Author Comment (Albert Ansmann on behalf of all the co-authors)

ACPD-2012-0355: Ansmann et al., Profiling of fine and coarse particle mass…..

First of all, we learned a lot from the three reviews, and considered most of the suggestions (including mentioned references). So, thank you ALL for taking the time to work on the manuscript and in this way to improve it.

All three reviews (plus our answers) are given in this document. Our answers start with: Our comment.

Reviewer #1

Specific comments:

1) Page 13365, lines 8 -12. It well known that Raman lidar technique provides important and useful data. At the same time, the claim that the Raman lidar technique provides "benchmark–like" data is greatly exaggerated. (Hereinafter, the quotation marks are used for the text from the discussion paper.) Unfortunately, the majority of the Raman-lidar community researchers, when they deal with aerosols, do not pay sufficient attention to the fact that the problem of numerical differentiation is known to be ill-posed in the sense that small perturbations in the function to be differentiated may lead to large errors in the computed derivative. As a consequence, one can found papers, published in highly ranked journals, where particle extinction profiles are oversmoothed. For example, in such a paper, one can see a well pronounced peak on a backscatter profile. The peak is due to an aerosol layer. But, the peak had disappeared on the corresponding extinction profile as if extinction and backscatter profiles vary independently and do not mainly follow variations of the aerosol concentration. Such oversmoothing can be avoided by the use of more sophisticated algorithms like proposed in the papers [1 – 3]. At the same time, the question whether the Raman lidar technique will be able to provide really benchmark–like data even with sophisticated algorithms needs further investigations.

Our comment: We changed the Introduction section and removed all these misleading and confusing statements. We now focus on the method presented only (combined polarization lidar plus sun-sky photometer), in a straightforward manner. We add the references 1-3 (see reviewer #1, references, two pages later in this reply) in the introduction where we now only briefly state that Raman lidar can provide backscatter and extinction profiles, without saying whether such observations deliver benchmark-like data or not.

Correct smoothing is always an issue. We may only reply: we are not just beginners, after 20 years of experimental experience with Raman data we believe that we know how to smooth the data and how to avoid oversmoothing, etc.

2) Page 13366, line 14. It is difficult to understand reasons why the proposed approach is "more general". The algorithm employs only a few parameters derived from the photometer observations (the aerosol optical thicknesses and the column-integrated volume concentrations of two modes). There were proposed much more sophisticated approaches (see, e.g., [4])

Our comment: We skipped …. More general…. We just state now that we extend the method to cover not only volcanic dust but also Saharan dust cases…..

3) Page 13371, lines 27 – 29. The relative uncertainties of the backscatter coefficients are done in the *papers Tesche et al. (2009b) and Ansmann et al. (2011b); the detailed error-discussion is done by Tesche in [5, Ch. 8.1]. The estimated uncertainties can be used for cases when the two modes have backscatter coefficients of the same order.*

The following example is done for the height of 2 km (see page 13400 Fig.2 of the paper under reviewing), i.e., for the case when the coarse mode largely dominates the fine mode. When the a priory depolarization-ratio of the coarse mode is taken to be of
0.35 (all other parameters being the same) the retrieved value of the fine-mode particle backscatter coefficient is three times higher; when that a priory parameter is taken to be of 0.29, the retrieved value of the fine-mode particle backscatter coefficient becomes negative. In other words, nonlinear properties of used equations can sometime lead to much higher errors than it follows from standard estimations.

Our comment: We improved the uncertainty discussion, along the suggestions made above. Errors are now discussed for each case separately in the Measurement section. We recalculated all results by varying the basic assumption of the depol ratio from 0.27-0.35 (desert dust), 0.30-0.38 (volcanic dust), and 0.00-0.04 (spherical particles) in order to determined the uncertainty range of all results. Based on these computations, the uncertainties in the coarse mode and fine mode backscatter coefficients are then quantified. These errors together with the other uncertainties (in the lidar ratio, vol-ext conversion factor, particle density) then allow us to realistically estimate the mass concentration uncertainties. Indeed, sometimes, the errors are very layer, but if we look at pronounced, well-detected layers of dust or of sulfate only, than the errors are of tolerable size and in the range suggested in the submitted version of the manuscript.

4) The depolarization ratios of dust and nondust modes are introduced as free parameters in the paper Tesche et al. (2009b) and [5]. The used values are justified by published experimental data. It is also worth noting that some constraints on variations of those parameters follow from measured profiles of backscatter coefficient and particle linear depolarization ratio.

Our comment: We enlarged the discussion on observations of desert dust depol ratios and volcanic dust depol ratios so much that we believe further statements are not necessary in this respect.

5) Page 13372, lines 5 -12. The overall relative uncertainties are underestimated. Please, use the standard "JCGM 100:2008" [6, Ch.5] to provide more reliable estimations on the base of the relative errors indicated on the lines 5 -12.

Our comment: As mentioned (point 3) we improved the uncertainty discussion significantly and discuss all uncertainties for each case separately based on extended error computations. This is sufficient, we think.

6) Eqs. (3) – (4) and Figures 2 – 5 (panels c, d, e). As a matter of fact, the extinctioncoefficient-and the mass-concentrations profiles are proportional to the corresponding backscatter-coefficient profiles at the wavelength of 532 nm. Thus, there is no much difference between the curves of the same color on the panels c, d, e. This leaves some doubts about the usefulness of the results knowing the large variability of aerosol integral-parameters as functions of microphysical characteristics. Moreover, the information from other lidar wavelengths is lost. When nonlinear algorithms are avoided, the method of linear estimation seems to be much more promising (see, e.g., [7] and references therein).

Our comment: First of all, reference 7 is now included in the paper. To point 6, I am personally not convinced that the more sophisticated multiwavelength lidar methods are better (more accurate) than the one proposed here. The problem with the shape model (spheroid model) in the case of 180 degree scattering is completely unsolved. The spheroid model is obviously well applicable in the case of sun photometer observation, but this is not the case for lidar inversion methods as e.g. the Gasteiger SAMUM paper in Tellus 63B (2011) indicates. I (AA) personally do not trust lidar inversion results too much when dust is present. There will be always large uncertainties when focusing on volcanic or desert dust by means of multiwavelength lidar inversion techniques. Nevertheless, the statement of the reviewer forced us to improve the introduction and to clearly formulate what the real goal of the method is: And this goal is (as stated now in the introduction) to present a simple, practical, and robust technique which is of advantage for networking and long term monitoring and when huge amounts of data are there for automated retrieval. If networking comes into play, then simple, robust methods are always better then complicated ones (that is at least my opinion, based on EARLINET experience). Furthermore, how many multiwavelength lidars do we have globally 10?, 20?, and how many of them are well calibrated 2? 4? And if they are well adjusted, how long does the good alignment hold (2 days, one week)? So, not easy to keep a high standard of observations and high quality of the results in case of sophisticated multiwavelength lidars (is at least our own EARLINET experience). Last point: If I look into the Veselovskii paper (AMT 2012, reference 7), then I can find a figure (I guess Fig4) with 6 profiles, all parallel to each other, 3 time backscatter, 2 times extinction, and one volume concentration profile. There is no difference to our figures!

References.

1 Shcherbakov V., “Regularized algorithm for Raman lidar data processing,” Appl. Opt. 46, 4879–4889, 2007.

2 Samoilova S. V. and Y. S. Balin, “Reconstruction of the aerosol optical parameters from the data of sensing with a multifrequency Raman lidar,” Appl. Opt. 47, 6816–6831, 2008.

3 Pornsawad P., G. D’Amico, Ch. Böckmann, A. Amodeo, and G. Pappalardo "Retrieval of aerosol extinction coefficient profiles from Raman lidar data by inversion method" Appl. Opt. 51, 2035-2044, 2012.

4 Chaikovsky A. , A. Bril, O. Dubovik, B. Holben, A. Thompson, Ph. Goloub, N. O’Neill, P. Sobolewski, J. Bösenberg, A. Ansmann, U. Wandinger, I. Mattis. "CIMEL and multiwavelength lidar measurements for troposphere aerosol altitude distributions investigation, long-range transfer monitoring and regional ecological problems solution: field validation of retrieval techniques” Optica Pura y Aplicada, Vol. 37, No. 3 -2004, 3241-3246, 2004.

5 Tesche M., “Vertical profiling of aerosol optical properties with multiwavelength aerosol lidar during the Saharan Mineral Dust Experiments” Universität Leipzig, Dissertation, 163 p., 2011. (see URL: http://www.qucosa.de/fileadmin/data/qucosa/documents/7125/doktorarbeit_published.pdf last access: June 2012)

6 JCGM 100:2008. Evaluation of measurement data -Guide to the expression of uncertainty in measurement. (see URL: http://www.bipm.org/utils/common/documents/jcgm/JCGM_100_2008_E.pdf last access: June 2012)

7 Veselovskii I., Dubovik, O., Kolgotin, A., Korenskiy, M., Whiteman, D. N., Allakhverdiev, K., and Huseyinoglu, F.: Linear estimation of particle bulk parameters from multi-wavelength lidar measurements, Atmos. Meas. Tech., 5, 1135-1145, doi:10.5194/amt-5-1135-2012, 2012.

Our comment: References 1,2,3,5,7 are now considered!

Reviewer #2

The authors' fundamental assumption throughout this study is that coarse particles fraction is associated with the fraction of light-depolarizing, non-spherical particles. The authors discuss the limitation of this assumption in the case of high water content of the coarse-mode and in the case of the coarse spherical maritime aerosol. However, the authors do not discuss the possible influence of coating of the coarse particles. This may have been relevant for example in the description of the West African case study of dust mixture with biomass-burning smoke. As Figure 3 shows, the low altitude transport of desert dust, which is typical for this winter case study, allows the smoke aerosol to lift through the dust layer and adhere to the dust particles which may then produce a coating effect. I assume this would comprise the limitation case for the methodology.

Our comment: The influence of size (sedimentation) and shape effects (coating) on the depolarization ratio of mineral dust is now discussed in the Method section. However, this coating issue remains to be investigated (in the field and in the lab). Until now there are no hints that coating leads to a decrease of the dust depol ratio. Our own observations during SAMUM (Jan-Feb 2008) and during the Eyjafjallajökull event (Leipzig, Cabauw, April-May 2010) with aged volcanic plume embedded in thick, optically dense sulfate aerosols do not indicate that there is a coating effect. If there would be a coating effect we should see it when we compare the lidar column backscatter values and related AOTs (for spherical and non spherical particles) and AERONET AOTs (fine mode, coarse mode). But we do not see it! We get good agreement between lidar and photometer results by assuming that there is no coating effect, no decrease of the depol value from 0.31 (Saharan dust) to may be 0.25, or from 0.34 (volcanic dust) to 0..25 or so. But as mentioned, the coating effect remains a topic, and must be investigated. As said, the coating effect is discussed in the revised version.

p. 13368, line 15: Difference between the depolarization ratios attributed to desert dust (0.31+-0.03) and volcanic dust (0.34+-0.03) seems to be not very strong, in particular when considering the uncertainties. As I understand these are averages+-StDev from a few studies that use quite the same technique. I would suggest consideration/discussion of possible values provided in other independent studies as well. For example, for the same volcanic aerosol event in 2010 there are at least two manuscripts in the JGR special issue that discuss the depolarization ratio,e.g., using CALIPSO [Chazette et al., 2012] or using photometer observations [Derimian et al., 2012]. By the way, agreements or discrepancies with the lidar ratio calculated using only the photometer observations for the same volcanic aerosol and for the key aerosol types, e.g. [Cattrall et al., 2005] could have been mentioned in the paragraph of line 18.

Our comment: We agree! We should provide more observational evidence that the desert dust lidar ratios are usually between 0.3 and 0.35 (and that 0.31 plusminus 0.04 is a good choice in our retrieval). This is now done in large detail. The same is done in the case of volcanic depol. values. They are between 0.34 and 0.38. Now the entire discussion is based on a larger number of desert dust publications as well as on recently published volcanic dust observations (Chazette, ACP, JGR, 2012, Winker, 2012, Miffre 2012, Gross 2012). Finally, we can conclude that 0.34 plusminus 0.04 is a good choice for volcanic dust.

We leave out to mention Derimian et al. (values are too low, as we already know from the Mueller and Gasteiger papers for SAMUM, Tellus and JGR). Photometer-based lidar ratios are widely model-based and thus uncertain because of the spheroid model problems in case of 180 degree scattering as mentioned in point 6 (reviewer #1). We also leave out to mention Cattrall for the same reason. We could also include the recently published Schuster et al. paper (ACP 2012). But it is not the aim of the paper to discuss the obvious shortcomings of photometer-based depolarization ratios and lidar ratios.

p.13382, line 27-29: it seems there is a typo, replace BC by CB.

Our comment: Yes! Is changed!

References Cattrall, C., Reagan, J., Thome, K. and Dubovik, O.: Variability of aerosol and spectral lidar and backscatter and extinction ratios of key aerosol types derived from selected Aerosol Robotic Network locations, Journal of Geophysical Research-Atmospheres, 110, D10, 2005.

Chazette, P., Bocquet, M., Royer, P., Winiarek, V., Raut, J. C., Labazuy, P., Gouhier, M., Lardier, M. and Cariou, J. P.: Eyjafjallajkull ash concentrations derived from both lidar and modeling, Journal of Geophysical Research-Atmospheres, 117, 2012

Derimian, Y., Dubovik, O., Tanre, D., Goloub, P., Lapyonok, T. and Mortier, A.: Optical properties and radiative forcing of the Eyjafjallajokull volcanic ash layer observed over Lille, France, in 2010, Journal of Geophysical Research-Atmospheres, 117, 2012

Our comment: As mentioned, we add Chazette ACP, 2012, Chazette JGR, 2012, and Winker, JGR 2012

Reviewer #3 (comments by G. P. Gobbi)

The manuscript addresses the retrieval of profiles of mass-specific extinction coefficients on the basis of colocated polarization-Raman Lidar profiles and sun-sky photometer inversions. The point of using sun-sky radiometers inversions to evaluate the extinction/volume relationships of fine and coarse aerosol modes is potentially good. Employing "pure" dust or "pure" ash depolarization as reference to separate fine and coarse aerosol components in the lidar trace may, however, bear some drawbacks. Here follow some relevant comments.

Our comment: We removed PURE from the entire text.

Page 13365 Line 15-21:
The sentence is misleading and should be reconsidered: Extinction to Backscatter ratios (LR) are always needed to evaluate aerosol backscatter coefficients in aerosol-laden atmospheres. Raman lidars retrieve LR by means of minor assumptions on spectral extinction, Mie lidars require a full assumption. This latter assumption can be assisted by further knowledge about aerosol type. In this respect, depolarization or color ratios may help such a choice. In fact, the solution of the Mie-Lidar equation is commonly obtained in terms of extinction coefficients (Klett, Appl. Opt., 20, p211, 1981; Fernald, Appl. Opt., 23, p652, 1984). Some comments about the Raman technique retrievals can also be found in the Referee Comment C3792.

Our comment: We changed the Introduction section and skipped this paragraph and changed others, with the goal to remove in this way all the confusing information. We now focus only on the method discussed in the paper (combined polarization lidar plus sun-sky photometer). We only add the advantage of Raman channels providing extinction coefficients, however without any comment, whether these are benchmark-like data or not…

Furthermore, model relationships linking Backscatter to: 1) Extinction, 2) Surface area and 3) Volume for various aerosol types (Marine, Dust, Continental) have been published in 2001 (Barnaba and Gobbi, JGR-D, p.3005, 2001; correction in JGR-D, 10.1029/2002JD002340, 2002). These relationships have been validated in 2003 (Gobbi et al., ACP, p.2161, 2003) by comparing polarization lidar and in situ aerosol measurements. The model computing such aerosol optical properties is the same one that generated the Extinction/Volume relationships of Barnaba and Gobbi (2004) cited here. This to say that mineral dust volume (or mass) could be estimated from Backscatter observations with acceptable accuracy (e.g. Gobbi et al., ACP, p. 2161, 2003) before the Barnaba and Gobbi (2004) paper and that the latter originated from the two previous ones.

Our comment: This is now considered in the Method section, as suggested here, almost word by word….

Page 13366
Line 2: The term "Sun-Sky Photometers" should be used instead of "Photometers".

Our comment: Done!

Page 13367
Line 12: Lidar depolarization measurements can be affected by a variety of uncertainties (bandpass-filter width, channels cross talk, calibration method, etc.). Lidar depolarization is, to some extent, "system dependent". To provide the reader with a reference about the quality of the measurement, the manuscript should report both the receiver band-pass filter width and the degree of depolarization the Lidar system measures in "pure" molecular conditions (i.e., where the signal could be calibrated). These issues should also be commented in the text.

Our comment: We now make a general statement that a well-calibrated polarization lidar is a basic prerequisite for the success of the approach and give proper reference to this topic (Freudenthaler et al., 2009), but we avoid to discuss this point in too much detail. We mention the Rayleigh calibration value (about 1%). But because we used several lidars, we do not mention any of the bandwidths of the receiver bandpass filters.

Page 13368
Line 15: Depolarization of "pure" mineral dust has been observed to show a certain variability. For instance, after laboratory depolarization measurements Sakai et al. (Appl.Opt, p. 4441, 2010) report "The values obtained from Asian and Saharan mineral particles were 0.39±0.04 (mean±standard deviation) for a high number of concentrations in the supermicrometer range and 0.17±0.03 to 0.14±0.03 in the submicrometer range". The first values are close to the ones (0.41±0.008) observed in Crete in 1999 and reported in a paper employing lidar depolarization to separate various aerosol types, including Saharan dust (Gobbi et al., Atmos. Env., p.5119, 2000). The lower depolarization values are closer to what is often measured after long-range transport. References in Sakai et al (2010) as well as in Miffre et al., (GRL, 2011) confirm such a variable behavior of depolarization. This to say that it is incorrect to expect and assume a universal "pure dust depolarization" level and that a depolarization

decrease in dust clouds (as well as in volcanic plumes) can be due to both decrease in effective size or mixing with non spherical aerosols. These points and references should be addressed in the paper.

Our comment: This is now discussed in extended detail, including the points mentioned above. It took me (AA) a week, to go through all relevant papers (about 40). A short review of this literature study is now given in the paper in the Method section. However, we avoid to cite and discuss the few papers as Gobbi et al. (2000) or Ansmann et al. (2009) with very large depol ratios of larger than 40% or even larger than 50%. It is not clear (at least from my point of view, AA) whether these values are trustworthy at all or not. So, to avoid a too long discussion on these extremely large depolarization values, we simply leave this point out. But it remains an interesting topic, these extremely large depol. values as they are predicted by the laboratory study of Sakai et al. (2010).

Page 13369
lines 22-24: In fact, Barnaba and Gobbi(2004) show the ratio Ext/Vol to vary mostly for dust-like particles. Conversely, continental type aerosols show quite a constant behavior of such ratio.

Our comment: Because we do not just see such a clear behaviour in Table 2, we better leave out to discuss this.

Line 25: Coarse mode Aeronet inversions are size limited (<15um). Could this introduce errors in the presence of large volcanic/dust particles?

Our comment: At the end of the Method section, we discuss the errors, and finally we mention that the presence of very large particles (in dust outbreaks, 24-36 hours after emission) may introduce large errors in the Vol values and thus errors larger than 100% in the retrieval results because of this possible 15 micron cut off problem.

Page 13371
Lines 24-25: The authors should address the fact that several Lidar systems are "blind" below a certain height (overlap issue). An important part of the optical depth (particularly in winter months) observed by the sun-sky photometer may then be not seen by the Lidar. Furthermore, cirrus clouds as well as aerosol inhomogeneity could also introduce biases in the sun-sky photometer inversions. To provide a "general approach" the authors should specify how to address these issues. Finally, the manuscript should indicate which Aeronet data (inversion method (1 or 2) and level (1, 1.5, 2)) is employed in each of the case studies addressed.

Our comment: After briefly discussing the pol lidar technique (calibration issue) in the Method section we add a few sentences regarding a potential overlap problem. G. P. Gobbi is right, this can be a big problem! However, in the text we make a more soft statement ….. that one has to take care, and usually errors may be of the order of 10%-20% in the AOT estimation by assuming certain extinction values in the blind near field area. This is at least found by Mattis et al. (2004), from multi-year EARLINET data analysis compared with AERONET data. Mattis et al. (2004) is cited now... But sure, we need 2 RFOV lidars to cover the entire troposphere. But this is not the main topic of the paper, so I tried to keep it short (and may a bit below the table).

Sections 3.1 and 3.2
The authors should address the issue of Aeronet data quality and time coincidence between Lidar and Sun-Sky photometer observations. For instance, very few Level 1.5 and 2 measurements were available

on 29MAY-08 in Leipzig, and these ended at 17 UTC (e.g., Aeronet site). Conversely, the reported lidar observations (Fig 2) span the 21.47-23.20 time range. How to address the relevant (4-6-hours) time variability of dust properties? A similar issue could be raised for the 22-JAN-08 Capo Verde Aeronet measurements which ended at 18.30 UTC while Lidar profiles start at 20 UTC (Fig 3).

Our comment: We improved the information content (give information concerning data used, level 1.0, level 2.0, in the Table 3 caption text). Regarding the dust on 29 May, there were very constant dust AOT conditions (28-30 May, 500nm AOT was always 0.7-0.8), we state that. Furthermore, there are only a few level 1.5 or 2.0 data for 29 May because of a thick cirrus throughout the day, only close to sunset the situation improved, as our lidar observations indicated. The statement that the AOT was constant over two days (28-30 May) should be enough to indicate that our approach to compare lidar observations (taken a few hours after the last AERONET obs.) with the AERONET data is justified. And we find the results that we expected, so this seems to be fine. For the 22 January case we explicitly state that obviously the smoke AOT changed from the AERONET to the lidar observation period according to the extinction observation with the Raman lidar. We discuss this and that should be sufficient.

Table 2: The values reported from Barnaba and Gobbi (2004) are wrong:
In fact, Fig.13 of that paper (reported below) shows that over the extinction coefficient range (5-300 Mm-1) kext varies from 3.56 to 3.44 m2 g-1 for continental aerosols (density=1.6 g cm-3) and from 2.7 to 0.46 m2 g-1 for Saharan dust (density=2.6 g cm-3). The maximum value of 1.35 reported in Table 2 for Saharan dust is found (in B&G2004) for Ext=20 Mm-1, conversely, the reported minimum value of 0.30 is never found. Fig 13 (B&G 2004) indicates dust kext can strongly vary as a function of Ext and that at the typical extinction levels encountered in Southern Europe (20-200 Mm-1, e.g. Fig 13b) kext span the range 1.5-0.54 m2 g-1 .
As specified before, continental aerosols show a very weak variation in kext (3.44 to 3.56 m2 g-1, Fig.13, B&G 2004) much weaker than the range (3-4 m2 g-1) reported in Table 2 of this manuscript.
Table 2 should be corrected according to the previous discussion.

Our comment: We improved the numbers accordingly! Thank’s!!