This paper comprehensively reports the results of optical properties measurements made as part of the CARES campaign. This is a very weighty paper and while the conclusions could really be regarded as game-changing, the high quality of the measurements and the depth of the analysis means that these results are still very important and relevant to ACP, having a wide range of potential applications in radiative transfer and the interpretation of remote sensing data. I would recommend that this be published subject to minor corrections.

General: I found the various combinations of sampling conditions referred to a bit be-
wildering at times. It would greatly help the reader if a schematic figure could be given showing the different sampling arrangements for the different sites. On a related note, I also found the large number of mathematical symbols to be a bit confusing at times, so it would be useful to compile these as a table.

Line 122: The method used to humidify the sample flow to 85%, including the methods to monitor and control the humidity, should be described here.

Line 128: A reference should be supplied for the mobility conversion for the aerodynamic diameters. Rather than assume sphericity (which is probably not valid for dust and dry sea salt particles), it would probably be more correct to refer to the assumed 2 g/cc density as the ‘effective’ density.

Line 138: These mobility diameters should be qualified as ‘approximate’ because they have each been clearly rounded off compared to the actual theoretical values.

Line 166: The factor of 1.66 is contrary to the factor of 1.33 recommended by Laborde et al. (2012b), so the nature of the ‘personal communication’ cited may need expanding on here.

Line 233: A reference should be included here because the exact effect BrC has on AAE is by no means certain.

Line 238: Similarly, include references for examples of how SAE is ‘commonly used’.

Line 384: What AAE would be necessary to cancel out any influence from BrC? How does this compare with DOI:10.1002/2014GL062443?

Line 402: A more fundamental reason the PALMS and SPLAT II are not capable of quantifying contributions from BC internally mixed on dust particles is the matrix effects associated with each instrument, such that the mass fractions reported on individual particles are not quantitative.

Line 453: Shouldn’t the qualitative influence of sea salt particles be evident in the single
particle mass spectrometer data?

Line 514 (and elsewhere): “consistent with that of (Russell et al., 2010)” should read “consistent with that of Russell et al. (2010)”

Line 531: This opening statement should be made more descriptive, as it isn’t specified what exactly the effect on climate is. It may also be worth mentioning that this is an important parameter for remote sensing retrievals.

Line 573: Was any correction for thermophoretic losses invoked? If not, the authors should comment on how much of an issue they believe this to be.

Line 593: According to conventional wisdom, supermicron sulphate and nitrate tends to be in the form of salts of calcium and sodium rather than ammonium, which makes them quite involatile, so I would doubt that this is significant.

Line 599: A related hypothesis could be that undenuded sea salt particles do not completely effloresce during drying due to the presence of magnesium salts (doi: 10.5194/acp-15-11273-2015) and organics. If the denuded particles are more completely dried out (due to the water being boiled off) then this would increase their apparent hygroscopicity further.

Line 620: How does this fit in with the conclusions of Doi:10.1038/Ngeo2220?

Line 646: The caveat should be added that this is assuming that the particles have not acquired an involatile coating (e.g. coagulation with sea salt, condensation of humic-like SOA), because this could confound any attempt to isolate the effect of morphological changes on SSA.

Line 653: It should be noted that if the particles are thought to be very non-spherical, the SMPS sizing is likely to be overestimated to a large degree

Line 653: If the particles are fundamentally different to ‘normal’ black carbon particles, then the SP2’s calibration could be invalid and the equivalent core sizes reported
inaccurate. If the instrument had a narrow band incandescence detector, it may be informative to compare the ratio of this to the broadband detector to see if the apparent colour temperature had changed.

Figure S1: Adjust the colour scale so that areas covered by land are green rather than blue.

Figure S9: Given that Babs of black carbon is more closely related to mass than number, a mass-weighted distribution comparison would be informative here.