Long-term aerosol optical hygroscopicity study at the ACTRIS SIRTA observatory: synergy between ceilometer and in-situ measurements (acp-2019-12)

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Author's response

We thank the anonymous reviewer for his/her comments and suggestions that have helped to improve the quality of the manuscript. According to the referees' reports, the following changes has been performed on the original manuscript and a point-by-point response is included below, where blue colour is related with answers for referee#1 and red colour for referee#2.

Answers to Referee#2:

Specific comments

1. **Introduction: Literature in which they examined the association between lidar- derived aerosol hygroscopic properties and in situ aerosol chemical composition should also appear in the introduction. Some references are given later in the manuscript (ex, zhang et al. 2015), but they could already appear in the introduction. See also: Lv et al., Hygroscopic growth of atmospheric**

aerosol particles based on lidar, radiosonde, and in situ measurements: case studies from the Xinzhou field campaign, J. Quant. Spectrosc. Ra., 2017.

Following the reviewer's suggestion, these references are now included in the new version of the manuscript:

P02, L29-32: **"**Finally, fewer studies have been performed by crossing information between remote sensing and in situ setups. In Lv et al., (2017) these synergies present interesting approaches for comparing chemical concentrations with hygroscopic growth properties mainly retrieved from lidar and radiosondes vertical cases studied."

2. **Check and revise all the equations carefully! Some comments here:**
   - **Use of ≡ or = for equation?**
     We will use "=".
   - **P4,L26: check the equation numbering**
     Done
   - **In eq1, z1,z2 are used as variable of T, so it should be T(z1,z2) instead of T(z). Or you can just use z1 = 0, z2 =z.**
     From now, we use T (z_1, z_2).
   - **Explain td before equation 7. You used "time window", mention it here.**
     Done in P06, L15: **"**where t_d refers to the dry state of the aerosols within the temporal window of evaluation**"**
   - **Eq7, I suggest not use "zref" here, the z used in your study is a fixed height of 30m which is not a "reference height", as you used RHref for the calculation, it could be a bit confusing. Or you should mention the zref at page 6, line 5.**
     We added in P06, L13: "RH changes at 30 m a.g.l (z_{ref})."
   - **Eq13 only give the expression of nwv(t)**
     Done

3. **Section 3.1. There is no really new methodologies proposed, too many equations (11 equations) in this section, they can be simplified. Ex, Eq8 no need**

We have reduced the number of Equations to 9, combining some of them.

4. **Section 3 and 4 can be one section of methodology.**

As it was proposed by the reviewer, we have modified the title of section 4, becoming it a part of section 3. This modification can be found in P08, L24: "3.3. Aerosol hygroscopic optical enhancement identification"

5. **Supplement: It seems that this supplement is related to your previous version of manuscript, please update it (e.g., the cross-reference).**

Now the Supplement has been carefully checked and the some references have been deleted.

6. **No references cited in the text, whereas you have a reference list in the supplement.**

This aspect has been corrected and the supplement has been accordingly updated.

7. **"From now, we will use beta instead of betaatt for simplicity", but you forgot to mention it in the manuscript."**

Now this is mentioned in P07, L19-20: "Therefore, in the manuscript, we will assume hereafter that $f_{\beta^{att}}(RH) \cong f_{\beta}(RH)$; therefore, β^{att} will be treated as β from this point forward."

8. **It would be better to make the table and figure captions directly with the tables and figures.**

Done

Minor comments:

1. **"Hänel" not Hännel. Also "Hänel parameterization" not Hännel or Hannel, please change them all.**

Done

2. **Make it clear when you use aerosol backscatter or attenuated backscatter coefficient throughout the paper.**

Done

3. **In situ or in-situ, water vapor or water vapour.**

Done

4. **P2, L16-18, introduce "enhancement factor" before the description of its magnitude.**

Done, P02, L19-20

5. **P3, L29, V-Trafic report, 2014 is not in the reference**
   According to the reviewer suggestion, this reference have been deleted, because it is a local information but nothing to be referenced.

6. **P4, L6, are the uncertainties mentioned here for raw data?**
   Yes, these are the uncertainties for the raw data measured by the instrumentation. In P08, L13: "The error associated with γ was calculated using the Monte Carlo technique, modeling raw measurements of β and RH as normal distributions and β_{ref} and RH_{ref} as the respective values calculated for each case and finally assuming the error as the mean standard deviation of all simulations"

7. **P7, L7, q(td)-q(d) if you keep using td.**
   Done

8. **P8, L22, "the Hännel parameterization (Eq. 9)", it is not eq9**
   Done

9. **P8, L25, ii rephrase the sentence**
   Following the reviewer's suggestion, we rephrase the sentence as follows:

i. P09, L10: "ΔRH > 30 %, where ΔRH refers to the difference between final and initial RH within the temporal window under study. This criteria is used to apply the Hänel parameterization over a sufficient RH-range."

10. **P9, L20, some introduction here will be better**

    This section was restructured in order to follow the suggestions from both reviewers:

P10, L1 to P11, L3:

"As an example of the methodology implementation, this section shows two of the final eight hygroscopic growth cases found in this study (Fig. 2 and Fig. 3). These examples correspond to 25 June 2013 from 07:17 to 10:17 UTC (case 3) and 17 May 2016 from 07:40 to 10:40 UTC (case 8). Fig. 2a and Fig. 3a present the time evolution of β, T, RH, q, W_s, W_d, dew point temperature T_d, and 1-h averaged aerosol chemical composition (BC, OA, NH_4^+, NO_3^- and SO_4^{2-}). Figure 2b and Fig. 3b show $f_\beta(RH)$ and $f_{PM1}(RH)$, and Fig. 2c and Fig. 3c contain a pie chart with the mean contribution of each chemical compound during the hygroscopic event. These cases were selected to show two different situations found in this study (the other six cases are shown in Figures S5-S10 of the supplementary material).

Case 3 presents lower value of hygroscopicity parameter, with values of $\gamma = 0.5 \pm 0.4$ and $f_\beta\ (RH = 85\ \%) = 1.7 \pm 0.2$. During case 3, the predominant wind direction was NW with relatively low wind speed ($W_s = 2.5$ m/s) and some variability up to ΔW_s =24.5 % and $\Delta W_d = 33.9$ %, and

the chemical composition was relatively constant in most compounds over the time window studied. The average chemical composition (Fig. 2c) indicated a high contribution of OA (58 %) and BC (17 %) particles, and the total aerosol mass (PM_1) was almost constant during the hygroscopic case (from 7:15 to 10:15 UTC), showing no correlation with RH. The relative high presence of BC and OA (less hygroscopic compounds) may reduce the hygroscopicity properties. These findings are consistent with results from rural and suburban sites presented by Chen et al. (2014) in Wuqing (China), Zieger et al. (2014) in Melpitz (Germany) and Titos et al. (2014a) in Granada (Spain), where low value of hygroscopicity parameters were observed due to high contribution of OA and BC. A detailed discussion of the origin of the air masses will be given in Sec. 4.2.

Case 8 presents predominant westerly wind with a relatively high mean wind speed (5 m/s) and low variability in both wind speed and wind direction (ΔW_s =20.7 % and ΔW_d = 6.4 %), and a slight increase in PM_1 with RH was observed (Fig. 3b). However, the enhancement of β is significantly higher with respect to the variation in PM_1. In fact, the RI remains within the selected range (RI= 0.60), denoting that most of the increase in the attenuated backscatter coefficient can be attributed to hygroscopic growth. The chemical composition during case 8 shows a predominance of OA (46 %) but also with important contribution of secondary inorganic compounds SO_4^{2-}(19 %) and NH_4^+(12 %), which are highly hygroscopic, and low contribution of BC (8 %). This case exhibited higher aerosol hygroscopic properties than case 3 with $\gamma = 0.9 \pm 0.6$ and f_β $(RH = 85\ \%) = 2.5 \pm 0.3$. This behavior might be linked to the lower contribution of OA and BC and higher contribution of inorganic aerosols (IA). Studies performed close to the SIRTA site by Randriamiarisoa et al. (2006) at Saclay (France) report a high $\gamma = 1.04$ and f_σ $(RH = 80\ \%) \sim 2.0$ linked to a low contribution of OA and high IA contribution associated with anthropogenic and marine aerosols. A more in-depth description of this case will be given in the following sections."

11. **P9, L22, in the text, 07:15 to 10:15 UTC, but in figure 07:17 to 10:17 UTC, check.**
    The correct one is 07:17 to 10:17 UTC, and it was changed in the manuscript in P10, L5.

12. **P9, L23 is beta here correspond to attenuated backscatter coefficient?**
    Yes, this beta corresponds to attenuated backscatter coefficient according to the statement set in P7, L19-20.

13. **P9, L27 "high contribution of OA (58 %) and SO4 2-(15 %)," for case 8 the contribution of so4 2- is higher than case 3, bus in case 3 there is higher contribution of BC,**
    According to the reviewer's suggestion, this section has been restructured, in addition the discussion was modified clarifying the inconsistent statements that reviewer highlighted. The new discussion section can be found in P10, L1 to P11, L3:

14. P9, L28 mention that the r here is for beta not for PM1

According to the reviewer suggestion the following phase was added in P10, L15: "Case 3 presents lower value of hygroscopicity parameter, with values of $\gamma = 0.5 \pm 0.4$ and f_{β} $(RH = 85\ \%) = 1.7 \pm 0.2$."

15. P9, L29 change 14% to 12%

This section has been restructured, therefore this line was suppressed.

16. P10, L19 do you mean "than case 3"?

Yes, it is correct, case 3.

17. P11, L23-24 Change the expression "beta"

Done

18. P11, L30 explain sp here, even though with the definition in section 3.1.

"sp" refers to scattering coefficient. This clarification has been included in P12, L14-16: "in situ measurements are performed by controlling RH (starting mostly from RH_{ref}=40 %) and uses σ_{sp} (scattering coefficient) as the optical property"

19. P11, L31 please rephrase the sentence

The phase has been rephrased in P12, L13-15 "…in situ measurements are performed by controlling RH (starting mostly from RH_{ref}=40%) and it uses σ_{sp} (scattering coefficient) as the optical property."

20. P12, L4 table1 do not have information about what you discussed here

The reviewer is right. Therefore, the reference to Table 1 has been deleted

21. P13, L12 "4.5 years" dataset

Done in P14, L6.

22. Figure 1, it would be nice to introduce the in-situ monitoring station here, as fig1a and fig1b

Thank you for your suggestion, Fig.1b it was added to introduce the in-situ station.

23. Figure2, specify the beta

Done

24. Table 1, please specify the RHref value, also specify the beta

Done

25. Table2, check caption and the table content, to be consistent.

Done

26. Fig S5, change the value-range (y-axis) of the wind speed

Done