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Interactive comment on “Measurements of size-resolved hygroscopicity in the California coastal zone” by D. A. Hegg et al.

Anonymous Referee #3

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General Comments

The manuscript describes a significant difference in the aerosol hygroscopicity at different altitudes in the marine boundary layer off the coast of southern California. Based on the chemical composition data, oxidation of organic films is suggested as the cause of the difference. Further, the contributions of different aerosol types/sources are assessed using receptor models. The observed large difference in the aerosol hygroscopicity at two altitudes is surprising, which is an important finding if it is real. Its relevance to the organic composition is not assessed in a fully statistical manner (e.g., correlation coefficient for individual data points), yet at least the composition data presented in this manuscript (averages of the bulk composition and two examples of the size-resolved composition) suggest the relation of organics to the difference in hygro-

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scopicity. The oxidation of organic films is, however, suggested without sufficient evidence. The receptor model successfully distinguishes possible sources of aerosols, whereas the essential meaning of the closure of DHGFs is not very clear.

While the manuscript presents interesting results, it needs major revisions based on the above comments and more specific comments listed below.

Specific Comments

Pages 10536-10538, Sect. 4.1 and Figs. 2, 4 and 6: It is reasonable to infer that organics, although the total amount is not quantified, were related to the observed difference in the aerosol hygroscopicity with altitude. However, it is not clear why only the oxidation of insoluble organics is discussed as the cause of the possible difference in the organic composition. Why is the possibility of different relative contributions of organic aerosols from different sources at two heights not discussed? If it is not realistic because of vertical mixing, is it realistic that the oxidized organics are not mixed vertically during their residence time in the marine boundary layer? These points may need to be addressed in the manuscript.

Page 10536, lines 17-28: Aerosol hygroscopicity is directly related to the relative abundance of chemical components in the particles, rather than to the absolute mass concentrations. (The explanation in lines 24-28 may be based on this theory.) Therefore, it is more appropriate that the mass fractions of water-soluble species in aerosols are presented in Fig. 2. This is also the case for Figs. 4 and 6. Even if the total dry mass is not available for the data in Figs. 4 and 6, the importance of mass fractions should be addressed so that the readers correctly interpret the figures.

Page 10537, line 10: Although there are several studies suggesting the "coating" of marine aerosol particles by fatty acids, my opinion is that it is not fully proven but is still a hypothesis.

Page 10538, lines 9-17: The discussion on the oxidation of organic films is some-

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what confusing. Theoretically, the particle hygroscopicity is governed by the number of soluble ions/molecules in the particle, and thus a substantial amount of water-soluble organic ions/molecules needs to be formed to account for the observed large increase in the particle hygroscopicity with altitude. The explanation on the oxidation of thin organic films with little mass contradicts the theory of particle hygroscopicity.

Page 10543, 2nd paragraph and Fig. 9: The authors use variables calculated from aerosol hygroscopicity ($H_2O_{\text{submicron}}$, $H_2O_{\text{supermicron}}$, and $H_2O_{\text{scattering}}$) in the receptor model (Table 1), assign them to three component loadings (Table 2), and then reconstruct the hygroscopicity (Fig. 9). In this case, isn't it trivial that some agreement is found between predicted and measured DHGFs because aerosol hygroscopicity is a part of the variables in the receptor model? The essential importance of the analysis in Fig. 9 needs to be clarified.

Technical Corrections

Page 10534, line 5: Hegg et al. (2003) is not given in the reference list.

Table 2: The abbreviations in Tables 1 and 2 should be harmonized.

Fig. 2: cl → Cl, so4 → SO_4 , na → Na, no3 → NO_3 , k → K, mg → Mg, bc → BC

Figs. 3, 5 and 7: The unit of the particle diameter seems incorrect.

Figs. 4 and 6: cl → Cl, so4 → SO_4

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