Author's response to Referee #2

The thoughtful reading and the time dedicated by the reviewer are highly appreciated. The provided major as well as the minor comments are an important feedback that enabled better focusing of the scientific content and improvement of the manuscript quality. Below please find our point-to-point replies. The responses to the reviewer comments are given in blue text; the original reviewer comments are in black text.

Anonymous Referee #2

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This is an interesting and well written article that describes how the composition of aerosols at an inland site can change dramatically on a daily basis because of the influence of sea breezes. The authors point out that this can have a significant impact on the atmospheric radiative effect at the site. I have only a few suggestions for improvement.

Major issues

Page 11, lines 1-24 and Figure 7: Interesting discussion about how the refractive index changes with air mass and water vapor. The authors use standard deviations for the error bars in panels c-f of Figure 7 to understand the differences in the observations during low and high water vapor periods. However, it would be more useful to use the standard deviation of the means for this application (i.e., SDOM, or standard errors). This will decrease the contribution of random noise to the size of the errorbars, and it will provide the reader with an understanding of whether these differences are statistically significant at the 1-sigma level (i.e., datasets with overlapping SDOM errorbars are not significantly different). The authors should also indicate how many data points are used to compute the means in panels c-f.

Thank you for this suggestion, the standard errors are presented in the revised version. It is indeed appropriate. The cases where the standard errors overlap the means are indicated in the text as non-significant (imaginary refractive indexes during dust season, Fig. 7f). The number of data points used is added as well.

Page 11, lines 18-24: The authors bring up the topic of ssa in this paragraph, but don't really take it anywhere. You could isolate the effect of refractive index on the ssa for Aug 16 w/o much work though. . . that is, compute the ssa of the sea breeze aerosols using the SD of the pre-breeze particles. This will provide a Delta ssa associated with the size change. Similarly, you could compute the ssa of the pre-breeze particles using the refractive index of the sea-breeze particles; this will provide a Delta ssa associated with refractive index. This type of calculation can provide the reader an idea of how much of the ssa change is associated with size and how much is associated with composition, and it will make this paragraph more interesting.

Absolutely agree that the suggested calculation makes the analysis more interesting. The Delta SSAs due to the size changes and the compositional changes are added, and the next discussion is provided: "Because both the size distribution and the complex refractive index change during the sea breeze, it is interesting to evaluate their specific contribution to the changes in the SSA. To address this, we calculate the SSA assuming that only the size distribution is changing while the refractive index is the same and vice versa. The difference

in the SSA of the before-sea-breeze aerosol model minus the SSA of the modified aerosol model is -0.002/-0.001/0.001/0.003 for the size change and 0.015/0.009/0.003/0.003 for the refractive index change. The calculated differences show that the scattering effectiveness increases at the shorter and decreases at the longer wavelengths due to the size change, and decreases at all the wavelengths due to the compositional change. It shows that there is a partial compensation of the decrease in SSA at the shorter wavelengths because of the size shift."
It has also be mentioned that a mistake in the reported SSA values was found during the revision. However, it does not change the reported tendencies, conclusions or any other reported results.

Figures 8 & 9: Is there a discrepancy here?... The coarse mode is dominated by dust before the sea breeze in Figure 8, but Figure 9 indicates that there are more marine particles than dust particles at all radii > 0.5 um.

It should be clarified in the text that Figures 8 & 9 are not directly comparable. In fact, one must keep in mind the differences in measurement techniques (aerodynamic diameter for cascade impactors in Figure 8 versus geometric diameter determined by electron microscopy in Figure 9), that is:
- Figure 8 shows the relative proportions of particle types as a function of the aerodynamic size range. The size-segregated sampling by cascade impaction is based on an aerodynamic cut-off diameter with 50% efficiency $D_{ae,50}$ depending on particle density. The Dekati impactor used in this study is calibrated for a particle density of 0.93 g cm^{-3} (Marjamaki et al., 2000). This last information is now added in the manuscript (page 6, line 30-31 of the initially submitted version). As can be read on page 11, line 31, the "coarse fraction" refers to particles collected on the stage with the aerodynamic diameter range 2.5-10 µm and the "fine fraction" refers to particles collected on the stage with the aerodynamic diameter range 1-2.5 µm.
- On the other hand, the number size distribution presented in Figure 9 reports radii values for all analyzed particles collected on both stages (1-2.5 µm and 2.5-10 µm). The geometric radius in this analysis is derived from equivalent circle area of the 2D-projected particle on SEM images.
The next clarification is added in the text of the revised version (at the beginning of section 5.2): "In addition, it should be realized that the size distributions of the particle types in Fig. 9 are not directly comparable to the relative proportions of particle types per size fraction in Fig. 8. This is because the particle type proportions reported in Fig. 8 are for the size fractions of a cascade impactor, which are defined by an aerodynamic cut-off diameters, while Fig. 9 presents the geometric radius derived from equivalent circle area of particles observed by SEM."

I really enjoyed the analysis of the effect of core-shell morphology on the AERONET retrievals (Section 7). I have a couple of additional points that I believe are worth including in the manuscript:
+ Water shell thicknesses of 10% and 40% correspond to geometric hygroscopic growth factors of 1.11 and 1.67 (GF = r / r_core). A value of GF = 1.11 seems reasonable, but GF=1.67 is a rather large value to obtain at ambient relative humidities (your figures indicate typical RHs of 60% for the sea breezes). These large growth factors are not impossible (especially since you are observing significant fractions of marine aerosols), but it would be worthwhile

to discuss these GFs in the context of TDMA measurements found in the literature. Swietlicki et. al. (Tellus 2008, 60B), for instance, provides a nice overview for measurements at 90% RH.

Thank you, it is indeed important to link between the geometric hygroscopic growth factor and thickness of shell that is used in the simulations. The next discussion is added on page 17, after line 2 (initial version):

"Three simplified scenarios are considered: first – the particles are homogeneous, second and third – a liquid water layer coats the particles with a thickness that corresponds to 10 % and 40 % of the total particle radius, respectively. This percentage is assumed because at a thickness of about 10 % the differences in optical characteristics become notable and for about 40 – 50 %, the residual of the fit in the inversion procedure reaches a maximum. This indicates the largest discrepancy between the core-shell model and the particle homogeneity assumption as used in the inversion. To put the percentages used here in the context of real observations, it can be represented in terms of the widely used geometric hygroscopic growth factor, which is the ratio between humidified and dry particle diameter. Thus, 10 % corresponds to a growth factor of 1.11, which can be defined as a low to moderate value, and 40 % corresponds to 1.67, which is near the upper limit of values in the review by Swietlicki et al., (2008), for instance. It is noteworthy that our tests show important differences in optical characteristics and increased residuals of fit also for 30 and 20 % shell thickness. In fact, the effect of the coating also depends on the shape of the particle size distribution and the contrast in refractive indexes of core and shell, therefore, the subject merits some more detailed studies."

+ Level 2 AERONET retrievals do not include retrievals with residuals greater than 5- 8% (depending upon AOT); thus, the 40% coating cases would not make it through the Level 2 AERONET screening, since the residual for that case is 14%. It is important to point this out to the reader, as it demonstrates that AERONET has the ability to omit cases where the aerosol morphology differs drastically from the morphology assumed in the retrieval. This is a much different conclusion than "the retrieval gets it wrong" for such cases.

I appreciate sharing of this thought, it is included in the related section of the manuscript. The phrase in bold is included (p.18, line 21, initial version): "The residual of the fit is quite high, which means that a physical interpretation of the retrieved microphysical parameters should be done with caution. **In addition, retrievals with high residuals are generally screened in final products and therefore cases where the aerosol morphology differs drastically from the morphology assumed in the retrieval algorithms may be omitted.** However, the obtained high fit error show the sensitivity of the measurements to the core-shell structure."

Page 17, line 33: "The retrieved refractive indexes significantly exceed those of the core,..." This is somewhat unintuitive, so it would be worthwhile to explain why this happens in a sentence or two.

The next sentences are added: "It is expected that in the case of mixed aerosol the values of the retrieved refractive index will be in between the refractive indexes of the two components. The fact that the retrieved values are greater and that the size distribution is modified suggests that the inversion algorithm attempts to compensate the specific particle morphology by an exceptional aerosol model."

Minor issues

Page 4: Authors discuss the relationship between the Angstrom exponent and aerosol particle size, without citing the literature. They should provide one or more citations for uninitiated readers.

The citations are added.

Page 10, line 24: "Figures 5c and d..." should refer to Figure 6.

Corrected.

Page 10, line 34: "Also the maximum of the coarse mode..." should be "Also the maximum radius of the coarse mode..."

Corrected.

Page 10, line 34: Replace millimeters (mm) with micrometers (um).

Corrected.

Figure 11, upper right panel: label should be Ca instead of C, right?

Corrected.

Page 14 and Figure 12: There is much discussion about the colored arrows in Figure 12, but I do not see any arrows in my copy.

Corrected. Sorry for this moment of distraction during the image conversion.

Page 15, line 6: What is the wavelength range covered by the SolRad-Net pyranometer?

The wavelength range is 0.3 – 2.8 μm. The information was provided on page 5, line 30. However, the information that is missing is the wavelength range of the calculated solar flux (0.2 – 4 μm). The discrepancy between the spectral range of measurements and calculations should be mentioned because it leads to about 3 % bias. At the same time, this discrepancy is important only for an inter-comparison of the measured and the calculated flux (note that 3 – 5 % is a usual accuracy of the flux measurements), but not for the presented analysis of relative perturbation. It is because the perturbations in the measured and the calculated solar flux are estimated separately. The sentences in bold are added on page 15, lines 9-11: "To evaluate the sea breeze induced radiative effect, we calculated the solar fluxes and the net instantaneous direct aerosol radiative effect using a computational tool described in (Derimian et al., 2016). **Note that the calculated solar flux is for the wavelength range of 0.2 – 4.0 μm, while the measured is for 0.3 – 2.8 μm, which implies about 3 % bias due to the cut-off of the spectral range (note that the accuracy of the measurements themselves is about 3 – 5 % as well). Nevertheless, this discrepancy in the spectral ranges does not affect analysis of the relative perturbation of the solar flux when evaluated using the measurements or the calculations separately.**"

Page 15, line 26: Figures 12c,d should be Figures 13c,d...

Corrected.

Page 16, line 2: The atmospheric radiative effect is related to the SSA, so you could tie this into your earlier discussion of SSA. That is, you could compute the radiative effect using pre-breeze SD and sea breeze refractive indices to estimate the effect of size on the radiative

effect (by comparing to the sea breeze computations that you have already done); likewise, computations utilizing the pre-breeze SD with both pre-breeze and sea-breeze refractive indices can be used to estimate the effect of composition on the radiative effect. I include this item as a "Minor Issue" because it would be a nice addition that will make the paper more interesting, but it is not something that is absolutely necessary for publication.
We thank the Reviewer for this suggestion. This indeed could be an interesting addition. However, there is a problem to distinguish between the effect of the changing microphysics/composition and the changing AOT on the radiative effect. That is, not only the shape of the size distribution will change, but also aerosol volume concentration, which means a change of AOT. In order to isolate the effect of microphysics, the radiative efficiency should be used (radiative effect normalized by AOT). However, a nonlinear dependence exists between aerosol radiative effect and AOT. Even the small effect of this nonlinearity can be comparable with the fine effect of difference in aerosol microphysics. All this makes this type of analysis quite delicate. We therefore, prefer to avoid this complex discussion.

Page 17, line 27: "Note that the refractive index used in the case of homogeneous particles is the same as that of the core." I think that you should move this sentence to the end of the previous paragraph, as I was looking for this information earlier on.
It is moved up.