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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "A Backscatter Lidar Forward Operator for Particle-Representing Atmospheric Chemistry Models" *by* Armin Geisinger et al.

Anonymous Referee #2

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General remarks:

The paper deals with an attractive and important research topic. The development of a backscatter lidar forward operator is very useful to support aerosol transport modeling and efforts to forecast aerosol conditions.

However, the paper is a pure (100%) technical, methodological paper and thus appropriate for AMT. I therefore recommend to move it to AMT in case that it gets finally accepted.

Unfortunately (I must say), the paper is not acceptable in its present form. But I have hope that it can survive. I was expecting a straight forward paper, a clear, compact

description of the nice method and then also a nice and easy to follow (and convincing) case study (demonstration case) which clearly corroborates the usefulness of the methodologic concept and developments. But all this is not presented! The method is introduced in a rather lengthy way, text-book knowledge is outlined in extended detail. This can easily be avoided. But! .... The demonstration case (Eyjafjalla volcanic aerosol scenario) is simply the worst case they could select! This case must be substituted by a case for which Mie scattering can be applied.

The methodology is based on the fundamental assumption that the aerosol particles can be handled as spheres (as so called Mie particles such as marine aerosol particles or urban haze or biomass burning smoke particles). This is well justified in the case of anthropogenic pollution including biomass burning smoke. However, the demonstration case, the authors selected, deals with volcanic dust particles crossing Germany quite shortly after emission so that nothing is well known, size distribution, chemical composition, shape properties. Such a situation occuring on 16-17 April 2010 has never been observed before with advanced atmospheric technology. Fortunately, several EARLINET lidars were able to pick up even the first traces of the heavy dust front which crossed Germany on 16 April 2010. The measured lidar ratios indicated irregularly shaped volcanic dust particles. The AERONET observations revealed that these fresh plumes were coarse mode dominated.... So it was by far not a normal aerosol situation. It was an extreme situation! As mentioned, nothing is well known, the size distribution of the volcanic dust particles is not known, the shape properties are not well known. Even the chemical composition could not be well characterized. Aircraft observations were started several days later, when the aerosol mixture and physico-chemical properties already had changed significantly, and spherical sulfate particles had formed from the emitted SO2, and were mixed with the voclanic ash and dust. On 16 Apri 2010, it is most likely that only dry ash and dust particles were present. And for such a case, Mie-scattering-based methods (including this backscatter lidar forward operator method) can definitely be not applied, as it is shown in this paper!

The authors are forced to discuss this case in a very speculative way so that the main question at the end was not: What do we learn from this paper? No, the main question was: Why did they choose this incredibly complicated case so that it was, at the end, impossible to demonstrate the usefulness of the new technique?

So, my conclusion is not only,. . .. the paper needs major revision. The conclusion is, we need an easy case that shows the applicability of the forward operator model! This paper definitely fails to show that. Without such a simple case, the paper must be rejected! To be fair, many papers of this quality get rejected at this stage, already. However, I believe that there is enough and substantial material to save it! But only if we have another, a much better, much easier demonstration case.

The conclusions based on this volcanic case the authors selected will always remain rather uncertain and thus speculative because of lack of sufficient knowledge of the relationships between the optical properties and the size/shape/composition properties of Islandic volcanic ash 2 two days after emission. There is no hope!

The paper has many other weak points. It is not well written. The reference list is far away from reflecting the aerosol lidar science field in a proper way. Even in the case of the volcanic aerosol plumes the authors leave out to mention essential papers. All this must be improved.

Point by point....:

Abstract:

The abstract must be rewritten: please provide a compact text with the goal of the article, the methods used, and the essential finds. That's it! All the motivating and explaining statements should not be given in an abstract. The right place for such information is the Introduction (section 1).

Section 1 Introduction

P 2-4: The introduction can be kept much shorter and more compact. Many details are

given that have nothing to do with the goal of the paper.

P2, l30: Please check Cuevas, MPL observations and lidar data assimilations into models (see my reference list below).

P3, l1-15: please remove the first paragraph, please focus on aerosol lidar only. The work of Japanese groups to provide lidar data for the modeling community should be mentioned in this context. They were pioneering in the 1990s and are even active presently to combine lidar and atmospheric aerosol modeling. There are also papers in which lidar data are used to validate models and verify results from the EARLINET community (check the speciall issue of EARLINET in AMT, and the papers including there reference lists). There were also efforts to combine lidar and model results in the Tellus SAMUM special issues of SAMUM (Tellus 2009, 2011). In the case of multiwavelength lidar (here the authors probably mean inversion techniques), I was surprised that the authors left out to provide the references to the fundamental Mueller, Veselovsky and Boeckmann papers! These authors are active in this field since more than 15 years. Mueller (JGR 2010, 2012) published several papers on the effects in optical closure studies when the scatterers are non spherical. . .and , if remember right, even not of spheroidal nature.

P3: Please add somewhere the basic work of Heese et al and Wiegner and Geiss in AMT on the characterization of ACLs (see my list below). All in all I was not very happy with the introduction section. It gave me the impression that the authors were not willing or not able to provide a well-balanced literature overview on aerosol lidar. This needs to be significantly improved and will certainly strengthen the quality of the paper as a whole. A good literature review always provides the impression, the work is done in a professional way. The other way around, ... provides the opposite feeling.

Here are some papers, that should be cited. . .

Heese, B., Flentje, H., Althausen, D., Ansmann, A., and Frey, S.: Ceilometer lidar comparison: backscatter coefficient retrieval and signal-to-noise ratio determination,

Atmos. Meas. Tech., 3, 1763-1770, doi:10.5194/amt-3-1763-2010, 2010.

Wiegner, M. and Geiß, A.: Aerosol profiling with the Jenoptik ceilometer CHM15kx, Atmos. Meas. Tech., 5, 1953-1964, doi:10.5194/amt-5-1953-2012, 2012.

An introduction of a paper dealing with a ceilometer forward operator should include such 'fundamental' papers to my opinion, especially devoted to Jenoptics ceilometers.

Here, some further papers (and one PhD work) on lidar data assimilation, and the references in these articles may help. . .. Please have a look and check!

http://www.mri-jma.go.jp/Dep/ap/ap1lab/member/tsekiyam/files/sekiyama_thesis.pdf

Wang, Y., Sartelet, K. N., Bocquet, M., Chazette, P., Sicard, M., D'Amico, G., Léon, J. F., Alados-Arboledas, L., Amodeo, A., Augustin, P., Bach, J., Belegante, L., Binietoglou, I., Bush, X., Comerón, A., Delbarre, H., García-Vízcaino, D., Guerrero-Rascado, J. L., Hervo, M., Iarlori, M., Kokkalis, P., Lange, D., Molero, F., Montoux, N., Muñoz, A., Muñoz, C., Nicolae, D., Papayannis, A., Pappalardo, G., Preissler, J., Rizi, V., Rocadenbosch, F., Sellegri, K., Wagner, F., and Dulac, F.: Assimilation of lidar signals: application to aerosol forecasting in the western Mediterranean basin, Atmos. Chem. Phys., 14, 12031-12053, doi:10.5194/acp-14-12031-2014, 2014.

Janiskova et al., Assimilation of cloud information from space-borne radar and lidar: experimental study using a 1D+4D-Var technique, QUARTERLY JOURNAL OF THE ROYAL METEOROLOGICAL SOCIETY Volume: 141 Issue: 692 Pages: 2708-2725 Part: A Published: OCT 2015

Campbell et al., CALIOP Aerosol Subset Processing for Global Aerosol Transport Model Data Assimilation, IEEE JOURNAL OF SELECTED TOPICS IN APPLIED EARTH OBSERVATIONS AND REMOTE SENSING Volume:3 Issue: 2 Pages: 203-214 Published: JUN 2010

Cuevas, E., Camino, C., Benedetti, A., Basart, S., Terradellas, E., Baldasano, J. M., Morcrette, J. J., Marticorena, B., Goloub, P., Mortier, A., Berjón, A., Hernández, Y.,

Gil-Ojeda, M., and Schulz, M.: The MACC-II 2007–2008 reanalysis: atmospheric dust evaluation and characterization over northern Africa and the Middle East, Atmos. Chem. Phys., 15, 3991-4024, doi:10.5194/acp-15-3991-2015, 2015.

I also found recently somewhere (unfortunately I do not remember the web page) a workshop report on MPL lidar data assimilation into the NMMB model. The first author was again: Cuevas. So there are presently data assimilation efforts in the dust forecast scene.

P4, l8: I sounds attractive and convincing when essential input parameters have not to be assumed. But in the case of the lidar ratio, it is frequently even better to calculate the robust extinction properties and then take a lidar ratio of 50 sr to obtain the backscatter coefficient. I would like to see if this option is mentioned too. For example, for volcanic dust (your case study) it is shown that the lidar ratio is around 50sr (Gross et al. 2012), and it would be simply much more straight forward to use such a number of 50sr than your rather unrealistic values close to 'modeled' 5-15 sr. I will come to this point later again.

To my opinion, one of the best Eyjafjalla volcanic lidar ratio papers is (please add it to the references and include the findings in the discussion) :

Groß, Silke und Freudenthaler, Volker und Wiegner, Matthias und Gasteiger, Josef und Geiß, Alexander und Schnell, Franziska (2012) Dual-wavelength linear depolarization ratio of volcanic aerosols: Lidar measurements of the Eyjafjallajökull plume over Maisach, Germany. Atmospheric Environment, 48, Seiten 85-96. DOI: 10.1016/j.atmosenv.2011.06.017.

To my understanding, the Ansmann 2010 lidar ratios of 50-60sr (similar to the ones in Gross et al, for the lofted isolated volcanic ash layer. . .) probably correctly describe the dry volcanic aerosol lidar ratios in the beginning of the volcanic episode. Later (18-19 April and afterwards. . .) the pure volcanic dust and ash conditions were gone. Volcanic sulfate aerosols formed from the emitted volcanic SO_2 so that a mixture of spherical and irregular shaped particles were always present. I mention it only to avoid that the authors misinterpret the findings in the Gross paper. The lidar ratios were definitely in the range of 50-60sr for dry volcanic dust (in the absence of volcanic sulfate). All this is very similar to desert dust, and the desert dust lidar ratio at 1064nm seems to be equla to the lidar ratios for 532 nm or even slightly higher than the ones for 532nm, gained from 1064nm lidar/ 1020nm photometer observations.

The authors mention the Greek paper on volcanic dust of Kokkalis et al., but if they would read Papayannis et al., (Atmos Env. or Atmos Res., 2012?) they would see that it was rather complicated to identify the volcanic aerosols in the mixtures of urban haze, marine particles, Saharan dust etc over the Eastern Mediterranean„ far away from Iceland, so that information of these articles (even Mona et al., 2012) should be handled with caution.

Section 2 Methods

Section 2.1 can be shortened drastically, the lidar equation is given in so many text books, so many details are not needed. Focus on the parameters needed for Eq.2.

P4, l30: Lidars are never elastic (hopefully…), so please write: elastic backscatter lidar Also sections 2.2.1 and 2.2.2 contain just text book knowledge. Please keep the discussion as short as possible, use references.

P7,l7: Buchholtz's formula holds for 200 to 1000nm, but you use 1064nm (please mention the used wavelength much earlier in the paper, for example already in this section!)? Did you extrapolate? Or ignore it? Please state the wavelength immediately, and give all the Rayleigh parameters for this wavelength, including the lidar ratio…

Section 2.2.3, even here, please provide the wavelength of the ceilometer, and then discuss all the different size scenarios for this wavelength, give examples for which the Mie theory may holds, for what size range (Aitken mode, accumulation mode, etc…), the same for DDA, T-matrix etc. Which of the natural particles have almost spheroidal

shape, which look like cyclinders? Make the full discussion more realistic, more lively.

P7,l20: So you definitely use Mie scattering codes for your backscatter lidar forward operator! This is ok, and the best first step you can choose!

BUT!!! … as a consequence, please provide us with an application (demonstration case) for which Mie computation really holds (fine-mode aerosol, urban haze and smoke) . This is a mandatory condition for the final acceptance of this paper from my side!!!

Again, in conclsuion: Sections 2.2.3 and 2.2.4 are rather lengthy, not necessary, text book knowledge, sections should be as short as possible.

Section 3 Case Study

I could simply leave out the rest because I am not willing to accept a revised version with such a complicated case. But again, many figures could be left out because they are already presented in the literature, e.g., that the extinction coefficient is robust with respect to shape effects (see for example Dubovik JGR, 2006, but this is shown in many other papers too). So, one could easily skip Figs.8, 10, 12, 14.

Here is the Dubovik reference:

Dubovik, O., Sinyuk, A., Lapyonok, T., Holben, B., Mishchenko, M., Yang, P., Eck, T., Volten, H., Munoz, O., Veihelmann, B., van der Zande, W. J., Leon, J. F., Sorokin, M., and Slutsker, I.: Application of spheroid models to account for aerosol particle non-sphericity in remote sensing of desert dust, J. Geophys. Res., 111, D11208, doi:10.1029/2005JD006619, 2006.

The paper has 19 Figures, and 50% are just required to explain the complexity of this crucial volcanic ash case and the impact of size, shape and composition of volcanic particles on the related optical properties. Alone this fact corroborates that we need a better demonstration case. The main goal of the paper is: presentation of the backscatter lidar forward application plus demonstration case. And without presentation of a convincing demonstration case, the paper must be rejected because the main message and conclusion then becomes completely unclear.

P10, l23: One of the best lidar-ratio papers in this field is not mentioned in the article: Groß, Silke und Freudenthaler, Volker und Wiegner, Matthias und Gasteiger, Josef und Geiß, Alexander und Schnell, Franziska (2012) Dual-wavelength linear depolarization ratio of volcanic aerosols: Lidar measurements of the Eyjafjallajökull plume over Maisach, Germany. Atmospheric Environment, 48, Seiten 85-96. DOI: 10.1016/j.atmosenv.2011.06.017.

One needs the depolarization ratio to see in which of the layers pure or almost pure volcanic dust was observed. In the lowest layers a mixture of local pollution, sulfate particle from the volcanic emission of SO2 and volcanic ash prevailed. Nevertheless, the lidar ratio for the volcanic ash was always close to 50 sr, and in the mixtures even larger, probably because of the freshly formed quite small sulfate particles. None of the publications on the Eyjafjalla eruptions, showed lidar ratios below 40sr in the volcanic layers. And this must be the guide for all your modeling studies which to my opinion are at all rather speculative. Fact is: The reality did not provide volcanic lidar ratios of 10-20sr.

Add to the references. Pappalardo, G., Mona, L., D'Amico, G., Wandinger, U., Adam, M., Amodeo, A., Ansmann, A., Apituley, A., Alados Arboledas, L., Balis, D., Boselli, A., Bravo-Aranda, J. A., Chaikovsky, A., Comeron, A., Cuesta, J., De Tomasi, F., Freudenthaler, V., Gausa, M., Giannakaki, E., Giehl, H., Giunta, A., Grigorov, I., Groß, S., Haeffelin, M., Hiebsch, A., Iarlori, M., Lange, D., Linné, H., Madonna, F., Mattis, I., Mamouri, R.-E., McAuliffe, M. A. P., Mitev, V., Molero, F., Navas-Guzman, F., Nicolae, D., Papayannis, A., Perrone, M. R., Pietras, C., Pietruczuk, A., Pisani, G., Preißler, J., Pujadas, M., Rizi, V., Ruth, A. A., Schmidt, J., Schnell, F., Seifert, P., Serikov, I., Sicard, M., Simeonov, V., Spinelli, N., Stebel, K., Tesche, M., Trickl, T., Wang, X., Wagner, F., Wiegner, M., and Wilson, K. M.: Four-dimensional distribution of the 2010 Eyjafjallajökull volcanic cloud over Europe observed by EARLINET, Atmos. Chem. Phys., 13,

4429-4450, doi:10.5194/acp-13-4429-2013, 2013.

See also. . ..

Wiegner, M., Gasteiger, J., Groß, S., Schnell, F., Freudenthaler, V., Forkel, R., 2011. Characterization of the Eyjafjallajkull ash-plume:potential of lidar remote sensing. Physics and Chemistry of the Earth. doi:10.1016/j.pce.2011.01.006.

Section 3.2

I miss a Table with technical details of the ceilometer, what, for example, is the pulse rep rate? And what does it mean. . .. Assuming that the pulse energy of 8 micro Joule given by Flentje is true. Do you know the technical details or not? Are you sure that the overlap is completed in 1500m height? Do you know it? Yes or no? A calibration measurement of the ceilometer was not provided. . .. . We take one from CALIPSO for 1064 nm. All this is really not easy to accept and tolerate by a reviewer trying to teach his student day by day to do always careful calibrations and high quality observations.

Section 3.3.

P12, l9: COSMO-ART uses six size classes, and class 1 includes all particles with diameters up to 2 micrometers, so all Aitken and accumulation mode particles and the rest of all fine mode particles (up to a diameter of 1 micrometer), and then also a large fraction of the coarse mode (particles with diameters from1 to 2 micrometers) are altogether in just one single class. Is that justified from the optics point of view? I do not believe. . .

P12, l22: molecular number density of standard air was calculated. What do you mean? Do you use, for simplicity, standard atmospheric conditions? Is that justified? Schumann paper is referenced asd ACPD version, this in not acceptable, you need to take the ACP version. . . What size distributions did Schuman et al measure. . .? Should be used as a guide in all the modeling approaches.

Section 4 is really not needed if you take a simple demonstration case. . . I will not give comments, except one: You cannot provide any uncertainty estimation (e.g. Fig 15) as long as you do not know what the truth is. We know already that neither the spherical nor the spheroidal particles shape models works in the case of irregularly shape dust particles (Gasteiger, 2011, Kemppinene, 2015).

Section 5 Results

The Schumann et al size distributions may show approximately the size distribution characteristics. Your size distribution cases 1 and 2 are just speculations. Case 1 (this COSMO modeled size distribution) appears to some extend realistic. Table 3 contains the modeled lidar ratio, and for the main classes 1,2 and 3 (case 1) the lidar ratios are 13, 4.6 and 10 sr, and on average probably close to 10sr and therefore far far away from the real-world values of 50-60sr.

As mentioned be careful when using Mediterranean Eyjafjalla dust papers, the dust maybe already widely mixed with other aerosols. And the paper of Mortier provides some column integrated lidar ratios and not values for well defined isolated lofted dust layers.

All in all, section 5.2 is so confusing and speculative, and simply ignores literature findings. There is also attempt to check the literature for observational hints that would corroborate all there speculative arguments. . . This part of the paper is clearly inacceptable.

Section 5.3 Qualitative Comparison

. . .provides no concluding remarks and any take-home messages (zero!) Again, and very clear: The shown demonstration case is a disaster for entire article and must be removed.

Section 5.4 Quantitative Comparison

Because of lack of any harmony between the model results and ceilometer observations (a factor of 60 difference!), the authors start to play around with the output (attenu-

ated backscatter values) of the forward operator and reduced the output by a factor 10, 20, 30 etc. Why do you not check other observations (EARLINET lidar observations, even the CALIPSO overflight is available) regarding the truth! So discussion remained open. . . at the end.

Figure 4 shows particles in diameters, Figs 8,9, etc show particles in radius, Figure 16 shows particle size. . ...., please harmonize!

Figure 17 shows COSMO ART results ..with shape sensitive lidar ratios at all around 10sr. All colors are just grey. Nevertheless this figure corroborates best that this demonstration case is a disaster. Measured ‘real world’ Eyjafjalla lidar ratios are a factor of 4 higher. The discrepancy is to my opinion just caused by the shape of the volcanic particles.

Figures 18 and 19 present almost the opposite of what I would like to see, and what would be possible to show if a simple European pollution day would be selected. It is hard to believe that the ceilometer output (top panels) have something to do with the COSMO ART output (bottom panel).

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