Supplement of

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

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1 Effect of beta-attenuated water vapor correction on calculated $f_\beta(RH)$ and $\gamma$ parameter

The $\beta_{att}$ signal presents a dependency on water vapor absorption as shown in Sec. 3.1 (Eq.8), associated with the wavelength emission. This dependency may cause direct effects over calculations retrieved by using $\beta_{att}$. Here, we present the correction applied to $\beta_{att}$ and the effects on $f_\beta(RH)$ and $\gamma$ calculations. Figure S1 (left panels) represents the temporal-evolution of $\beta_{att}$, beta attenuated water vapor corrected ($\beta_{att}^{wv}$) signals over 3 h time-window, together with the temporal-evolution of $q$. Figure S1 (right panels) shows the biases for beta ($bias_\beta$) and $\Delta q$ obtained.

The quantifications are performed by means of the bias$_\beta$ ($\beta_{att} - \beta_{att}^{wv}$) and $\Delta q$ ($q(t) - q(t_d)$) calculations. Fig. S1ac presents two cases (case 1 and case 2) with low absolute-differences in $q$, which produces slight changes in $\beta_{att}^{wv}$ signal respect to $\beta_{att}$. On the other hand, Fig. S1eg (case 3 and case 4) show that high absolute-differences in $q$ are linked to high changes on $\beta_{att}^{wv}$. The right side of the panel (Fig. 1Sbd, case 1 and case 2), presents the bias quantification, showing that low bias$_\beta$ are associated with low $\Delta q$ and, on the contrary, Fig. 1Sfh (case 3 and case 4), show that increases in $\Delta q$ makes that bias$_\beta$ becomes higher. This analysis leads us to conclude that no-water vapor correction will produce an overestimation of the total $\beta$ signal, being $\Delta q$ the parameter that rules the $\beta$ correction. From now, we will use $\beta$ instead of $\beta_{att}$ for simplicity.

To see the effect of $\beta_{att}^{wv}$ on $f_\beta(RH)$, we applied the Hänel parameterization (Eq. 7, Sec. 3.1 of the manuscript) to the same 4 cases studied above. Figure S2 presents the $f_\beta(RH)$ and the enhancement factor water vapor corrected, $f_\beta^{wv}(RH)$. The results reinforce those obtained above (Fig S1), where low/high changes in $\Delta q$ are linked with low/high bias$_\beta$ and, on this way, this would affect $f_\beta(RH)$ calculation. Additionally, the water vapor correction tends to decrease $\gamma$ ($\beta_{att}^{wv}$ is lower than $\beta$). Therefore, cases with lower bias$_\beta$ and $\Delta q$ (case 1 and case 2), exhibit lower bias$_\gamma$ (0.02 and 0.05, respectively), meanwhile on case 3 and case 4 instead bias$_\beta$ increase, the bias$_\gamma$ becomes higher (0.11 and 0.09, respectively).

Once we applied the phase 2 of the methodology (Sec. 5), we obtain 94 hygroscopic potential cases for 3h time-window (Fig. S3a), 9 cases for 4h time-window (Fig. S3a) and 4 cases for 5h time-window (Fig. S3a), resulting in a total of 107 cases. To establish a bias error for this hygroscopic study, we have calculated the median of the bias$_\beta$ and $\Delta q$, highlighting two main aspects: (i) The median bias$_\beta$ follows the median $\Delta q$ variability, remarking their dependency. This fact is well seen from the scatter plot (Fig. 3Sb), where these variables show a positive correlation, however the correlation coefficient is not too high ($R^2= 0.61$), mainly because the data dispersion increases for bias$_\beta > 1.5 \times 10^{-7}$ m$^{-1}$sr$^{-1}$ and $\Delta q > 3.0$ g/m$^3$; (ii) The mean bias error calculated for $\beta_{att}^{wv}$ over the 107 potential cases evaluated is lower than $2.5 \times 10^{-7}$ m$^{-1}$sr$^{-1}$ and the mean $\Delta q$ is lower than 5.5 g/m$^3$. 
Fig. S4 quantifies the effect of the $\beta_{\text{att}}^{\text{tot}}$ over $f_\beta(RH)$ and $\gamma$ hygroscopic properties, by means of the median bias$_{f_\beta(RH)}$ ($f_\beta(RH) - f_{\beta_{\text{att}}}(RHI)$) and the bias $\gamma$ ($\gamma_{\beta_{\text{att}}} - \gamma_{\beta_{\text{sat}}}$). Figure S4 reveals the no-correlation between bias$_\beta$ and bias$_{f_\beta(RH)}$. However, combining the results from Fig. S3 and Fig. S4, it is possible to establish that bias$_\beta > 1$ m$^{-1}$sr$^{-1}$ would cause an increment of bias$_{f_\beta(RH)}$ above 0.2, increasing the error on $f_\beta(RH)$. Finally, it was obtained that bias$_{f_\beta(RH)}$ and bias $\gamma$ for the whole study were lower than 0.3.

2 Results
2.1 Methodology applied to eight aerosol hygroscopic growth cases

The 8 hygroscopic growth cases reported in this study (Table 1, Sec. 5.2) were found at daytime in the early morning, with $RHI_{\text{ref}}$ around 50 % and the maximum RH reached was up to 90% over 3h time-window. For cases 1, 2, 4, 5, 6 and 7 the perceptual composition was dominated by OA with values up to 50 %, except on case 7 where OA decreased up to 38%. The BC concentration was relatively low almost for all cases found (close to 6 %). The concentration of inorganic compounds were dominated by $SO_4^{2-}$ (lower than 21 %) and $NH_4^+$ (lower than 19 %), however $NO_3^-$ reached values of 21% on case 6. The air masses that come mainly from W-NW direction are related to case 1 ($\Delta u=3.6$ m/s W), case 2 ($\Delta u=23.0$ m/s NW) and case 6 ($\Delta u=4.4$ m/s W), with speed variability up to 14.2 %, 20.7 % and 18.5 %, respectively; and the air masses that coming from W-SW direction at low wind direction variability are associated with case 4 ($\Delta u=2.7$ m/s W), case 5 ($\Delta u=2.4$ m/s SW), and case 7 ($\Delta u=1.7$ m/s SW), and wind speed variability about 15.4 m/s, 20.4 m/s and 10.9 m/s, respectively. All these cases fulfilled the threshold established for $\frac{\Delta F_{PM1(RH)}}{\Delta f_\beta(RH)} < 0.5$ indicating that increases/decreases in $f_\beta(RH)$ are not related with advected aerosol into the atmospheric volume studied. The Hänel parameterization is calculated for both $f_\beta(RH)$ and $f_{PM1}(RH)$ (see panel Fig. S4 to S9 b.). The hygroscopicity properties of the 6 cases presented here were evaluated and compared against literature in the Sec. 5.2 of the article.
Figure S1. Time evolution of $\beta_{att}$ (blue line), $\beta_{catt}$ (red line) and q (orange line) [left panels (a,c,e,g) bias (black line) and $\Delta q$ (green line) [right panel (b,d,f,h)].
Figure S2. Experimental data points (blue/red dots) and Hänel parameterization (blue/red lines). Case 1 and Case 2 show the effect of the lower bias $\beta$ and $\Delta q$ differences over $f_{\beta \text{att}}(RH)$ and $f_\beta(RH)$. Case 3 and Case 4 present the effect of the higher bias $\beta$ and $\Delta q$ differences over $f_{\beta \text{att}}(RH)$ and $f_\beta(RH)$.
Figure S3. Median of bias$_\beta$ and $\Delta_q$ for all potential cases of hygroscopic growth found from 2012 to 2016 at the ACTRS SIRTA observatory: (a) median of bias$_\beta$ and $\Delta_q$ to 3h time-window analysis (green bars), 4h time-window analysis (orange bars) and 5h time-window analysis (blue bars); (b) scatter plot correlating median of bias$_\beta$ and $\Delta_q$ for whole time-windows.
Figure S4. $bias_{f(RH)}$ and $bias_{\gamma}$ for all potential cases of hygroscopic growth found from 2012 to 2016 at the ACTRIS SIRTA observatory. $bias_{f(RH)}$ and $bias_{\gamma}$ for 3h time-window analysis (green bars), 4h time-window analysis (orange bars) and 5h time-window analysis (blue bars).
Figure S5. Criterion for data selection: case 1 on 29 July 2012 from 06:30 to 09:30 UTC.
**Figure S6.** Criterion for data selection: case 2 on 02 September 2012 from 10:30 to 13:30 UTC.
**Figure S7.** Criterion for data selection: case 4 on 28 July 2014 from 09:10 to 12:10 UTC.
Figure S8. Criterion for data selection: case 5 on 17 August 2014 from 06:40 to 09:40 UTC.
Figure S9. Criterion for data selection: case 6 on 21 May 2015 from 06:15 to 09:15 UTC.
Figure S10. Criterion for data selection: case 7 on 15 April 2016 from 07:05 to 10:05 UTC.