Dear AMT-Editor,

We thank the two reviewers for careful reading of the manuscript and the kind and constructive instructions how to improve the paper. We considered probably 95% of their suggestions. Our answers in bold.

We want to start with an overview of the main changes. In this way we already answer main questions of both reviewers #1 and #2.

1. **We do no longer argue (directly or indirectly) that the two-step POLIPHON method is better than the established one-step POLIPHON method. We now say that we have an extended POLIPHON method. The one-step and two-step method deliver a complementary set of solutions (height profiles of backscatter coefficients for three different aerosols). The one-step method needs less assumptions (and is thus a quality standard) whereas the two-step method provides more insight into the fine-mode and coarse-mode characteristics of observed dust (this is the step forward).**

2. **We introduce a new section (section 4) and provide a review of the depolarization ratio for all aerosol types (dust, soil dust, dry marine, urban haze, smoke, ….) and discuss in detail a potential interference of our dust separation and profiling technique by non-dust, but strongly depolarizing particle types such as pollen or dry sea salt particles.**

3. **We went carefully through the methodology section (section 5) and provide more explanations as requested and now clearly separate the one-step method (new section 5.1) from the two-step method (new section 5.2) development. We explain in detail our assumption on the used depolarization ratio for the residual aerosol depolarization ratio (i.e., the depolarization ratio of 0.12, we use,….for the total aerosol after removal of the coarse mode depolarization effect).**

4. **To better contrast the one-step method and the two-step method we improved Figure 5 which illustrates (sketch) the main steps of the one-step and the two-step method.**

5. **We provide a more extended uncertainty discussion (new section 5.4).**

6. **In the result section (section 6) we follow the main new strategy of argumentation, i.e., we added several paragraphs in which we compare the one-step and the two-step results and clearly give the impression now that they are complementary data set which can be used to estimate the overall uncertainties in the retrieval products.**

Anonymous Referee #1

General issues:

1) The authors should state more clearly the underlying assumptions that they use in the retrieval. This is especially true for the initial step, the separation of the three components of the backscatter coefficient. The assumptions made in this step may be invalid in many different atmospheric scenarios.

Regarding a clear statement of the assumptions…. this is done…, starting in the new section 4 and then, later on, within extended discussions when introducing the methodology in section 5, and in the new uncertainty section 5.4.

The assumptions on the residual aerosol (total – coarse dust) only hold for certain aerosol conditions, the reviewer argues…. That's it true. That is the same as with many other methods, e.g., in the Klett method, you need to know the aerosol type (conditions, location) you observed to know what lidar ratio you should use in the analysis (20, 50, or even 80sr). But it is not our topic to provide an automated code with look up tables etc…., we just introduce a new method.

Two distinct assumptions are made: a) When d_p > 0.12, the ratio of fine-dust and fine-non-dust backscatter coefficients if fixed. This is a strange assumption, as the

processes that generate these two types of aerosols are not related. It could be a reasonable assumption for situations of mainly background contribution to the fine mode.
However, is this still valid in cases of wildfires and urban pollution episodes?

We improved the text, we give more explanation. d_p=0.12 is to our opinion a very reasonable assumption which is height independent in cases with a homogeneous well-mixed aged aerosol plume. Now we better explain this topic (section 5.2). The lower boundary d_nd+df = 0.12 (input parameter for the residual aerosol consisting of non-dust and fine dust particles) represents a case with about 33% spherical non-dust particles (i.e., urban haze and biomass burning smoke in the case of free tropospheric aerosols) and 67% fine-mode dust. This is justified for our smoke-dust period we discuss in this paper. There is always a mix of dust and pollution as the AERONET Angstroem observations clearly indicate, at least for air masses from Turkey and the Middle East. We run simulations with d_nd+df = 0.08 (67% smoke and haze, 33% fine dust) and d_nd+df = 0.16 (100% fine dust). These simulations indicate that the uncertainty is of the order of 70-100% in the fine-mode dust profile, and less than 20% for the coarse dust profile in the case of pronounced dust layers. All this is now discussed in section 5.4. We did a lot of new simulations. But these error analysis figures will not be shown in the paper, we have already 18 figures, and the main message of the error analysis can be stated within a few sentences.

However, two error simulations (for the most important input parameters d_nd+df =0.08, 0.12, 0.16) and d_df = 0.1, 0.16, 0.22) are shown at the end of this reply letter.

Regarding … when is the method valid when not… we can only say: We always need backward trajectory analysis and AERONET observations. We need a careful pre-analysis of the polarization lidar data in the context of all these other information sources. This should be generally the case when doing atmospheric research. At the end, we need to have reasonable consistency between all avaialable data and informations. This is the case for all case studies in the result section 6.

b) When d_p < 0.12, there are no coarse-dust aerosols in the atmosphere. This may be applicable in more cases but still is it generally true?

We explained this issue just above…

Nevertheless, we believe we got the point of the reviewer. As a consequence, we changed the entire argumentation (throughout the paper): We state more carefully that the one-step method (which always gives a coarse mode backscatter coefficient! … when dust is there) and the new two-step approach are complementary. The profiles obtained with both methods show the full range of possible solutions for these two aerosol types (spherical particles, coarse dust) and thus provide a good basis to get a feeling for the uncertainty range of the two-step solution. Even for the fine mode dust, the one-step method (zero fine dust) and the two-step method (non-zero fine dust) deliver a range of solutions for the fine dust profile . With other words, the one-step method may be seen as a quality standard (needs less assumptions , but cannot resolve the fine mode dust backscatter) and from the point of view of an improved profiling … , the two-step method provides more insight in the vertical distribution of fine and coarse dust particles. All this is then checked with available AERONET column data. This is now our message, and discussed in section 6, and partly already in section 5.4.

The fine-dust aerosols in this case represent background contribution? Is it possible to have only fine mode aerosols in dust transport event?

Yes, fine dust can be an important background aerosol component, at least in aged biomass burning plumes. Biomass burning injects soil dust, and may contribute to a background aerosol with fine dust as the dominant dust component. This is described in more detail by Nisantzi et al. (ACPD, 2014) and supported by AERONET observations of high Angstrom exponents and simultaneous lidar observations with typical fine-mode depolarization ratios. All this is also supported by DLR Falcon in situ observations of size distributions of aged long-range transported smoke, where the coarse mode is no longer a dominant feature (Dahlkoetter et al., ACP, 2014, cited in our paper). So, yes, in biomass burning plumes the coarse dust

fraction is obviously almost deleted. The absence of the coarse mode is shown in Figures 9 and 10 in this paper (i.e., a missing coarse dust mode over Limassol on 26 and 27 Sep, biomass burning background aerosol in the free troposphere).

The assumptions and limitations are mentioned in various parts in the text but it will be beneficial for the reader to know clearly in which cases this algorithm can be applied as-is or when he needs to modify used assumptions. The method is not generally applicable, in contrast to what is claimed in the abstract and conclusions.

The method is generally applicable. Why should that not be the case? We should the methodology (itself) be restricted to specific aerosol scenarios? But, yes, the method needs assumptions, if that is meant. However, the method will work in all cases of atmospheric situations of cloudless and cloudy conditions. The method cannot be automated in that sense that you can do a blind data analysis, i.e., without checking the HYSPLIT analysis for example. In that point the reviewer is right. But this not the aim of the paper to present an automated version of a polarization lidar technique, as already stated above.

We will not repeat all statements (on assumptions) made above. We discuss all these points slowly and carefully now, in the way describe above (main changes). We introduced all influences, step by step, therefore we introduced the new section 4 and give extended discussions in section 5 (methodology). As a main conclusion, we use the one-step and two-step solutions as a whole to check the quality of the results (uncertainties) and as strong point, we compare this AERONET observations. We need HYSPLIT and AERONET data to get a clear picture of the atmospheric situation (even MODIS imagery is used). This is always our basic approach when doing atmospheric research based on observations. This is demonstrated based on the case studies in section 6.

A final remark: The method is simple and straightforward, much more simple and straightforward than these black-box LIRIC and GARRLIC approaches. The authors are convinced that this method is clearly better than any of the LIRIC or GARRLIC approaches. Until now, we have never got a clear pictures of the error ranges of the LIRIC and GARRLIC products.

2) The authors should briefly discuss their definition of fine and coarse aerosols. Are the definitions of these modes used in the cited works of e.g. Sakai, Barnaba, O'Neill, and Dubovik compatible? Could these different definitions contribute to the total uncertainty estimation, and how much?

We give the definition (fine mode, coarse mode) now already in the introduction and discuss this aspect in the AERONET observation section (section 3). The main optical contribution to the fine mode optical properties (after AERONET) are due to particles with radii less than 300nm (O'Neill et al., 2003). This is also stated now. We will not confuse the people with different numbers (defining the particle size spectrum), e.g., when using the Dubovik approach (here the fine mode ends in the size spectrum, where the minimum is found in the bimodal size distribution, between the two modes). We do not use the optical properties computed by the Dubovik approach, O'Neill is better in this respect, the more direct approach. Barnaba and Gobbi rely on AERONET size distributions and are in an overall good agreement with all the different AERONET products (O'Neill, Dubovik). The overall impact of different assumptions on the fine mode size distribution range is of minor influence. The main optical information comes from the numerous particles within the radius range from 50 to 250nm. We will not confuse the reader with too much (and different) information here.

We also discuss briefly (or indirectly) the approach by Sakai (section 4). Sakai just tried to filter out all supermicron particles to get the depolarization ratio for the rest. But these depolarization ratios are in good agreement with the Gasteiger simulations (mentioned in section 4), and with the overall depolarization observations (SAMUM, Asia, etc.). More cannot be done yet, is our feeling. Of course we need more research in this field. This is also clearly stated several times, including in the conclusions (section 7).

And all in all, the error analysis with different fine-mode dust depolarization ratio d_df = 0.10, 0.16 (used in the two-step approach) and 0.22 (section 5.4), which may result from different fine-mode particle size distributions, does not introduce so large errors. The errors are of the order of 20%-30% in the fine dust backscatter coefficients and less than 10% in the coarse dust backscatter (see the second figure at the end of the reply letter). As mentioned, more important (more sensitive impact) is uncertainty in the assumption of the residual aerosol depol ratio d_nd+df=0.12. The uncertainties are stated in section 5.4.

Specific comments:
p. 5176 l.6 What do you mean by **continental** dust particles and how can AERONET identify their presence?

Is changed (statement is removed), we always use HYSPLIT trajectories to check the air mass type (marine, continental and so on…) and to interpret our observations (lidar, photometer).

p. 5176 l.17 Why do you use 16% for fine dust instead of 15% mentioned in line 10?

This is just the average of the measurements of Sakai et al., we mention that in section 4 now. 16% or 15% would introduce only minor changes in the products, as just mentioned above (uncertainty discussion).

p. 5177 ll. 8-9 Mention that LIRIC can retrieve 3 different aerosol components, "fine-mode, coarse-mode spherical, and coarse-mode non-spherical particles."

We do it now in the introduction.

p. 5177 ll.17 -25 For clarity, consider separating the description of the one-step POLIPHON from the new developments (esp. ll. 19-20).

We keep the description short in the introduction (is already very long), but better separate the description of the two different methods in the method section (section 5.1, one-step method, section 5.2, two-step method).

p. 5178 l.9 Consider rephrasing.

Done.

p. 5178 l.18 *were* checked...

Done

p. 5179 ll. 19-20. The sphericity parameter etc are not calculated from the AOT but from the sky radiance measurements. You should also mention here the definition of FVF that you use in p.5180 l.11.

Is now improved accordingly in section 2.2

p. 5180 l. 17 Is the volume distribution retrieval of AERONET (and, consequently, the fine-mode volume fraction) reliable near the dust source? (see e.g. D. Muller et al., Mineral dust observed with AERONET Sun photometer, Raman lidar, and in situ instruments during SAMUM 2006: Shape-independent particle properties, JGR, 2010)

Is now also mentioned now accordingly in section 2.2. New Mueller reference is included.

p. 5180 l. 29 Clarify if AOTf/c are calculated using O'Neil or Dubovik method.

O'Neill method is now indicated by reference (section2.2).

p. 5186 ll. 12-14 Are these typical values actually used? If not, I don't think you need to mention them again.

We included the parameters now in Table 1 (as suggested by the reviewer, see below) and therefore kept the discussion of the parameters very short now at the end of section 5.2.

p. 5187 l. 7. The PBL height mentioned here is not consistent with the values of p. 5189 l. 10.

Is improved now

p. 5187 ll. 6-9. Clarify which method you use to separate the PBL and FT lidar ratio from AERONET measurements.

We give the reference of Mamouri et al. (2013). We do not like to discuss it here, paper is already long.

p. 5188 l. 16. I don't think it's possible to argue that the "uncertainties are usually much lower" based on a single case study.

Although this is the case if you look at all the comparisons with AERONET, we skipped this not very science-based statement.

p. 5194 s.5.4 What are the v/AOT values actually used for converting extinction to volume concentration? Consider adding these values also in Table 1.

Are now given in Table 1.

p. 5204. Fig 1. Consider marking the position of the measurement site.

Done (Figs. 1 and 6, yellow circle)

p. 5208 Fig.5 Verify that the numbers in the two-step diagram are correct. In the second step, pfmax = 0.12, so values above 0.16 should not be possible.

Figure 5 is largely improved. We think all inaccuracies are gone now.

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Anonymous Referee #2

…….. The technique, as described and applied, is only applicable to specific scenes, however, as a result. That is, specific assumptions used for depolarization ratios for non-dust fine-mode particles, fine-mode dust and coarsemode dust, would break down in the presence of other depolarization particles (pollens, ocean biogenics, aged smoke, etc...).

See our list of main changes, at the beginning of this reply letter. Sorry, we disagree! The technique is generally applicable. This tells us the long-term TROPOS experience (LACE98 in central Europe, SAMUM1, Morocco, SAMUM 2, Cape Verde, SALTRACE, Barbados, INDOEX, Maldives). In the new section 4, we discuss a potential interference or bias by, e.g., pollen or dry sea salt particles. We conclude, that there is practically no interference. The only significant depolarizing aerosol type is dust (volcanic dust, soil dust, desert dust) and nothing else. There may be a few exceptions, may be in the boundary layer at certain days in spring (pollen, see discussion in section 4).

I do have issues with the mostly "hand waving" arguments around uncertainties. Its seems to me that it would not be too difficult to do the math and solve an error model based on input uncertainties that propagate all the way through the solution. I recognize that the Authors are using AERONET to verify results, and that's okay. But, this portion of the manuscript is a bit wanting, and something that the lead should simply break down and try to formulate sometime. Its not a make-or-break request, however. As with Reviewer #2, I do think you should specifically define ranges for fine and coarse mode somewhere in the Introduction.

See the new section 5.4. Here we now provide an extended discussion on uncertainties. However, the application of the error propagation law and a respective error model does not help here. Simulations (varying the input parameters, one by one…) is the only useful technique to check the uncertainties. We did this and present the main results (resulting error ranges) of these simulations. Two of the error simulations are shown in this reply letter, see the figures below, at the end this letter.

Fine-mode and coarse-mode definition is already given in the introduction now, and discussed later on in section 3.

Also, with respect to some of the information used from AERONET inversion retrievals (mass volume concentrations), I wonder what the impact is on your verification considering that these retrievals are usually only conducted for relatively high optical depth loading cases (tau500 > 0.4). Its a minor point, however, given that this mostly a secondary verification parameter.

First of all, the O'Neill approach (FMF, fine-mode and coarse-mode AOT) is applicable even to marine particles with AOTs of 0.03 to 0.06. The Dubovik inversion method may be only applicable to larger AOTs (with low uncertainties). But the method provides also reasonable results for lower AOTs. This is just our experience when looking at all the combined lidar/photometer observations during the last 15 years.

Otherwise, I'm uploading my reviewer notes, which contain what is mostly minutiae and technical notes if anything. Lots of places to improve the narrative are highlighted. Sorry for the relatively small notation. I wish Copernicus would give us proper manuscripts to edit!

THANK YOU VERY MUCH! This was a very good support for us. We considered practically all statements. But some (may be two or three) of them we were not able to read. But these were of minor impact, we believe…

One final note, however, that I'd like the authors to consider. Typically, use of AERONET data for publication implies that you've talked with the site PIs before you use their data. In particular, Barbados is Joe Prospero's site. These guys like to be contacted ahead of time, and, if not at least acknowledged, offered full co-authorship. You folks would be wise to get in touch with these PIs and let them know that this exists. These data can't be collected if the PIs can't advise their sponsors that the data are being put to good use!

Yes, now we thank Joe Prospero for the Ragged Point measurements. We will send him the paper, when it is accepted. We know him personally. Albert Ansmann met him last time at the DUST 2014 conference in Italy in June 2014. Rodanthi Mamouri met Brent Holben (head of AERONET) at the last ACTRIS meeting in France and then she went to Barbados (SALTRARE) and changed the measurement configuration of the Ragged Point AERONET photometer in close cooperation with Brent Holben and Joe Prospero's group. So, there exist close links already.

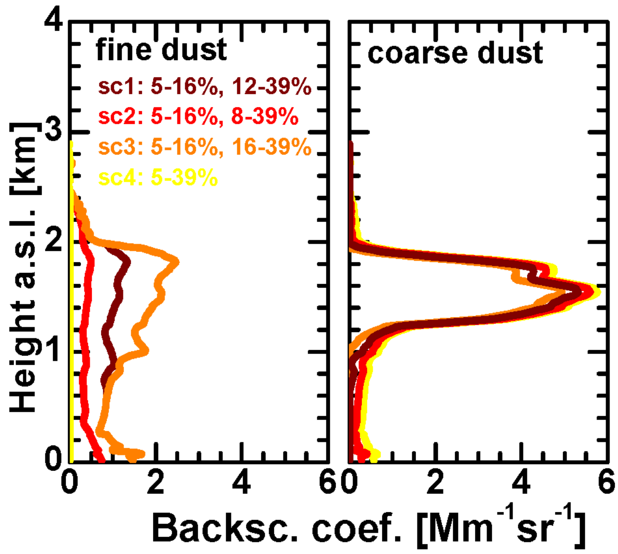

Figure 1: Here we simulated different residual aerosol (total – coarse dust) depol ratios: d_nd+df =0.08, 0.12, 0.16

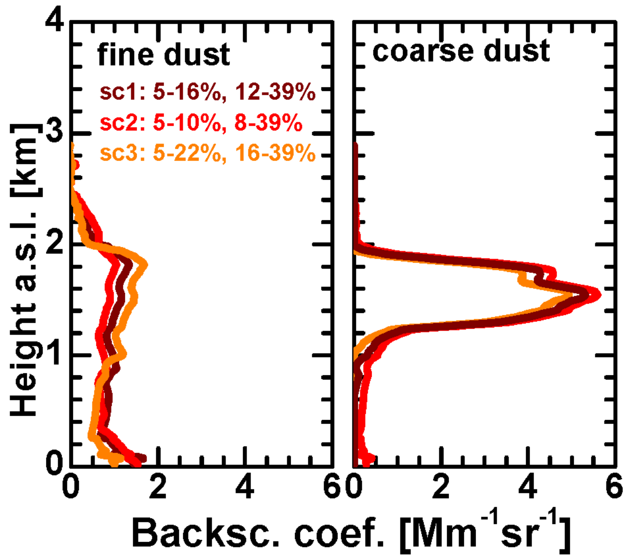

Figure 2: Here we simulated different fine-dust depol ratios: d_df = 0.1, 0.16, 0.22, which then also influences the assumption on d_nd+df (0.08, 0.12, 0.16).