

Anonymous Referee #1

We thank the Referee for the thorough review of the manuscript and the constructive comments, which contributed to the improvement of this manuscript.

In response, the manuscript is substantially revised with the following:

- 1) Updated analysis of global W data to develop $W(U10)$ parameterization.
- 2) Extended analysis of regional W data to develop $W(U10,T)$ parameterization with SST explicitly included; this was done for both quadratic and cubic wind exponents.
- 3) Analysis for statistical significance (with Student's T-statistics and ANOVA) of new and previous W parameterizations.
- 4) Extended 'Methods' section to justify and clarify approach, data, and implementations.
- 5) Revised and extended 'Results and Discussion' section to clearly describe results and give substantive and quantitative interpretations and conclusions.

The table of contents of the revised manuscript is added after the responses for reference.

Manuscript revisions with track changes are provided in a separate pdf file.

Several comments and questions are similar in all 3 reviews (e.g., uncertainty not reduced, quadratic wind speed exponent, embedded secondary forcing, intercept interpretation). To avoid repetitions, we attempted combining responses to these common points in one file. We found, however, that one-fits-all responses do not always address the reviewers' comments and questions fully. Thus, risking some repetitions, we proceeded with a specific response to each comment.

Responses are presented below in sequence: (1) the original comment from the Referee (in bold italic), there are 10 comments; (2) our response; (3) changes in manuscript.

1.1 In general, the manuscript has poor flow. The authors jump from topic to topic with little flow between main points. There is a lot of redundancy in the text that makes it hard to follow.

1.2 We can see how a perception of "poor flow" of the manuscript can arise. The study and results presented in the manuscript are on three somewhat distinct yet interweaved topics, namely: (i) assessment of satellite-based W data; (ii) parameterization of W; and (iii) application of new W parameterization to predict SSA production. Being well aware of this, we gave a roadmap of our approach in the end of the Introduction (page 21225, lines 3-16) and occasionally listed the points considered in each subsection in short preamble (e.g., p. 21225, lines 18-20).

1.3 The manuscript is now extensively revised. The flow is different and we believe much improved.

2.1 There is no independent verification of the parameterization. Without comparison with other measured, remotely sensed or modeled data I do not see sizable contribution to scientific progress in the field.

2.2 Our intended verification of the performance of the new $W(U10)$ parameterization was given with Fig. 12 and its discussion (old Sect. 4.2.2) as well as the extensive comparison and discussion of the SSA production estimates obtained with the new $W(U10)$ parameterization to previous SSA production estimates (old Sect. 4.3).

2.3 We added new Fig. 13a to compare W values, obtained with new parameterizations $W(U_{10})$ and $W(U_{10},T)$ for 10 and 37 GHz, to in situ and WindSat W data. Description and discussion of Fig. 13a are added in new Sect. 3.3.2.

Comparisons to the published in situ W data demonstrate order-of-magnitude consistency of the W values from the new parameterizations. Because there are no other remotely-sensed W data except those from WindSat, the most we can do at the moment is to evaluate how well the new parameterizations can replicate the trend and the spread of the satellite-based W . Recently, W values from a global wave model were compared to W from MOM80 and WindSat by Leckler et al. (2013), so one can evaluate where modeled W values stand in the comparison of data and parameterizations of W . All parameterized W values shown in Fig. 13a are calculated using U_{10} and T from the whitecap database, i.e., U_{10} from QuikSCAT and T from GDAS.

3.1 While the proposed parameterization for W has fair agreement with other parameterizations, the authors fail to distinguish the proposed formulation from previously proposed parameterizations after using similar retrieval algorithms (i.e. SAL13).

3.2 We do not expect to prove/show a distinctly different parameterization from that of SAL13 because, indeed, we and SAL13 use the same W data. What we show in this study (and have said in the initial text on p. 21243, lines 3-7) is that a different analysis (a ‘top down’ approach from global to regional scales) gives similar results and this proves that the outcome is robust. What is added with this work to previous analysis of the whitecap database (i.e., SAL13) is the analysis and quantification of the possible intrinsic correlation in the W data and how this could affect W predictions with the new $W(U_{10})$ expressions.

3.3 For the revised manuscript, in addition to the above, we extended the analysis to derive a $W(U_{10},T)$ parameterization from regional W data sets in addition to the $W(U_{10})$ parameterizations at 10 and 37 GHz from the global W data set. We discuss/justify the approach for the parameterizations (new Sect. 2.1) and its implementation (new Sect. 2.3) and present the results in Sect. 3.2. The comparison to previous parameterizations, including to those of SAL13, is now extended using two metrics—percent difference between different parameterizations and tests for significant differences (new Sect. 3.3).

4.1 When applying the new parameterization to a global model which predicted SSA flux, the authors showed their parameterization reduced SSA emissions in polar regions while increasing emissions in tropical regions. Model analyzes were in the context of mass concentration and was limited to supermicron sized SSA. The argument for using supermicron sized aerosols (i.e., that sub-micron size range additionally includes organic material) does not hold water. Organic enrichment becomes important for particles with <200 nm in diameter. Such particles do not contribute considerably to overall mass. At the end, the point of this exercise is not well explained.

4.2 We agree with the Referee that the justification with the organic content is not strong and acknowledge that we should have explained our choices for estimating SSA emissions better.

4.3 We revised Sect. 2.4 in Methods to give justification for our choice of size distribution with the following arguments (new Sect. 2.4.2).

Generally, the division of the SSA particles into small, medium, and large sizes is well warranted when one considers the climatic effect to be studied. For example, submicron particles are important for scattering by SSA (direct effect) and CCN formation (indirect effect), while supermicron particles are important for heat exchange (via sensible and latent heat fluxes) and

heterogeneous chemical reactions (which need surface and volume to proceed effectively). For the purposes of this study, we do not focus on how the choice of the size distribution will affect the SSA estimates. Rather, at a fixed distribution, we want to see how W data (and W parameterizations based on them), which carry information for the influences of many factors, would affect SSA estimates. In this sense, we can use any size distribution.

The size range of 1 to 10 μm that we have chosen is in the range of medium (supermicron) SSA particles (e.g., de Leeuw et al., 2011, §8). This is the range for which Monahan et al. (1986, or M86) size distribution is valid. Table 3 in Textor et al. (2006) shows that the M86 size distribution, in its original or modified forms, is widely used. Also, Table 2 of Grythe et al. (2014) shows that this size range is a recurring part of the size ranges used in all SSSFs. As the Referee has noted, the SSA particles below $r_{80} = 0.1 \mu\text{m}$ contribute little to the overall mass ($\sim 1\%$ according to Fachini et al. (2008)). We quantify the expected discrepancy due to neglecting particles for $0.1 < r_{80} < 1 \mu\text{m}$ to be 14% using Grythe et al. (2014) estimates of SSA with M86 over two different sizes. We use this assessment in our subsequent analysis of SSA emissions (Sect. 3.4).

5.1 *The total predicted sea spray aerosol mass varies by several orders of magnitude. So if the emissions inferred by the current parameterization are within this range, does that prove its validity?*

5.2 Yes, it does. That emissions inferred by our new parameterization are within this range shows that our modified SSSF gives consistent estimates, which effectively proves its validity. Certainly, we do not want to be an outlier among SSA emission estimates, especially for a variability range of 2 orders of magnitude. What is more important, however, is that the spatial distribution of this total SSA emission is significantly different from those of previous SSSF predictions. Our new Fig. 14 (old Figs. 10-11) illustrates the global spatial distribution of SSA emissions and a difference map with SSA estimate using MOM80 parameterization.

5.3 We added the following text in new Sect. 3.4.

Previously modeled total dry SSA mass emissions vary by two orders of magnitude because of a variety of uncertainty sources (Sect. 1): $(2.2\text{--}22)\times 10^{12} \text{ kg yr}^{-1}$ (Textor et al., 2006, their Fig. 1a; de Leeuw et al., 2011, their Table 1); and $(2\text{--}74)\times 10^{12} \text{ kg yr}^{-1}$ for long-term averages (over 25 years) (G14, their Table 2, excluding 3 outliers). The impact of the modeling method used has to be acknowledged too. Grythe et al. (2014) suggest that the spread in published estimates of global emission based on the same M86 SSSF (Eq. (4)), from 3.3×10^{12} to $11.7\times 10^{12} \text{ kg yr}^{-1}$ (Lewis and Schwartz, 2004), can be attributed to differences in model input data and resolution differences. An example of the same SSSF yielding different results when applied in different models is also seen in the work of de Leeuw et al. (2011, their Table 1).

For a meaningful comparison of our results to SSA emissions obtained with other SSSFs, we attempt to remove (or at least minimize) the impact of the modeling method. As in this study (see Sect. 3.4), G14 used the same model (i.e., input data and configuration) to evaluate 21 SSSFs, including that of M86, against measurements. We thus can infer a “modelling” factor using our and G14 results obtained with M86 SSSF. We find that the G14 estimate of SSA emission from M86 ($4.51\times 10^{12} \text{ kg yr}^{-1}$) is 1.55 times larger than our estimate of $2.9\times 10^{12} \text{ kg yr}^{-1}$ from M86 and MOM80. We apply this factor of 1.55 to our SSA emission estimated with the new $W(U_{10}, T)$ parameterization and obtain a “model scaled” value of $6.75\times 10^{12} \text{ kg yr}^{-1}$. Our “model scaled” estimate of the SSA emission is close to the median $5.91\times 10^{12} \text{ kg yr}^{-1}$ of the SSA emission reported by G14. This shows that an SSSF with a magnitude factor derived from satellite-based W data provides reasonable and realistic predictions of the SSA emission.

6.1 The submicron range is the most likely size range influencing direct and indirect radiative forcing. The authors' analysis of SSA emissions with the new parameterization fails to highlight this reality.

6.2 We are well aware of this reality. This is seen on page 21224 where we have mentioned the importance of SSA for the direct and indirect radiative effects on climate in Lines 1-4. The importance of SSA to other climate processes is listed in the same paragraph. We have not mentioned specific sizes of the SSA suitable for each of these processes, because in Lines 1-2 on page 21225 we state that for the objective of the study we focus on the effect of W on SSA estimate.

6.3 We revised the last paragraph on page 21224 (Sect. 1) to more clearly state the focus of this study on W (the magnitude factor in the SSSF), not on the size distribution (the shape factor in the SSSF); the magnitude and shape factors are now clearly introduced in Sect. 1. Specific sizes for specific climate effects are now mentioned in our justification for the chosen size distribution (new Sect. 2.4.2) (see our response to comment 4).

7.1 There are lots of speculations in the paper that are not supported by the facts. For example, the discussion regarding 37GHz vs 10 GHz intercept is not convincing.

7.2 We agree with the Referee that our discussion on this subject could have been presented better. Yes, the interpretation of the y-intercept was speculative at the moment, and we did admit this on page 21231 (lines 21-22). Still, by providing data points globally and over all seasons, the satellite-based W data offer possibilities for new insights. The observation of different W variability for active and decaying whitecaps (approximated by W values at 10 and 37 GHz, respectively) is one example for such new insight.

7.3 We revised the manuscript to introduce the currently accepted interpretation of negative y-intercept (Sect. 2.1). Then in Sect. 3.1.1, we propose broader interpretation of the y-intercept in W(U10) expressions, be it negative or positive. Briefly, we promote the hypothesis that positive y-intercept could be interpreted as a measure of the capacity of seawater with specific characteristics, such as SST (thus viscosity), salinity, and surfactant concentration, to affect the extent of W. These secondary factors do not create whitecaps per se. Rather, they prolong the lifetime of the whitecaps thus contribute to W by altering the characteristics of submerged and surface bubbles such as stabilization and persistence by surfactants or rise velocity variations that replenishing the foam on the surface at different rates. These processes ultimately augment or decrease W and the y-intercept can be thought of as a mathematical expression of this static forcing (as opposed to dynamic forcing from the wind). In this light, our data showing negative y-intercept for W values at 10 GHz is consistent with our and SAL13 analysis that active whitecaps are less affected by secondary factors. However, secondary factors do affect strongly residual whitecaps and the positive y-intercept for our W values at 37 GHz can be interpreted and used to quantify this static influences. This is a hypothesis which is worth promoting for consideration, debate, and further verification by the community.

8.1 The discussion about the "secondary factors" being "imbedded in the exponent of the wind speed dependencies" is misleading. The influence of secondary factors can only be ascertained by the satellite based estimates of W augmented by additional data sets for directional wave spectra, currents (speed and direction), and proxies for surfactants such as ocean color, chlorophyll a, or oceanic primary production. Such studies should be conducted as case studies on regional scales.

8.2 We agree with the Referee that the most rigorous way to fully parameterize the influence of secondary factors on W is to have a large database of W values concomitant with additional variables such as those the Referee has listed. The need for such a database has justified the work of Anguelova and Webster (2006, their Sect. 2 and specifically §16 and §22) on obtaining W from satellite-borne radiometric measurements. Initial version of the database of W and additional variables built by Anguelova et al. (2010, <https://ams.confex.com/ams/pdfpapers/174036.pdf>) and described by SAL13 (their Sect. 3.1) is used in this study.

We respectfully disagree with the Referee's descriptor "misleading." Our approach to parameterize secondary forcing is now extended and clearly presented in new Sect. 2.1. We show the concept that the variability of W caused by secondary factors is expressed as a change of the wind speed exponent is not new. The Monahan and O'Muircheartaigh (1986) analysis of five data sets showed that the variability of W caused by SST (and the atmospheric stability) affect significantly the coefficients in the wind speed dependence $W(U10)$, especially the wind speed exponent. The survey of $W(U10)$ parameterizations by Anguelova and Webster (2006, their Tables 1 and 2) also clearly shows that each campaign conducted in different regions and conditions comes up with a specific wind speed exponent. This strongly suggests that the influence of secondary factors is expressed as a change of the wind speed exponent.

8.3 We extended our regional analysis to develop $W(U10,T)$ parameterizations using empirical (adjusted) and cubic wind exponents. We used significance tests (Student's T-statistics and ANOVA) to establish similarity and differences between $W(U10)$ and $W(U10,T)$ with both empirical and cubic exponents. We found that the $W(U10)$ trend predicted with a quadratic wind speed exponent does not differ significantly from the $W(U10)$ trend predicted either with quadratic or cubic $W(U10, T)$. This result clearly shows that to a large extent, the adjusted wind exponent accounts for the change in the trend caused by SST and other secondary influences. Our new Sect. 3.3.2 shows that explicitly accounting of SST (and eventually other factors) helps to model the spread, not the trend, of the W data.

We describe our approach in Sect. 2.1, the significance test used in Sect. 2.3, and give the results regarding differences between parameterizations that account for variability implicitly or explicitly in Sect. 3.3. Through the text, with each new result presented, we drive the point that the adjustment from cubic to quadratic wind exponent accounts to a large extent for secondary forcing.

9.1 The new parameterization fails to reduce uncertainty in predicting sea-spray aerosol (SSA) flux. To what degree is the uncertainty in SSA flux attributed to uncertainty in predicting W versus other aspects of traditional sea-spray source functions (SSSF)?

9.2 Indeed, we do not report reduced uncertainty in predicting SSA flux. There are many uncertainty sources yielding wide spread of predicted SSA emissions. With our study, we address only one of the uncertainty sources—that associated with the natural variability of the whitecaps.

The Referee's question prompted us to use comparisons between our and Grythe et al. (2014) results for SSA fluxes to examine and quantify variations of SSA emissions attributed to magnitude and/or shape factors.

9.3 New Sect. 3.4 is revised and extended to give our new results.

These results are summarized in the Conclusions as follows:

With or without the SST effect included in the SSSF, SSA emissions obtained with the new $W(U_{10}, T)$ parameterization vary by ~50%. Different approaches to account for SST effect yield ~67% variations. Different models for the size distribution applied to different size ranges lead to 13%-42% variations in SSA emissions.

We conclude Sect. 3.4 with the following:

On the basis of these assessments, we can state that the inclusion of the SST effect in the magnitude factor and/or the choice of the shape factor (size range and model for the size distribution) in the SSSF can explain 13%-67% of the variations in the predictions of SSA emissions. The spread in SSA emission can thus be constrained by more than 100% when improvements of both the magnitude and the shape factor are pursued. Our results on the W parameterization (Fig. 13a) suggest that accounting for more secondary forcing in the magnitude factor would explain more fully the spread among SSA emissions. Because, after wind speed, the most important secondary factor that accounts for variability in W is the wave field (SAL13), efforts to include wave parameters in W parameterizations are well justified.

10.1 Figures appear to have been generated with different software packages.

10.2 Yes, indeed. We have used Python, IDL, and Excel. Respectfully, we do not see this as a problem in presenting our results and drawing conclusions.

10.3 No changes were made regarding comment 10.

Specific Comments

Page 21221; Line 1: Awkwardly worded first sentence which fails to highlight the importance of reducing uncertainty of SSA flux.

Agreed. We removed it. The importance of reducing the uncertainty of the SSA flux is mentioned on page 21223 lines 24-25 and page 21224 lines 1-14.

Page 21223; Line 15: Acronym SSA used prior to defining SSA. SSA acronym is defined on Page 5, Line 24.

Now fixed, acronym SSA introduced on first use of “sea spray aerosols” in the Introduction.

Page 21224; Line 11: Neither evidence nor citation is made to support this statement. Suggest this as an explanation versus declaring as fact.

Yes, this was our explanation. The extensive investigation of Salisbury et al. (2013) on the W variability using year-long satellite-based W data was cited in Line 12 as a basis for this explanation. With the extensive revisions of the manuscript, this statement now is lost.

Page 21225; Line 23: Continue to use whitecap fraction instead of “ W ”. Authors flip back and forth (e.g. Page 7; Line 24) on notation. Please use W to represent whitecap fraction after defining whitecap fraction as W .

Yes, we use both “ W ” and “whitecap fraction” depending on the context. Following the Referee’s comment, the specific example and other cases have been changed from “whitecap fraction” to “ W ”

Page 2138; Line 27: Please reword.

This text is removed.

Abstract

1 Introduction

2 Methods

2.1 Approach to derive whitecap fraction parameterization

2.2 Data sets

2.2.1 Whitecap database

2.2.2 Regional data sets

2.2.3 Independent data source

2.3 Implementation

2.4 Estimation of sea spray aerosol emissions

2.4.1 Use of discrete whitecap method

2.4.2 Choice of size distribution

3 Results and Discussion

3.1 Parameterization from global data set

3.1.1 Wind speed dependence

3.1.2 Intrinsic correlation

3.2 Regional and seasonal analyses

3.2.1 Magnitude of regional and seasonal variations

3.2.2 Quantifying SST variations

3.3 New parameterization of whitecap fraction

3.3.1 Comparisons to W parameterizations

3.3.2 Comparisons to W data

3.4 Sea spray aerosol production

4 Conclusions

Data availability

Acknowledgements

References

Table 1

Figure captions