please consider the following part of the reply to General comment #1: "it should be considered that quantification methods properly targeted on the SEM XEDS microanalysis of individual particles from environmental matrices does not exist, as discussed in the Manuscript (Section 2.3), and standard materials of environmental particulate matter properly dedicated to the quantification of the elemental composition of individual particles (relating to individual particles from environmental matrices) are not available. For this reason, the authors applied an internal standard approach to achieve the goal of quantification of particle elemental composition, which is an unavoidable step in the analytical structure of this study, and assessed the reliability of this approach (and of the procedure of particle allocation to mineral classes) by comparison with the quantitative results of elemental composition obtained by EDXRF on the bulk PM10 dust samples."

p. 1 13361 L. 18. PCA cannot be used in your case because itc allows negative concentrations (or mass). PMF (positive matrix factorization) is the reference method in this case.

In this work the PCA is employed to discuss results of the elemental ratios obtained by SEM XEDS microanalysis of individual dust particles; no limitation exists, to the author's knowledge, in applying the PCA to this type of data. This issue has been yet discussed in the reply to General comments # 2.

p. 13362 L.7. What did you do in order not to get any negative values in figure 2? If you modified the standard PCA technique you should explain what was done…

Please refer to reply to the previous comment and to General comments # 2.

p. 13363 L. 16. I don't see how this inference can be made, since you did not measure atmospheric aerosol particles. For the lab. Method you used, you should already know if weathering is most important.

In the discussion related to this point of the manuscript, the term 'weathering' is used to indicate processes of rock alteration, and not weathering from atmospheric factors. The suitability of using this term in this case is linked both to the fact that we are discussing the possible lithological processes which are responsible of the mineralogical composition of the PM10 dust samples obtained "at source" from the outcropped rocks (or topsoil, depending on the samples), as clarified and discussed in the previous comments, and to the fact that this term is commonly used in the geochemistry research field to indicate rock alteration processes. The sentence related to this comment has been rephrased as follows:

"The mineralogical composition of the silicate component in marlstones and siliciclastics dust is strictly related to the originating materials. Rock-forming processes (erosion, fluvial and marine transport, sedimentation) support, in this case, the presence in the PM10 fraction, as detected by XRD, of stable silicates (plagioclase and quartz), the reduced presence of inosilicates and the presence of alteration by-products, such as phyllosilicates. Different processes must be considered in volcanic rocks, which explain the mineralogical composition of silicates observed in the PM10 resuspended from this geological material; specifically, crystallization is the main responsible process, in this case. Thus, the presence of most minerals observed in the PM10 from volcanic rocks is coherent with the magmatological framework of Central Italy. Differently from the above considerations, however, the association kaolinite – quartz, observed by SEM XEDS microanalysis in this PM10 dust type, has to be ascribed to rock alteration (weathering). In this case quartz is

thus the product (with kaolinite) of the hydrolysis reaction of feldspars (Jackson et al., 2010), and not a crystallization-derived phase."

p.1 13363 L. 19. Presence where?

Please refer to reply to previous comment.

p. 13368 L. 17. Isn't the lab method to produce these particles much more important?

Please refer to reply to General comments #3, where this issue has been extensively discussed.

p. 13369 L. 19. Why is this figure so much different from fig. 4? In Fig. 4 the largest size are > 5 µm, but in figure 5 it is < 2 µm. The max concentration is also different. The data points should include the uncertainties as well (and those should be used in the fit). Moreover, as you don't see the decrease for large radius, the uncertainty associated with the fitted std will be very large and should be discussed. Last, quality quality of this figure does not fit publication standards. Are you should you included the right figure?

Figure 5 shows the probability density function (PDF) obtained from the fitting to Log normal curve of the number size distributions experimentally obtained by SEM XEDS microanalysis data, which is reported versus the physical radius of particles, while Figure 4 shows volume size distributions experimentally obtained by the same SEM XEDS dataset, which are reported versus the aerodynamic diameter of particles, therefore these two figures are necessarily different.

The whole procedure to obtain results of figures 4 and 5 is described in details in the Manuscript.

The uncertainty of each bin was estimated associating a Poisson error to the bin weight (Liley, 1992), that is calculating the square root of the total counts of particles observed in each size range.

Figure 5 has been replaced in the revised Manuscript, including uncertainties. Furthermore, as the PM10 samples of this study have been obtained by sampling with a PM10 sampling head compliant with EN12341 standard (as reported in the paper by Pietrodangelo et al., 2013, cited in Section 2.1 of the Manuscript), particles have aerodynamic diameter below 10µm, which is coherent with the fact that in the PDF data related to particle radii larger than 3 µm are not present (considering an average particle density of 2.71). Furthermore, our results concerning the fitted PDFs are in line with results reported by Mahowald et al. (2014), which review the data reported by many studies dealing with size distribution of mineral dust samples obtained by chamber resuspension or by field sampling at source. An extract from this paper, reporting details on this issue, is reported in the reply to General comments # 3.

Following the Reviewer's suggestion, the following sentence has been added in the revised Manuscript (Section 3.5.1):

"Results of fitting are in line with findings discussed by Mahowald et al. (2014). "

Finally the quality of figure 5 has been checked by the editorial office of Copernicus during the first submission process, and any problems have been evidenced on it; indeed, it was provided as .eps file.

p. 13370, values of r and σ: What are the uncertainties associated with these values? What are the units?

Uncertainties and units of r and σ values have been added in the revised Manuscript.

p. 13370 L. 12. how can you be sure that these dust measurements correspond to the optical properties of your samples? what are the associated uncertainties with the following estimates?

"The other microphysical property required for 6SV run is the refractive index. In Fig. 6 the real (n) and imaginary (k) part of the refractive index have been interpolated at the 6SV twenty wavelengths (350; 400; 412; 443; 470; 488; 515; 550; 590; 633; 670; 694; 760; 860; 1240; 1536; 1650; 1950; 2250; 3750 nm), following the spectral data of water-insoluble (Kokhanovsky, 2008; WCP-112, 1986) and calcite-rich dust (Ghosh, 1999) refractive index, respectively related to volcanics and travertine."

In the 6SV, the Mie theory is used to estimate optical properties of an aerosol type on the basis of its microphysical properties (i.e., size distribution and refractive index). The authors have yet discussed, in the previous replies, the need of adopting from literature values of the real and imaginary parts of the refractive index in the spectral range within which the 6SV performs the radiative transfer modelling. As experimental measurements of the optical properties of dust types of this study are not available, it is not possible to give an estimation of the uncertainties of the 6SV modelling results. It has to be also taken into account that the 6SV simply apply the Mie Theory with the required assumptions, particularly referring to the assumption of particle sphericity.

p. 13370 L. 26. these.

“this” should be used, in this case, as it is referred to “dust type”.

p. 13371 L. 1. rephrase

“The radiative modeling has been focused on the downward component of the radiative impact at BOA due to the volcanics and travertine dust in Rome area.” has been rephrased in *“The radiative modeling has been focused on the downward component of the radiative impact at BOA influenced by the volcanics or the travertine dust in Rome area.”*

p. 13371 L. 18. this is not what is shown in Fig.9. It shows that both give the same BOA irradiance. To say they don't affect direct radiation you would need to simulate the same atmosphere with any dust at all.

“Both volcanic and travertine dusts leave the direct component unchanged, while the diffuse component depends strongly on the mineral composition.” This sentence has been rephrased in *“Direct components calculated in presence of volcanic-only and of travertine-only dusts **shows negligible differences**, while the diffuse component depends strongly on the mineral composition.”* The authors have previously explained that the simulations have been performed by using each one of the two components, separately. The direct component of the BOA irradiance is the same whereas the diffuse component depends on the dust component used for the simulation as reported in Fig. 9.

p. 13372 L. 26. You could have concluded that without any RT simulation... Just the large difference in your ADHOC index of refraction for the two species were enough to justify it.

The following sentence has been deleted in the revised Manuscript: *“Nevertheless, the charge **(???)** of differences existing in the Rome local mineral dust composition on the variability of optical and radiative properties of the airborne aerosol appears as a key issue, to be further considered in the radiative balance analysis.”.* The real and imaginary parts of the complex refractive index can justify, but the RFE cannot be evaluated without a RT model. Furthermore, the RT models are a powerful and necessary tool recognized for accurate simulation of the radiative field and widely applied to the Earth Observation data.