Response to Anonymous Reviewer #1

We would like to thank Reviewer #1 for his/her comments. We have done our best to address each of the points as detailed below.

Note: All reviewer comments are in *italics*. All author responses are in normal format.

#1. *Title: The title is a bit misleading because it does not highlight what "a new study" means (new observations? new method?). In the abstract and in the conclusions it is specified that a new method id described. Therefore, I suggest to make the title clearer by using "a new method", "a new algorithm", etc.. or anything else equivalent.*

We agree with the referee, the title has been modified as: A new method deriving sea spray optical properties from multi-sensor spaceborne observations.

#2. *Abstract: Line 2: Replace the sentence "the assumption of an extinction-to-backscatter ratio" with "the assumption of the extinction-to-backscatter ratio".*

The abstract line has been changed.

#3. *Introduction: General remark: It would be good to provide a list of state-of-art available methods, which allow the retrieval of the lidar ratio. This could be done just in 1-2 sentences.*

A table with references of the currently available state-of-the-art retrieval methods has been added to the revised manuscript. We also added the following to the last paragraph of the introduction: "Other techniques like the inversion of AERONET radiometer data (Holben et al., 1998) along with Mie theory can also provide the lidar ratio. The supplementary Table S1 summarizes available methods to retrieve the lidar ratio along with some measured S_p values for marine aerosol. A more comprehensive list can be found elsewhere (Cattrall et al., 2005; Smirnov et al., 2001)."

#4. *Page 214, line 23: The acronym SSA sounds misleading in this context because usually it refers to "single scattering albedo". The authors should find an alternative acronym for the sea spray aerosol. Maybe SS (sea spray) is sufficient.*

In the sea spray aerosol remote sensing/modeling community, it is common to use the abbreviation SSA. Therefore, we have retained the abbreviation throughout the manuscript.

#5. *Page 214, line 24: Replace the sentence "SSA contributes an aerosol optical depth (AOD) of approximately 0.15" with "the contribution of sea salt aerosol to AOD if approximately 0.15".*

Corrected as suggested.

#6. *Page 216, line 21: Replace the sentence "Because of this" with "For this reason" or something equivalent.*

Corrected as suggested.

#7. *Page 216, line 23: The acronym S_p is introduced here for the first time with no clear meaning of the subscript p. Later in the manuscript, it's clear that p stands for particulate but it should be explicitly defined here.*

Corrected as suggested.

#8. *Page 217, line 17: Replace the sentence "we present a new method for deriving lidar ratios" with "we present a new method to derive lidar ratios"*

Corrected as suggested.

#9. *Methods: General remark: The title of this paragraph does not seem consistent with the ones of the sub-paragraphs. Please consider it to change it, or to change the ones of the sub-paragraphs or to re-organize this section.*

The header of the section was changed to "Instrumentation and Methods"

#10. *Page 218, line 5: Replace the sentence "The CALIPSO mission was launched on April 28, 2006. CALIPSO has been able to provide the scientific community with vertically resolved measurements of both aerosol and cloud optical properties like depolarization ratio (a measure of particle sphericity), AOD, and ice/water phase since June 2006" with "The CALIPSO mission (INSERT A REFERENCE HERE), launched on April 28, 2006, has been able to provide the scientific community with vertically resolved measurements of both aerosol and cloud optical properties like depolarization ratio (a measure of particle sphericity), AOD, and ice/water phase since June 2006".*

This was changed as suggested and the reference Winker et al. (2009) was added.

#11. *Page 219, lines 10, 12: Repetition of "Therefore" at the beginning of the sentences. Please modify.*

Corrected as suggested. The revised manuscript reads:

"Since the radar signal attenuates mostly due to water vapor and the lidar signal weakens mostly due to aerosols, after the radar signal is corrected for attenuation by water vapor and oxygen, the change in the radar-to-lidar signal ratio is directly related to aerosol abundance (Josset et al., 2008; 2010a). Therefore, by using observations from two different sensors, SODA can eliminate uncertainties induced by the CALIOP aerosol extinction algorithm over oceans."

#12. *Page 220, line 14: Replace the sentence "With these assumptions in mind, integration: : :" with "Based on these assumptions, the integration: : :".*

Corrected as suggested.

#13. *Page 220, line 20: Replace the sentence "we get an equation for a columnar particulate* *lidar ratio as" with "the equation for a columnar particulate lidar ratio is".*

Corrected as suggested.

#14. *Page 221, line 1: Replace "The equation" with "Eq. 4".*

Corrected as suggested.

#15. *Page 225, line 11: Provide a reference.*

The following references have been added to the revised manuscript:

Smirnov, A., Holben, B., Eck, T., Dubovik, O. and Slutsker, I.: Effect of wind speed on columnar aerosol optical properties at Midway Island, J. Geophys. Res., 108(D24), 4802, doi:10.1029/2003JD003879, 2003.

Sayer, A., Smirnov, A., Hsu, N. and Holben, B.: A pure marine aerosol model, for use in remote sensing applications, J. Geophys. Res., 117, doi:10.1029/2011JD016689, 2012.

#16. *Page 226, line 9: Replace "this figure" with "Fig. 3".*

Corrected as suggested.

#17. *Page 231, line 10: Avoid repetition of "that" in the sentence, if possible.*

Corrected as suggested.

Response to Anonymous Reviewer #2

We would like to thank Reviewer #2 for his/her comments. We have done our best to address each of the points as detailed below.

Note: All reviewer comments are in *italics.* All author responses are in normal format.

#1. *The paper shows that the lidar ratio has considerable spread globally. Because of this, changing the lidar ratio from the CALIOP default to 26 sr, while improving bias in the retrieved CALIOP AOD from SSA, will not drastically improve the error in spatial and seasonal variability. The current paper appears a half-step towards formulation of a lidar ratio that is a function of the wind speed that would significantly improve the CALIOP retrieval relative to the proposed change in global lidar ratio here. Perhaps the step of parameterizing the lidar ratio with wind should be included in this paper? It appears that minimal extra work would be required and the value of this paper to the community would be increased substantially. Unless there are significant reasons for leaving that work to a separate paper I think effort should be made to parameterize the SSA lidar ratio in this study.*

We agree with the reviewer that a parameterization of lidar ratio with wind speed could add value to the paper. However, we note that the lidar ratio is dependent on far more properties than wind speed: mainly atmospheric variables like relative humidity and boundary layer depth and to lesser extent chemical properties. Thus, parameterizing the lidar ratio as a function of ocean surface wind speed may provide an oversimplified expression to the scientific community. Nevertheless, we agree with the reviewer and have included the parameterization as online supplementary Figure S2 with a word of caution. The equation for the least squares linear regression is $S_p = -0.5U_{10} + 28.4$ with an $R^2 = 0.76$.

#2. *Marine organic aerosol may contribute to the spatial and temporal changes in aerosol properties (e.g. O'Dowd et al., 2004). There is limited mention of the impact of marine organic aerosol on the lidar ratio, beyond mention of general chemical composition. Might marine organic be expected to have different optical properties to pure sea salt and affect the retrieval? Might this contribute to the remaining spread and seasonal geographical distribution once wind speed has been accounted for? It may be worth investigating this in future and going into a more detail in this paper how the chemical composition of marine aerosol would be expected to alter the retrieval.*

The reviewer is correct. Organic aerosol is expected to contribute different optical properties than pure sea salt. These differences will indeed propagate through to the lidar ratio retrievals, contributing to the results in this paper. Unfortunately, exactly how we expect these differences to manifest is unclear. A significant amount of laboratory and in-situ evidence has shown that increases in the organic content of inorganic aerosol can influence hygroscopic properties. These changes in aerosol hygroscopicity will probably have an effect on the optical properties for marine aerosol as well. The following was added to the conclusions section of the manuscript: "A large body of experimental data suggests that increases in the organic fraction of marine aerosol can have implications on hygroscopicity (e.g., Saxena et al., 1995; Fuentes et al., 2011; Ovadenevaite et al., 2013) and could potentially influence our results."

#3. *pg221 line 16 - should this be less than (<) 0.01 sr^{-1} based on Omar et al., (2009)?*

We thank the reviewer for catching this inconsistency. The revised manuscript reads: "We start with clean marine aerosol that is identified based on surface type (as determined by the location of the satellite) and either total integrated attenuated backscatter $\gamma' > 0.01$ km^{-1}sr^{-1} or total integrated attenuated backscatter $\gamma' < 0.01$ km^{-1}sr^{-1} and volume depolarization ratio $\delta' < 0.05$ (Omar et al., 2009)."

#4. *Figure 2 - Just a suggestion, but rather than the rainbow color scale it may be informative to use another color scale (e.g. blue-white-red) to highlight the difference relative to the CALIOP assumed values.*

We decided to retain the current figure since in Fig. 1 we show the difference between SODA and CALIOP AOD, which can be a proxy for the difference map suggested by the reviewer.

References:

Fuentes, E., Coe, H., Green, D. and McFiggans, G.: On the impacts of phytoplankton-derived organic matter on the properties of the primary marine aerosol - Part 2: Composition, hygroscopicity and cloud condensation activity, Atmos. Chem. Phys., 11, 2585-2602, doi:10.5194/acp-11-2585-2011, 2011.

Omar, A. H., Winker, D. M., Kittaka, C., Vaughan, M. A., Liu, Z., Hu, Y., Trepte, C. R., Rogers, R. R., Ferrare, R. A., Lee, K. P., Kuehn, R. E. and Hostetler, C. A.: The CALIPSO Automated Aerosol Classification and Lidar Ratio Selection Algorithm, J. Atmos. Ocean. Technol., 26, 1994-2014, 2009.

Ovadnevaite, J., Ceburnis, D., Martucci, G., Bialek, J., Monahan, C., Rinaldi, M., Facchini, M. C., Berresheim, H., Worsnop, D. R. and O'Dowd, C.: Primary marine organic aerosol: A dichotomy of low hygroscopicity and high CCN activity, Geophys. Res. Lett., 38, doi:10.1029/2011GL048869, 2011.

Saxena, P., Hildemann, L. M., McMurry, P. H. and Seinfeld, J. H.: Organics alter hygroscopic behavior of atmospheric particles, J. Geophys. Res., 100, 18755-18770, doi:10.1029/95JD01835, 1995.

Response to the comments provided by M. Tesche

We the authors thank Dr. Tesche for his attention to our study and his insightful critique of the manuscript. We have done our best to address each of his points as detailed below.

Note: All comments by M. Tesche are in *italics.* All author responses are in normal format.

#1. *To avoid confusion, I suggest the use of the term marine aerosol instead of sea spray aerosol (SSA). 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. It should be borne 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). I am aware that SSA is the term commonly used by the sea spray aerosol community so at the very least this should be clarified when the SSA acronym is introduced in the manuscript.*

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.

#2. *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 South American and African landmasses. This seems to be an artifact that is related either to a strong contribution of non-marine aerosols or an effect of clouds. In the same figure, there appears to be at best a weak relationship between wind speed and AOD; those regions generally expected to have higher wind speeds do not show markedly 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."

#3. *This seems to be an artifact that is related either to a strong contribution of non-marine aerosols or an effect of clouds.*

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.

#4. *Generally, a quantitative discussion and critical assessment of the manuscripts findings is missing. The AOD obtained with the SODA algorithm must also be compared to direct measurements of AOD. Therefore I strongly suggest to validate the SODA AODs with actual measurements at suitable AERONET stations.*

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.

#5. *In Section 2.4, the text from page 221, line 8 to page 222, line 14 simply describes the same procedure as the one from page 222, line 15 to page 223, line 11. I suggest the authors harmonize these descriptions omitting one of them.*

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.

#6. *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.

#7. *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.

#8. *I suggest the authors 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.

#9. *In order to strengthen the authors' findings, it seems worthwhile to use these different size distributions to investigate by means of scattering calculations whether 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.

References:

Josset, D., Pelon, J., Protat, A. and Flamant, C.: New approach to determine aerosol optical depth from combined CALIPSO and CloudSat ocean surface echoes, Geophys. Res. Lett., 35, L10805, doi:10.1029/2008GL033442, 2008.

Josset, D., Pelon, J. and Hu, Y.: Multi-instrument calibration method based on a multiwavelength ocean surface model, Geoscience and Remote Sensing Letters, IEEE, 7, 195-199, doi:10.1109/LGRS.2009.2030906, 2010.

Josset, D., Rogers, R., Pelon, J., Hu, Y., Liu, Z., Omar, A. and Zhai, P.: CALIPSO lidar ratio retrieval over the ocean, Opt. Express, 19, 18696-18706, doi:10.1364/OE.19.018696, 2011.

Omar, A. H., Winker, D. M., Kittaka, C., Vaughan, M. A., Liu, Z., Hu, Y., Trepte, C. R., Rogers, R. R., Ferrare, R. A., Lee, K. P., Kuehn, R. E. and Hostetler, C. A.: The CALIPSO Automated Aerosol Classification and Lidar Ratio Selection Algorithm, J. Atmos. Ocean. Technol., 26, 1994-2014, 2009.

Sayer, A., Smirnov, A., Hsu, N. and Holben, B.: A pure marine aerosol model, for use in remote sensing applications, J. Geophys. Res., 117, D05213, doi:10.1029/2011JD016689, 2012.

Vaughan, M. A., Powell, K. A., Kuehn, R. E., Young, S. A., Winker, D. M., Hostetler, C. A., Hunt, W. H., Liu Z., McGill, M. J. and Getzewich, B. J.: Fully automated detection of cloud and aerosol layers in the CALIPSO lidar measurements, J. Atmos. Ocean Technol., 26, 2034-2050, doi:10.1175/2009JTECHA1228.1, 2009.

Winker, D. M., Vaughan, M. A., Omar, A., Hu, Y., Powell, K. A., Liu, Z., Hunt, W. H. and Young, S. A.: Overview of the CALIPSO Mission and CALIOP Data Processing Algorithms, J. Atmos. Oceanic Technol., 26, 2310-2323, doi: 10.1175/2009JTECHA1281.1, 2009.

Young, S. A. and Vaughan, M. A.: The retrieval of profiles of particulate extinction from cloud-aerosol lidar infrared pathfinder satellite observations (CALIPSO) data: algorithm description, J. Atmos. Oceanic Technol., 26, 1105-1119, doi:10.1175/2008JTECHA1221.1, 2009.