Reply on RC1
Bastiaan van Diedenhoven et al.

We thank the reviewer for the positive feedback and helpful comments. Comments are repeated below in italic script, followed by our responses.

Line 150: The authors assumed the size distributions of the insoluble component and the dry soluble component of the mixture are the same. Is there any back up reference?

Little information is available. We added a discussion on this assumption and a sensitivity study at line 183 and further. In the discussion we include the sentence:

“Some observations, such as from Holmgren et al. (2014) and Kim et al. (2020), suggest that hygroscopicity increases with particle size.”

Please also see our reply to comment 5.

Line 254: Why only data over ocean are used in this manuscript? Please specify.

To specify we added the following to the text at line 275:

“The majority of the data during these campaigns were collected over ocean. Furthermore, the RSP aerosol retrieval algorithm (section 3.2) is currently limited to ocean surfaces. Hence, only data over ocean are used here.”

Section 4.1: The Figure 5 showed that the water volume fractions derived from the retrieved refractive indices show a larger peak at zero for all three campaigns, corresponding to κ = 0. Why the retrieved aerosol hygroscopicity can equal to zero? As I understand, even the even the water-insoluble particles will be somewhat hygroscopic rather than absolutely not hygroscopic.

Petters and Kreidenweis (2007) define kappa as equal or greater than 0 (e.g., see their Fig 1). Note that these insoluble particles can be wettable at supersaturation above 1, as
discussed by Petters & Kreidenweis: “As kappa approaches zero, the particle becomes nonhygroscopic and the slope approaches that expected for an insoluble but wettable particle as predicted by the Kelvin equation, i.e. –1.”

Since we already refer to Petters and Kreidenweis (2007) for our definition of kappa, no changes were made to the manuscript related to this comment.

Line 448: Why is there no difference in the retrieved results assuming different f values? This is confused. Because the solube aerosol fraction has lagre effect on aerosol hygroscopicity and thus influence particle size distribution.

The reviewer is correct that soluble fraction has a large effect on the aerosol growth as a function of relative humidity. However, note that we derive dry effective radius and variance from the ambient values using the simulated relationship between r_e and v_e and water fraction in combination with the retrieved water fraction. Figure 1 shows that the relationship between r_e and water fraction is rather similar for f_sol values >0.05. The relationship between v_e and water fraction varies somewhat more with f_sol, but this variation is still limited for f_sol between about 0.1 and 0.7.

To further clarify, we added the following to the text at line 473:

"Since the variation of re,mix and ve,mix with fw vary relatively weakly for 0.1 < fsol < 0.6 (Fig. 1), no noticeable difference in the histogram is seen if a different value fsol is assumed within this range."

Section 5: Same above. the authors assume that the aerosol consists of externally mixed soluble (κ > 0) and insoluble (κ = 0) components with equal dry size distributions. This will lead to large uncertainty in retrieved results. I suggest the uncertainty analysis about this assumption are needed.

According to the reviewer’s suggestion we added a sensitivity study to the supplement (section S2) and summarize the results in the main text. The following was added to the text starting at line 183 and further:

“To derive Eqs. 18–21, we assumed that the size distributions of the insoluble component and the dry soluble component of the mixture are the same. Some observations, such as from Holmgren et al. (2014) and Kim et al. (2020), suggest that hygroscopicity increases with particle size. To test the impact on assuming the same dry particle distribution of insoluble and soluble aerosols, we simulated re,mix and ve,mix as a function of fsol for a range of fw and for an aerosol with a soluble component with re,dry = 0.15 μm and an insoluble component that is Δre smaller. For both components we assume ve,dry = 0.15. Using these simulations as our dataset, we subsequently infer fsol, re,dry and ve,dry while assuming an aerosol mixture with equal soluble and insoluble components. The differences between the inferred fsol, re,dry and ve,dry and the true values in the simulations yield an estimated sensitivity to Δre. Detailed results are given in the Supplement. For a Δre of 0.03 μm, maximum underestimations of fsol, re,dry and ve,dry are 0.23, 9% and 17%, respectively, occurring at true fsol values of 0.65, 0.28 and 0.16, respectively. These biases scale approximately linearly with Δre. Hence, especially retrieved fsol and ve,dry are quite sensitive to our assumption of the same dry particle distribution of insoluble and soluble aerosols. However, a realistic estimate of the range of Δre is not available.”