Author's response to Referee #3

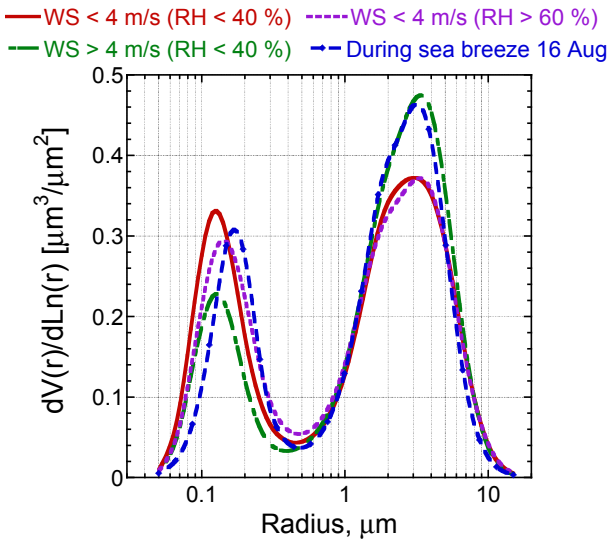
The thoughtful reading and the time dedicated by the reviewer are highly appreciated. The provided major as well as the minor comments are an important feedback that enabled better focusing of the scientific content and improvement of the manuscript quality. Below please find our point-to-point replies.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

Anonymous Referee #3
Received and published: 17 February 2017

Review of 'Effect of sea breeze circulation on aerosol mixing state and radiative prop- erties in a desert setting' by Derimian et al.
The paper studies the modifications in summertime sea breeze conditions of the aerosol compositional, microphysical, optical, and radiative properties at an inland lo- cation of the Negev Desert (Israel). It is well written and easy to read. The study is original and the scientific methodology sound. The complementarity of the inversion of remote sensing (sunphotometer) observations and of the direct, and I suppose time-consuming, off-line individual analysis for characterizing the particles is particularly interesting. The authors evidence for the first time the significant influence of the daytime intrusions of marine air on the aerosol characteristics at such a remote place.
Not only the composition, but also to internal structure of the particles greatly differ in the pre- and post- see breeze situations. In particular, the proportion of mineral (desert-dust) particles surrounded by a coating is unexpectedly large in both cases, which contradicts the common assumption that desert dust is hydrophobic. These modifications of the aerosols characteristics need to be taken into account for quantifying their radiative effect. Finally, the numerical simulations made by the authors show that the current remote sensing inversion algorithms need to be modified in order to take the core-shell structure of the particles into account. My opinion is that this paper deserves publication provided the following concerns are addressed: General comment: After reading the paper, one is left with the impression (see for instance lines 21-23, page 2) that the aerosols initially present at Sede Boker are modified by the arrival of the sea breeze. In fact, these pre-existing aerosols are most probably blown further downwind of the experimental site by the breeze and replaced by new freshly advected particles. The authors make an exhaustive and quite interesting comparison of the characteristics of these two sets of particles, but if the particles are not the same, is it possible to conclude that the size increase observed after the arrival of the sea breeze could be due to the water vapor uptake? More generally, the mineral particles observed during the marine intrusions probably have a long history of coexistence with the other species (sea-salt and anthropogenic aerosols and gases), what's more in humid air-masses. Therefore, they are more liable to have formed internal mixtures than the resident aerosol of Sede Boker.

Agree and reworked the text in order to emphasize that the new air masses arrive and replace the previously existing aerosol. Regarding the question: "…if the particles are not the same, is it possible to conclude that the size increase observed after the arrival of the sea breeze could be due to the water vapor uptake?"

Thanks for this question, indeed, the conclusion should be better focused. It should be emphasized that the water uptake is a hypothesis. This hypothesis is suggested as an explanation of the aerosol size shift based on the findings that: 1) the particles are more hygroscopic and 2) the air contains more water. Thus, the water uptake is suggested as the most probable reason for the aerosol size increase, relative to the background conditions. Of course, other reasons for the size increase are also possible, for example, it can be freshly advected dust. However, the freshly advected large particles are expected to settle quite fast or mostly be present near the surface. The photometric measurements, however, are for the total atmospheric column and are thus not expected to be significantly influenced by the near-surface conditions. In order to support this supposition, we conducted a supplementary analysis by sorting the retrieved size distributions (data for summer 2012) by near the ground measured wind speed and RH. The figure below shows normalized volume size distributions. It shows that the shape of the distribution does not change significantly for low versus high wind speed. However, it does change when the RH is changing; the tendency to the shift of the fine mode is consistent with the size distribution during the sea breeze on August 16th.



It is unlikely that the size shift is due to freshly emitted from the surface dust also because the main increase in the particles size is in the fine mode fraction, which is dominated by mixed dust/marine and pollutants and not by pure dust. The fine mode is more strongly influenced by pollutants and mixed dust/marine aerosol which are highly hygroscopic. It does not, however, diminish the role of the apparent hygroscopicity of dust once reacted with marine particles or pollutants.

In summary, following the reviewer's comment, it is realized that the related discussions and conclusions should be focused better. The text in the new version of the paper is reworked accordingly.

Miscellaneous:

1) P. 7: What can the origin of the Ti-rich particles be?

Ti-rich particles presumably consist of TiO_2. The main source of TiO_2 particles suspended in the atmosphere is most likely from windblown mineral dust (e.g., Chen, H., Nanayakkara, C. E., & Grassian, V. H., Titanium dioxide photocatalysis in atmospheric chemistry. Chemical Reviews, 112(11), 5919-5948, 2012). In our case, Ti was often found mixed with other metallic elements including Fe, Ca, Si and Al that are typical constituents of windblown eroded soils.

On overall, Ti-rich particles accounted for 1.4% of all analyzed particles and occurred in both fine and coarse fractions.

2) P. 8, line 25; on Fig. 4a, the Angström exponent increases with the arrival of the sea breeze on 14 August. Is there a plausible explanation for this exception to the rule?
The Angström exponent is indeed increasing, but only before the sea breeze, at the moment of the sea breeze arrival, it rapidly decreases as in all other cases.

3) P. 10, line 8: the nephelometer 'dries'. . .
Corrected.

4) P.10, lines 12-13: couldn't the 'abrupt response' also be due to the increase of the aerosols concentrations and to a shift in their size?
Yes, the response in optical characteristics is indeed due to the changes in both, concentration and microphysics. The text is clarified as follows: "In summary, all the above mentioned observations of the aerosol optical properties in the solar spectrum and radiation in the thermal infrared wavelength region manifest a coherent abrupt response associated with the sea breeze arrival. An abrupt response in the aerosol optical characteristics can be due to a higher aerosol concentration, but also a change in the aerosol microphysical characteristics and influence of the increasing atmospheric water content. In order to examine the possible change in the aerosol microphysical parameters that take place during the sea breeze, we use the remote sensing observations …"

5) P.10, line 24: This is Fig. 6 (not 5)
Corrected.

6) P.10, line 33: The unit is µm not mm
Corrected.

7) Fig 6c and d : the blue line corresponds to WV larger than
The notation is correct there.

8) P.11; line 5: As said in the following sentences, the large standard deviation does not allow concluding that there is 'a decrease' of the mean real refractive index. I would remove the 'Curiously a decrease. . .'.
Please note that, as suggested by Reviewer #2, the standard deviations are replaced by standard errors, which are more appropriate to the discussion about significance of the measurements. The standard errors do not overlap the mean in the case of the real part. The corresponding text was rewritten.

9) P.11; line 31 and the notations of Fig. 8: Usually, PM1 and PM2.5 correspond to particles with diameters smaller than 1 and 2.5µm, respectively. Here, PM1 corresponds to particles with diameters between 1 and 2.5, and PM2.5 to the range 2.5-10µm. This is confusing.
Agree, the notation in Fig. 8 should correspond to the aerodynamic cut-off diameter of the impactor and not to the conventional PM. It is corrected to PM1-2.5 and PM2.5-10 in Fig. 8 and corresponds now to the explanations in the text, a clarification is also added in the figure caption.

10) P. 12, lines 15-16: the 'other' particles represent 7% of the coarse fraction but are said to be smaller than 1µm in diameter. Isn't this contradictory?
There is no contradiction in the data, but a confusion due to a difference in the measurements techniques. These particles were analyzed by SEM/EDX and found on the PM2.5-10 stage of the impactor. Because 2.5 µm is a cut-off diameter at 50 % of collection efficiency, particles smaller than 2.5 µm can also be present on this stage (the size-segregated sampling by cascade impaction is based on an aerodynamic cut-off diameter and depends on particle density). The diameter obtained by SEM observation is derived from particle's 2D-projection and corresponds to the geometric diameter of the equivalent circle area. Depending on the density, the aqueous content and morphology of ambient particles, it is possible that the SEM technique will provide geometric diameter smaller or bigger than the aerodynamic cut-off diameter of the impactor. A clarification is added in the section about aerosol sampling methodology (3.3.2).

11) P. 13, line 1: the authors say that the shift towards larger sizes of the marine particles during the sea breeze could be due to hygroscopic growth. Aren't the SEM observations made under a vacuum, i.e. in dry conditions? Moreover, if we go back to my first comment, please consider that the marine particles observed at the inland site before the arrival of the sea breeze might be more aged than the new ones. Consequently, their size-distribution might have been modified by the size-selective dry deposition process. For instance, I cannot help observing on Fig 9 that the very fine and the coarse particles present in the fresh marine air-masses (Fig. 9 b) have disappeared on Fig 9a, and that on the latter figure, only the particles with a diameter corresponding to the smallest deposition velocity (around 1µm) subsist.
Cascade impaction is conducted at ambient temperature and RH, and a dry size distribution is derived following the SEM observation. Indeed, the SEM observations were performed under high vacuum and water-solvated ions dehydrate in the SEM chamber, but due to the wettability of the substrate, initially hydrated sea salt particles appear generally in SEM pictures as flat particles (larger than thick) and often as rounded shape for aged marine particles, consisting of a core and a shell formed by residues as shown in Figure 10d. Thus, the geometric size of marine particles given by SEM may be slightly underestimated due to the low thickness of the border of the dehydrated particle but not as much as if marine particles were dehydrated before sampling. That is why the shift towards larger sizes of the marine particles during the sea breeze could be due to hygroscopic growth.
However, we also agree with the referee on the possibility of a shift towards larger marine particles of the coarse mode during sea breeze due to size-selective dry deposition processes that can occur during the aerosol transport. Thus, the text was modified considering this hypothesis (page 13, line 1): "This can be due to hygroscopic growth caused by the higher RH during the sea breeze, but size-selective dry deposition processes (Seinfeld and Pandis, 1998) can also take place during the aerosol transport since the wind speed and atmospheric residence time of marine particles are different before and during the sea breeze."

12) P. 13, line 24: There is no Mg in the composition of the calcite.
Mg is referring to dolomite. The text is modified. Both dolomite and calcite are minerals containing calcium carbonate. Dolomite differs from calcite because of the presence of magnesium (i.e., calcite ($CaCO_3$) mainly contains calcium carbonate and dolomite $CaMg(CO_3)_2$ contains calcium magnesium carbonate).

13) Fig. 11: On the upper right-hand panel, this should be Ca (not C).
Corrected.

14) Fig. 12: I cannot see the arrows mentioned on page 14.
Corrected. Sorry for this lack of attention during the image conversion.

15) P. 15, line 19: In the reduction of 5%, what are the respective shares of the 1) aerosol changes and 2) WV increase?
Thank you for this question. It is indeed important to distinguish between the effect of water vapor and aerosol. Supplementary calculations were conducted and the next discussion is added to the manuscript:
"… This amounts to 4.6 % reduction of the total solar flux that would reach the surface without the sea breeze effect. It should be realized, however, that the reduction in the solar flux is not only due to the change in aerosol properties, but also due to the increase in the water vapor content. In order to estimate the role of each component, additional calculations were conducted assuming that only the increase in the water vapor takes place, and then, assuming that only the aerosol properties change. The results show that the increase in the water vapor (from 1.62 to 2.13 g cm^{-2}) is responsible for a loss of 7.5 W m^{-2} in the solar flux reaching the surface, while the change in the aerosol properties is responsible for 15.5 W m^{-2} of the total 23 W m^{-2} difference, which amounts to 1.5 and 3.1 %, respectively."

16) P. 15, line 26: this should be Fig 13, not 12.
Corrected.

17) Section 7: please, consider reformulating the whole section. It is much harder to follow than the rest of the paper. For instance, the reader discovers only on page 17 that forward calculations have been made (and with which inputs), then that different scenarios have been considered for inversion simulations.
Thank you for this feedback, the section was reorganized and some parts reformulated.

18) P.19, lines 2-5: could you be more specific regarding implications for the satellite and LIDAR inversions?
The sentences were modified as follows: "…we can also conclude that including backward scattering angles and polarimetric measurements present more sensitivity to the core-shell structure. This is because the main differences, due to the aerosol core-shell structure, are observed in the angular and polarimetric characteristics of the backward scattered light. Thus, since the backward scattering is a primary signal measured by satellites and LIDAR, important implications for these types of measurements are possible. For example, the aerosol core-shell structure will affect the lidar ratio and a parameterized core-shell aerosol model can be considered in satellites retrievals."