Review of the paper entitled

A new study of sea spray optical properties from multi-sensor spaceborne observations

by K. W. Dawson, N. Meskhidze, D. Josset, and S. Gassó

I would like to thank the editor for the opportunity to comment on the revised version of the manuscript. As most of the points in my short comment have not been addressed in the revised version, I will repeat them below. Answers by the authors are given in *italics*. My replies as well as updated and new comments are given in **bold**.

If I had been a reviewer from the start, I would recommend rejection of the paper but also encourage re-submission after thoroughly addressing the points below. As I entered the review process during its second round, I think it's fair to leave it to major revisions. I have some issues with the paper that I think should be addressed in a revised version (details follow below):

- **Use of the term sea spray aerosol as a synonym for marine aerosol.**
- **Results of direct measurements of the lidar ratio of marine aerosol are marginalized in the manuscript and not properly addressed in the discussion of the findings.**
- **A critical discussion of the AOD derived with SODA is missing. This makes it impossible to assess the reliability of subsequently deduced parameters, and thus, the scientific quality of this paper.**
- **There is not enough convincing evidence for a wind-speed dependence of the lidar ratio of marine aerosol.**

The manuscript presents a method to determine extinction-to-backscatter (lidar) ratios of marine aerosols from combined measurements of CALIPSO and CloudSat over the oceans during selected atmospheric conditions. While I think that the general idea of retrieving lidar ratios is presented in a convincing way, I have to criticize that the authors fail to convince me that the findings are reliable. This is mainly because the manuscript lacks a critical assessment of the representativeness of SODA AOD and a comparison of the retrieved lidar ratios to actual measurements of this parameter (which are available in the literature). Since the authors appear to be no lidar experts they might appreciate some input from the lidar community to strengthen the scope of this paper. The following comments follow the structure of the manuscript and are not ordered according to any priority.

To avoid confusion, I suggest to use the term marine aerosol instead of sea spray aerosol. If not defined otherwise, the latter refers to a production process (primary marine aerosol) while the former is associated with a location of origin and is commonly used by people that work with aerosol optical parameters. You have to keep in mind that a lidar will detect all aerosols within your layer of interest and not just the ones that are actual sea spray (i.e., of primary origin).

We have decided to retain the acronym SSA as it is now widely used in the sea spray remote sensing/modeling community. Concerning the point that a lidar will detect all aerosols, we believe there is a misunderstanding. Description of the scene selection algorithm (in section 2.4) clearly states that we start with clean marine aerosol Although we acknowledge that some natural continental aerosols and human-induced pollution can be miss-classified by CALIOP as clean marine, and caution readers when interpreting data near coastlines, suggestion that all aerosols within the layer of interest are included in the calculations is incorrect.

I realized the both sea spray aerosol and marine aerosol are used in the manuscript. In the first sentence of the second paragraph of the introduction, the authors even explain the difference between marine aerosol and sea spray aerosols! My comment regarding "all aerosols" was referring to the bulk of particles in the up to 2 km deep marine boundary layer. Most of these particles will not be the type of sea spray aerosol that is observed close to the ocean surface as a result of bubble-bursting processes. I am not familiar of anybody in the remote-sensing community who uses the term sea spray aerosols when referring to marine aerosol (i.e. aerosol of marine origin). In the remote-sensing community, SSA is usually used as an abbreviation for the single-scattering albedo. If the authors want to address the remote-sensing community, they have to adept to the terminology.

The authors should extend the literature review on the lidar ratio of marine aerosols. Currently, most references are of rather theoretical nature (i.e., modeling studies that obtain lidar ratios from measurements of the particle size distribution or lidar ratios obtained from measurements with passive sensors). Only one reference is given that presents a direct measurement (*Ansmann et al.*, 2001). However, this study did not focus on measurements of marine aerosol. While the authors state that direct measurements of the lidar ratio of marine aerosols are possible with HSRL (page 217, line 25), they miss to mention that this is also possible with Raman lidar (RL). More critically, the authors also forgot to review the rich literature of actual direct measurements of the lidar ratio of marine aerosols with well-calibrated HSRL or RL. Common values at the CALIOP wavelength of 532 nm are 23 ± 3 sr (RL, North Atlantic, *Müller et al.* 2007), 23 ± 5 sr (RL, Indian Ocean, *Müller et al.* 2007), 18 ± 2 sr (RL, Equatorial Atlantic, *Groß et al.* 2011), 20 ± 5 sr (HSRL, North Atlantic, *Burton et al.* 2012), and 18 ± 5 sr (HSRL, North Atlantic, *Groß et al.* 2013). I suggest that the authors revise the introduction to include a thorough review of **directly measured** lidar ratios of marine aerosol. Furthermore, the findings should be discussed in the context of these observed values.

Direct measurements of the lidar ratio with Raman and High Spectral Resolution Lidar are the only reliable benchmark the authors can compare their results to! It is unacceptable that these results are reduced to a side note in the end of the introduction and dumped in the supplement! Instead, the authors discuss a host of indirect results obtained from scattering calculations that need the assumption of a particle size distribution and a complex refractive index. Even though they are currently less becoming, the findings of this study have to stand up against actual lidar measurements rather than indirect results.

I can only repeat: please perform a proper literature study on lidar measurements of marine aerosols. I also recommend a look at *Weitkamp* (2005) as an introduction to lidar; particularly the chapters on Raman lidar, HSRL, and Lidar and Atmospheric Aerosol Particles.

Generally, a quantitative discussion and critical assessment of the findings in missing. This issue is already addressed for the lidar ratio in the previous comment. The AOD obtained with the SODA algorithm also needs to be put into the context of actual observations. I would assume that 500-nm AOD for conditions considered in this paper (only marine aerosols present in a single layer close to the surface, no elevated layers, no clouds) varies between 0.05 (*Kaufman et al.*, 2001) and 0.10 (*Smirnov et al.*, 2009, 2011) with a tendency towards the smaller values. For instance, *Smirnov et al.* (2002) report a mean AOD of 0.07 at 500 nm for marine aerosols observed at sites in the tropical Pacific. *Smirnov et al.* (2002) also give a review of AOD measurements over the oceans. However, it has to be noted that measurements at coastal sites are not synonymous with measurements of marine aerosols. I recommend that the authors could take a look at Sun photometer observations at remote islands or from ships (*Smirnov et al.*, 2002, 2009, 2011) to get a feeling for the values they can expect in pristine marine environment. Such values are much smaller than what is presented in Figure 1 and Tables 1 and 2 for SODA or given as a common value for marine aerosol in the introduction to this paper. In that context, CALIPSO-derived AOD seems more realistic to me. The complete lack of scrutinizing the representativeness of SODA AOD is the strongest weakness of the paper since it translates directly to the trustworthiness of the retrieved lidar ratios. I am afraid to say that without an assessment of the accuracy of the SODA AOD used for the situation considered in this study there is no value in the retrieved lidar ratios. My guess is that a decrease in the AOD used in the retrieval would lead to a decrease in the lidar ratio towards the values from measurements with HSRL and RL. This is an issue that deserves an equally extended discussion as the influence of the integrated attenuated backscatter coefficient on the accuracy of the retrieval.

This is a major issue that needs to be addressed to convince the reader that SODA AOT for marine aerosol is now flawed.

Following up on the previous comment, Figure 2 shows that SODA retrieves increased AOD for marine aerosol in the Yellow sea, around the Indian subcontinent, and to the west of the continents. This seems to be an artifact that is related either to a strong contribution of aerosols of other origin than marine or an effect of clouds. In the same figure, it is also not obvious that regions with generally higher wind speeds and more efficient production mechanisms would show increased AOD.

We believe the reviewer means Fig. 1, as Fig. 2 is reporting the lidar ratio, not the AOD. The increased AOD around the Indian subcontinent was identified as a probable artifact that was mentioned in the manuscript (section: Results, paragraph 1) and interpreted as contamination of continental pollution. The manuscript text has been revised to more clearly state this point: The region around the Indian subcontinent and over the Bay of Bengal is believed to be just a retrieval artifact.

Clouds always pose a challenge for satellite retrievals of aerosols. We have to the best of our ability, removed clouds by our described layer screening technique (see the *manuscript section 2.4). We have used the information from the CALIOP vertical feature mask to only analyze columns containing one aerosol layer (classified as clean marine) and no identified clouds. Nevertheless, hydrated aerosols near the cloud edges may lead to biases in CALIOP retrieved AOD. We attempt to mitigate this by requiring SODA aerosol retrievals to make up 70% or more of the 5 km CALIPSO aerosol layer product. In other words, there must be more than 10 retrievals in the 5 km averaging swath. This means that for any 5 km aerosol products that we use in our analysis, there are at least 11 CALIPSO shots (out of a possible 15) that make up the reported values. This ensures that aerosols near large clouds will not be included in the retrieval.*

Okay

The authors should state somewhere that CALIOP AOD is not considered as a reliable operational output. A comprehensive overview of CALIOP-derived AOD can be found in *Winker et al.* (2013). In my opinion, CALIOP AOD can only be used for cloud-free profiles for which a surface signal is detected. This information is provided in the 5-km aerosol profile product. I assume that using CALIOP observations according to the availability of SODA AOD intrinsically accounts for cloud-free conditions and for a surface signal being detected. Is that correct? It is worthwhile mentioning that somewhere in the Section 2.4 (Data selection method). The presence of clouds above the marine aerosol layer would increase the value of the total integrated backscatter coefficient.

In Section 2.4, the text from page 221, line 8 to page 222, line 14 basically describes the same procedure as the one from page 222, line 15 to page 223, line 11. I suggest to harmonize and shorten this section.

We believe that, for the sake of clarity, the text should remain as is. The first section referred to in the comment describes the scene selection component of the quality control algorithm (i.e., layer type and conditions of layer selection), whereas the text referred to in the second part of the comment explains the rest. We think the current state of the text helps readers easily understand what steps have been taken in the quality control algorithm.

It is my understanding that redundant text should be avoided in scientific writing. I still suggest to harmonize and shorten this section.

The description of the CALIPSO data retrieval lacks critical references regarding the instrument (*Winker et al.*, 2009), the feature-finding algorithm (*Vaughan et al.*, 2009), the lidar-ratio selection algorithm (*Omar et al.*, 2009), and the extinction-coefficient retrieval (*Young and Vaughan*, 2009). These references should be given in Section 2.1 and not in the discussion of the findings.

The references have been added to the revised manuscript.

Okay.

The authors use the terms integrated backscatter and integrated attenuated backscatter. My guess is that this is the parameter Column_Integrated_Attenuated_Backscatter_532 provided in the CALIOP aerosol product. It is not clear from the text if that is the case or if the authors obtain this parameter themselves. It is also not stated in Section 2.1 which CALIPSO products are included in the data analysis.

I found three different time periods for which data were considered: 2007 to 2010 (Introduction), Dec 2007 to Dec 2009 (Abstract, Conclusions), and Dec 2007 to Feb 2011 (Section 2.4). Please clarify which one is correct.

The text has been fixed; the proper dates are from December 2007 to February 2010.

Okay.

Please elaborate why there would be more misclassification in the low wind speed regime (page 225, line 23-26 and page 227, line 15-19). A lot of these cases occur over the southern oceans where the influence of anthropogenic pollution is almost negligible. I find it more convincing that the decreased signal-to-noise ratio of the CALIOP measurements during situations with low aerosol load introduces noise that leads to misclassification? Misclassification along the coastlines is more likely due to the effect of surface type on the aerosols detection algorithm (see, e.g., *Omar et al.* 2009 or *Kanitz et al.* 2014).

I suggest to move the supplementary material into the actual paper or at least add the occurrence rates of the different wind speed regimes to Table 2.

We have added the number of retrievals and the percentages to Table 2.

Okay. Now it is obvious that only 10% of cases are used to construct some kind of wind-speed dependence. The authors should clarify that the

numbers in parenthesis refer to the standard deviation of the lidar ratio per wind regime. This is not the actual retrieval error! I would be surprised if the relative (stochastic + systematic) error in retrieving the lidar ratio with the proposed method would be below 50%. In fact, it is presented in Figure 2 of (*Josset et al.*, 2011) that the error in retrieving lidar ratios with SODA inhibits reliable retrievals under low-AOT conditions.

If I understand this information correctly, 90% of all cases (wind speeds between 4 and 15 m/s) show lidar ratios of $S = 25 - 27\,\mathrm{sr}$. These values are slightly higher than the ones measured with RL or HSRL ($20 \pm 3\,\mathrm{sr}$) but show a comparable variation of only a few steradian **(This is not the actual uncertainty of the retrieval!)**. While it is conceptually comprehensible, I find it somewhat odd to deduce a wind-speed dependence of the lidar ratio from the remaining 10% of observations. Also, not much of a wind-speed dependence is left if the value for the lowest wind speed (that shows the highest standard deviation) is omitted.

I understand that wind-speed dependence is a major issue for the sea spray community. As a lidar person, I would like to comment that presenting more or less direct and systematic measurements of the lidar ratio of selected aerosol types is already an achievement that warrants publication—if it is done in a convincing way. Consequently (and as a reply to a comment by one of the official reviewers), I have to say that from a lidarist's point of view there is absolutely no value in a parametrization of the lidar ratio with wind speed!

I stand by this comment. As described earlier, I don't see enough evidence for a wind-speed dependence of the lidar ratio when considering the distribution of data points and likely retrieval errors. The authors also fail to mention why the lidar community should be interested in such a relationship.

Based on the values obtained from direct measurements of the lidar ratio of marine aerosols given above, it is my personal opinion that changing the value used in the CALIPSO retrieval by a few steradian would not lead to any improvements. I am afraid to say that there are too many stronger factors (clouds in a profile, signal-to-noise ratio, solar background, internal calibration, ...) that have an influence on the quality of CALIPSO products to seriously consider such a minor change. Since the extinction coefficient of marine aerosol is generally small, a slightly incorrect (if so!)

choice of the lidar ratio will only have small effects.

The authors state that the size distribution of marine aerosol changes with wind speed. It seems worthwhile to use these different size distributions to investigate by means of scattering calculations if they would lead to different lidar ratios.

These calculations were done with measured size distributions near Hawaii by Sayer et al. (2012). We have inserted the reference where appropriate.

It is customary in scientific writing to elaborate on the content of a specific reference instead of just adding it somewhere in the text. For instance, it would be of interest to the reader to learn about the outcome of these studies. Also it's worthwhile mentioning that these "measured" size distributions are actually the output of AERONET inversions and that a fixed refractive index has been used in the scattering calculations.

The authors state in the conclusions that the obtained lidar ratio of 26 sr leads to a better agreement of the AOD obtained through SODA and CALIPSO measurements alone. It would be just fair to also challenge the results obtained with SODA in the discussion of the findings. Following up on an earlier point of this short comment, I suggest to add a comprehensive assessment of the SODA AOD for the aerosol situation considered in this paper. It is hardly a surprise to come up with higher lidar ratios if SODA overestimates the AOD of marine aerosols. The authors spend the entire Section 4 on discussion one of the two input parameters to their retrieval. Why is the AOD as the second input parameter not assessed in the same way? This is an obvious flaw that should be corrected.

The SODA method has been extensively evaluated against High Spectral Resolution Lidar (HSRL) retrievals as well as MODIS observations (Josset et al., 2011, 2010, 2008). We strongly believe that the evaluation of the SODA algorithm against AERONET data is outside the scope of the current paper.

I strongly disagree for several reasons. The extensive evaluation the authors refer to consists of a comparison of (1) SODA AOT to four MODIS overflights (*Josset et al.*, 2008), of (2) SODA AOT to one month of MODIS AOT over the Gulf of Guinea (*Josset et al.*, 2010) which is not exactly representative for clean marine conditions, and (3) of SODA lidar ratios during three HSRL underflights of CALIPSO (*Josset et al.*, 2011). It would

also be of interest for the reader to know that *Josset et al.* (2008) state that the AOT calibration error of less than 15% only applies to wind speed between 3 and 10 m/s and that *Josset et al.* (2011) discuss the error in the lidar ratio retrieval as a function of AOT.

In that context I strongly believe that it is within the scope of the paper to convince the readers that SODA AOT for marine aerosol is reliable before any subsequent analysis is done with this parameter. As described in my previous comment on assessing SODA AOT, I am not convinced that reliable data are used in this paper.

Given that the conclusions of the paper still hold after the authors check for the issues raised above, I suggest going for a more daring title like: *Spaceborne observations of the lidar ratio of marine aerosols.* A paper with such a title would raise much more interest in the lidar community. From a lidar point of view, it would still be a strong and innovative paper if the authors omit the part on the wind speed dependence of the lidar ratio as the current evidence is not convincing enough to draw this conclusion.

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