Response to Referee #1 Comments

Characterisation of hygroscopic aerosol processes using remote sensing and in situ techniques at the MeteoSwiss Payerne station

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Reviewer comment

Overall, this manuscript presents a valuable and comprehensive dataset on aerosol hygroscopicity from the HYGRO19 campaign, combining Raman lidar, COBALD-RS41 balloon measurements, tandem nephelometry, and complementary aerosol observations. The large number of hygroscopic aerosol layers retrieved from RALMO is particularly valuable and demonstrates the potential of remote-sensing observations to provide vertically resolved information on aerosol hygroscopicity under ambient atmospheric conditions. The comparison with independent in situ techniques is also an important strength of the study. The observed variability in hygroscopicity between different periods and its association with aerosol chemical composition provide useful insights into the factors controlling aerosol water uptake. However, I believe that the interpretation of the intercomparison and the term "robust characterisation" requires some further clarification. The different techniques probe distinct optical properties (backscatter versus total scattering), wavelengths, particle populations, and sampling conditions; therefore, the retrieved hygroscopicity parameters are not directly equivalent. The authors appropriately acknowledge that the comparisons are primarily qualitative, but the implications of these methodological differences and the associated uncertainties should be discussed more explicitly before drawing general conclusions regarding the capability of remote sensing to quantitatively characterise aerosol hygroscopicity. Although the paper is too lengthy for a research article, some material could be included in a supplementary session. Here are my comments.

Response

We sincerely thank the referee for his/her time and suggestions, which have greatly contributed to improving the quality of this study. All the points raised have been carefully considered, and the corresponding revisions have been incorporated into the manuscript. Below we provide detailed, point-by-point responses to each comment (in blue).

Major comment # 1

The Comparability of f_β and f_σ. A core strength of this paper is its multi-instrument study, but the physical quantities compared are fundamentally different optical properties. The RALMO Raman lidar and COBALD backscatter sondes measure the aerosol backscatter coefficient β_{aer} at a 180^o scattering angle, whereas the tandem nephelometers measure the hemispheric scattering coefficient σ_{sp}. The authors acknowledge that these absolute values are not directly comparable due to differences in polar geometry and wavelength. They note that a "more quantitative comparison would require dedicated Mie simulations". However, despite this major physical caveat, the manuscript proceeds to make qualitative "good agreement" assertions throughout the text and presents them side by side in Tables 6 and 7. To elevate the scientific quality of this paper, given the importance of this issue to the intercomparison, I encourage the authors to conduct at least a preliminary Mie scattering analysis. Since they already have size-resolved particle distributions from the collocated SMPS system, they should perform a preliminary

Mie calculation for the case studies. Specifically, they can estimate the theoretical ratio of the backscatter Hänel parameter $\gamma_\beta(355\ \text{nm})$ to the scattering $\gamma_\sigma(550\ \text{nm})$, providing a physical baseline for the systematic discrepancies observed in Tables 6 and 7 (e.g., why γ_β) is frequently higher than $\gamma_\sigma(550\ \text{nm})$ on the same day).

Response

We sincerely thank the reviewer for this valuable comment. Given the importance of the different optical properties involved in the intercomparison, we performed preliminary, surface-based Mie simulations for the study cases with available SMPS size distributions and consequently added a new subsection entitled *"Assessment of aerosol hygroscopicity using Mie simulations" (Sect. 4.3.5)*. In these simulations, aerosol backscatter was calculated at 355 and 455 nm, corresponding to the RALMO and COBALD wavelengths, respectively, while aerosol scattering was calculated at 550 nm, corresponding to the tandem nephelometers. The simulations used measured dry SMPS size distributions and daily surface aerosol chemical composition to estimate the dry-particle refractive index and hygroscopicity parameter, while hygroscopic growth was calculated using the κ-Köhler theory.

The simulated enhancement factors and Hänel parameters are presented in Fig. 13 and Table 8 of the revised manuscript, with $\gamma_\sigma(550\ \text{nm})$ values ranging from 0.31 to 0.41 and $\gamma_\beta(355\ \text{nm})$ values from 0.30 to 0.32, and $\gamma_\beta(355\ \text{nm})/\gamma_\sigma(550\ \text{nm})$ ratios ranging from 0.77 to 0.99. These results suggest that differences in wavelength and optical property alone may not explain the differences observed between the RALMO and tandem-nephelometer hygroscopicity retrievals. Under the same assumed aerosol population, surface chemical composition, and hygroscopic growth, the simulations showed similar or larger hygroscopicity parameters for scattering at 550 nm than for backscatter at 355 nm. Differences among the dry size distributions could partly account for the range of simulated hygroscopicity parameters, since hygroscopic growth modifies particle size relative to wavelength and therefore could affect scattering and backscatter differently. The simulations also suggest larger γ_β values at 355 than at 455 nm, in qualitative agreement with the RALMO and COBALD-RS41 retrievals.

The results also showed that the simulated hygroscopicity parameters (Table 8) are generally closer to those obtained with the surface tandem of nephelometers (Table 6). This numerical similarity should not be interpreted as a quantitative closure, since the sampling times and reference RH conditions are not identical in all comparisons. Nevertheless, it may partly result from the fact that both the SMPS size distributions and the chemical composition used as inputs to the Mie calculations are measured at the surface. Consequently, differences between the simulated and RALMO-derived hygroscopicity may also reflect changes in aerosol size distribution, composition, morphology, and refractive index with altitude, particularly for decoupled aerosol layers.

The following changes have been introduced in the manuscript:

1. The text in **Lines 490–492 of the original manuscript** has been revised as follows (**Lines 530–534 in the revised manuscript**): This comparison should be interpreted with caution, since the retrievals are based on different wavelengths and optical properties, and the limited number of available RALMO cases should also be considered. To provide a preliminary physical baseline for assessing the effects of wavelength and the optical property considered under a common set of surface-based aerosol assumptions, Mie simulations were performed (Bohren and Huffman, 2007) and are discussed later in Sect. 4.3.5.

2. **A new Sect. 4.3.5**, *Assessment of aerosol hygroscopicity using Mie simulations*, has also been added to **the revised manuscript (Lines 675–707)**. The corresponding results are presented in the **new Fig. 13 and Table 8**. This subsection clarifies the surface-based nature of the simulations and the limitations of their comparison with vertically resolved retrievals.

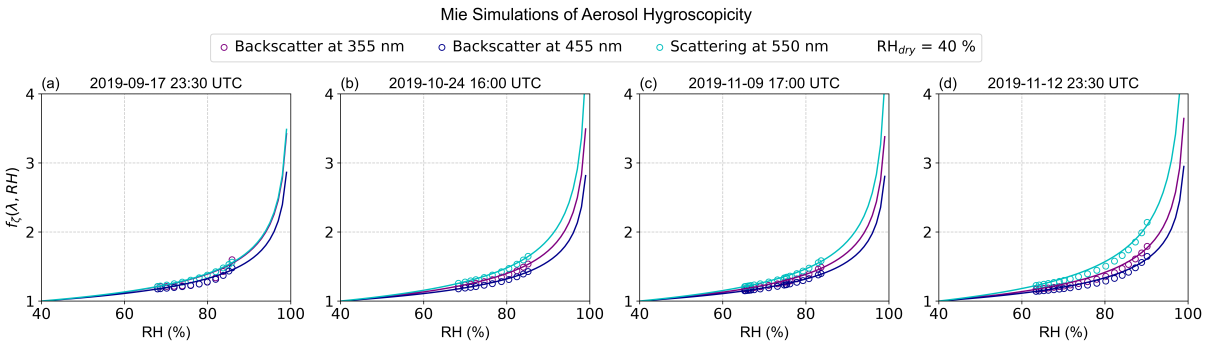


Figure 13: Enhancement factor ($f_\zeta(\lambda, RH)$) retrieved from Mie simulations for (a) 17 September 2019 at 23:30 UTC, (b) 24 October 2019 at 16:00 UTC, (c) 9 November 2019 at 17:00 UTC, and (d) 12 November 2019 at 23:30 UTC. Backscattering simulations are shown at 355 and 455 nm, and scattering simulations at 550 nm. Note that the humidograms were normalised to 40 % RH.

Table 8: Values of the enhancement factor ($f_\zeta(\lambda, RH)$) and Hänel parameter ($\gamma_\zeta(\lambda)$) retrieved from Mie simulations at 355 and 455 nm for aerosol backscatter and at 550 nm for aerosol scattering. Note that all values are normalised to 40 % RH. R_{f_ζ} and R_{γ_ζ} denote the ratios between the corresponding enhancement factors and Hänel parameters at the indicated wavelengths, respectively.

		$f_\zeta(\lambda,\ 85/40\ \%)$				
Date	SMPS interval	$f_\beta(355\ \text{nm})$	$f_\beta(455\ \text{nm})$	$f_\sigma(550\ \text{nm})$	$R_{f_\zeta}^{355/455}$	$R_{f_\zeta}^{355/550}$
17 September 2019 23:30 UTC	22:00 – 24:00	1.52 ± 0.08	1.43 ± 0.04	1.53 ± 0.02	1.06 ± 0.06	0.99 ± 0.05
24 October 2019 16:00 UTC	16:00 – 18:00	1.53 ± 0.02	1.42 ± 0.01	1.64 ± 0.02	1.08 ± 0.02	0.93 ± 0.02
09 November 2019 17:00 UTC	16:00 – 18:00	1.51 ± 0.02	1.42 ± 0.01	1.62 ± 0.02	1.07 ± 0.02	0.93 ± 0.02
12 November 2019 23:30 UTC	22:00 – 24:00	1.55 ± 0.02	1.44 ± 0.02	1.77 ± 0.03	1.07 ± 0.02	0.88 ± 0.02
		$\gamma_\zeta(\lambda)$				
Date	SMPS interval	$\gamma_\beta(355\ \text{nm})$	$\gamma_\beta(455\ \text{nm})$	$\gamma_\sigma(550\ \text{nm})$	$R_{\gamma_\zeta}^{355/455}$	$R_{\gamma_\zeta}^{355/550}$
17 September 2019 23:30 UTC	22:00 – 24:00	0.30 ± 0.04	0.26 ± 0.02	0.31 ± 0.01	1.12 ± 0.17	0.99 ± 0.13
24 October 2019 16:00 UTC	16:00 – 18:00	0.31 ± 0.01	0.25 ± 0.01	0.36 ± 0.01	1.21 ± 0.05	0.86 ± 0.03
09 November 2019 17:00 UTC	16:00 – 18:00	0.30 ± 0.01	0.25 ± 0.01	0.35 ± 0.01	1.18 ± 0.05	0.86 ± 0.03
12 November 2019 23:30 UTC	22:00 – 24:00	0.32 ± 0.01	0.26 ± 0.01	0.41 ± 0.01	1.19 ± 0.06	0.77 ± 0.03

3. We have also included in the **Supplement (Sect. S3)** the dry aerosol refractive indices **(Table S1)** and the corresponding SMPS size distributions for the analysed cases **(Fig. S4)**, both used as inputs to the Mie simulations.

Major comment # 2

The chemical composition data (NABEL station filters) are collected strictly at the surface, whereas hygroscopicity is evaluated at different heights, including the Free Troposphere (FT). The assumption of using surface chemistry as a proxy for decoupled FT layers. May request a more thorough discussion of the potential errors introduced by this extrapolation, especially in cases where boundary-layer air is not well mixed into the free troposphere. They can use their own RALMO profiles of potential temperature and water vapour mixing ratio (r).

Response

Thank you for highlighting this important point. We agree that surface aerosol chemical composition measurements cannot be assumed to represent aerosol layers at different altitudes. Chemical composition data are available at daily resolution and correspond to surface measurements, whereas hygroscopicity parameters are derived from RALMO measurements at higher temporal resolution and at different altitudes. The RALMO profiles of potential temperature and water vapour mixing ratio are used to identify the well-mixed layers required for the lidar hygroscopicity analysis. These layers may be decoupled from the surface, particularly when located in the free troposphere, and therefore well-mixed conditions within the layer do not necessarily imply that the atmosphere is well mixed from the surface up to the identified hygroscopic layer. Additionally, these conditions are met during specific periods and may not persist throughout the day to which the surface chemical composition measurements correspond. Consequently, a direct relationship between surface chemistry and individual hygroscopicity retrievals may not be representative.

We have therefore revised the manuscript to clarify the role and limitations of the surface chemical measurements. They are now consistently described as complementary information used to characterise the near-surface aerosol chemical composition during the campaign and are not used as a proxy for the aerosol chemical composition at different altitudes. We have also revised the corresponding interpretations throughout the manuscript to avoid implying a direct extrapolation of surface chemical composition to the aerosol layers identified by RALMO. Potential links between surface composition and vertically resolved hygroscopicity are treated as hypotheses rather than demonstrated explanations. This distinction also applies to the surface-based inputs used in the preliminary Mie simulations, which provide a conditional physical baseline rather than a reconstruction of the elevated aerosol layers.

The following changes have been introduced in the manuscript:

1. The text in **Line 6 of the original manuscript** has been revised as follows (**Lines 8–9 in the revised manuscript**): complementary surface aerosol chemical analyses.

2. The text in **Lines 16–17 of the original manuscript** has been revised as follows (**Lines 17–18 in the revised manuscript**): Surface chemical analyses showed a predominance of organic matter, with inorganic species accounting for approximately 11 % of the aerosol mass.

3. The text in **Lines 76–78 of the original manuscript** has been revised as follows (**Lines 82–88 in the revised manuscript**): Additionally, complementary in situ measurements, including tandem nephelometers were performed at ground level, enabling the evaluation of lidar retrievals in selected cases where the sampled layers were connected to the surface under well-mixed conditions. Nevertheless, some limitations arise from the different approaches and wavelengths used to obtain aerosol hygroscopicity parameters, since lidars rely solely on backscatter measurements, whereas tandem nephelometers use the scattering coefficient. Differences in sampling time and vertical representativeness further limit the direct comparability of the retrieved parameters.

4. The text in **Lines 389–393 of the original manuscript** has been revised as follows (**Lines 420–430 in the revised manuscript**): Note that chemical composition data are available at daily resolution and correspond to surface measurements, whereas hygroscopicity parameters are derived from RALMO measurements at higher temporal resolution and at different altitudes. These hygroscopicity studies require well-mixed conditions within the

analysed layers, which may be decoupled from the surface, particularly when located in the free troposphere. Therefore, thermodynamic homogeneity within a selected layer does not establish that it is mixed with the surface or that its aerosol chemical composition is represented by surface measurements. Additionally, these conditions are met during specific periods and may not persist throughout the day to which the surface chemical composition measurements correspond. Therefore, the surface chemical measurements are used to characterise near-surface aerosol composition, rather than to attribute the hygroscopicity of individual elevated layers to specific chemical components. Such attribution would require vertically resolved chemical measurements, and the uncertainty associated with extrapolating surface composition cannot be quantified here.

5. The text in **Lines 402–406 of the original manuscript** has been revised as follows (**Lines 439–451 in the revised manuscript**): Results from Tables 1 and 4 show that organic matter was the dominant component of the near-surface aerosol (4.9 ± 2.5 $\mu g/m^3$ (83 %)), whereas inorganic species accounted for approximately 11 % of the aerosol composition. The inorganic fraction was mainly composed of sulphate (0.30 ± 0.13 $\mu g/m^3$ (4.4 %)), nitrate (0.11 ± 0.08 $\mu g/m^3$ (4.1 %)) and ammonium (0.17 ± 0.07 $\mu g/m^3$ (2.8 %)). Elemental carbon showed a low concentration (0.28 ± 0.13 $\mu g/m^3$ (4.6 %)). Previous studies have also reported an important contribution of organic aerosol at Payerne (e.g., Minguillón et al., 2012; Hüglin and Grange, 2021; Dada et al., 2025). During the campaign, this contribution was particularly high in summer, especially in June (Tables 1 and 4), and may partly reflect enhanced photochemical activity during this season (Kammermann et al., 2010). In contrast, during autumn and winter, black carbon concentrations increase due to greater residential wood burning and a shallower boundary layer in the region (Minguillón et al., 2012; Dada et al., 2025), and these particles are typically less hygroscopic (Deshmukh et al., 2025; Lorenzo et al., 2025). The predominance of scattering and fine particles, together with lower concentrations of carbonaceous species during summer, may be associated with the higher hygroscopicity observed during this period compared with the second part of the campaign (Fig. 2 and Table 1). However, the surface composition does not establish the chemical composition of the layers analysed with RALMO and cannot, by itself, explain their retrieved hygroscopicity.

6. To consistently distinguish the surface measurements from the vertically resolved lidar observations, the following expressions were revised throughout the **case-study** discussion:

   - **Line 512 in the original manuscript** was revised to: daily surface aerosol chemical composition **(Lines 554–555 in the revised manuscript)**.
   - **Line 566 in the original manuscript** was revised to: Daily surface aerosol chemical measurements **(Line 609 in the revised manuscript)**.
   - The text in **Lines 602–603 of the original manuscript** has been revised to: daily surface aerosol chemical composition **(Line 647 in the revised manuscript)**.

7. The text in **Lines 622–626 of the original manuscript** has been revised, and the statement suggesting that surface aerosol chemical properties could be extrapolated to higher altitudes has been removed (**Lines 666–668 in the revised manuscript**): Regarding the surface chemical measurements, used here to characterise the near-surface aerosol chemical composition, the enhanced EC content combined with the lower SO_4^{2-} concentration (4.1 vs 4.7 %) may explain why this case exhibited lower hygroscopicity ($f_\beta(355\ \text{nm}) = 1.89 \pm 0.24$) than on 17 September ($f_\beta(355\ \text{nm}) = 6.98 \pm 0.37$).

8. The text in **Lines 674–678 of the original manuscript** has been revised as follows (**Lines 733–741 in the revised manuscript**): The highest occurrence was in summer (177 layers, 78 % of all cases), with aerosols being more hygroscopic in June ($f_\beta(355\ \text{nm}) = 3.65 \pm 0.70$ and $\gamma_\beta(355\ \text{nm}) = 0.94 \pm 0.15$). Fewer cases were detected during October–December, when lower hygroscopicity was observed ($f_\beta(355\ \text{nm}) = 2.36 \pm 0.26$ and $\gamma_\beta(355\ \text{nm}) = 0.60 \pm 0.08$), limiting the representativeness of comparisons with summer. Complementary surface chemical measurements showed a predominance of organic matter (OM) during both campaign periods, with a higher sulphate mass fraction in June–August and higher elemental carbon (EC) and nitrate mass fractions in September–December. This contrast in surface aerosol chemical composition between the two periods may help explain the observed differences in hygroscopicity. However, the surface observations provide context for the campaign and do not establish the chemical drivers of the temporal variability in vertically resolved aerosol hygroscopicity.

9. The sentence in **Line 694 of the original manuscript** was removed as part of the restructuring and shortening of the **Conclusions section**.

Major comment # 3

In section 4.3.3, the authors state: "Overall, no consistent spectral dependence of the retrieved hygroscopic parameters was identified across the different instruments". However, in Section 5 (Conclusions), they state: "A weak wavelength-dependence was also observed, with larger hygroscopicity parameters at shorter wavelengths". The authors need to clarify this apparent contradiction. They should explicitly define where and under what conditions the "weak wavelength-dependence" was statistically supported.

Response

Thank you for pointing out this inconsistency. We agree that the statement in the Conclusions referring to a weak wavelength dependence was not supported by a statistical analysis. The spectral comparison in this study is limited to a small number of case studies. In particular, only the cases on 17 September and 12 November include backscatter-derived hygroscopicity parameters for the same case studies from RALMO, COBALD-RS41, and the ceilometer at different wavelengths. The case on 10 December includes RALMO and ceilometer measurements, while the tandem nephelometer provides scattering-based hygroscopicity parameters at 550 nm under near-surface conditions and is therefore not directly comparable in terms of absolute values. Furthermore, comparisons across instruments combine differences in wavelength with differences in sampling and retrieval methodology, limiting the attribution of observed discrepancies to wavelength alone. Moreover, the values summarised in Table 7 do not show a consistent monotonic dependence on wavelength and therefore cannot support a systematic increase or decrease in aerosol hygroscopicity with wavelength. Therefore, no statistically supported wavelength dependence can be established from the available observations. We have removed the statement *"A weak wavelength-dependence was also observed, with larger hygroscopicity parameters at shorter wavelengths"* from the **Conclusions** and clarified that the available case studies show no consistent spectral dependence of the retrieved hygroscopicity parameters.

The following changes have been introduced in the manuscript:

1. The text in **Line 551 of the original manuscript** has been revised as follows (**Line 593 in the revised manuscript**): The ceilometer retrieval, however, yielded lower hygroscopicity parameters, with
2. The text in **Line 693 of the Conclusions in the original manuscript** has been revised as follows (**Lines 754–760 in the revised manuscript**): No consistent spectral dependence was identified from the available observational case studies. The small number of cases and the differences in sampling and retrieval methodology between instruments prevent a robust assessment of a general wavelength dependence. Since the different techniques rely on different aerosol optical and microphysical properties, measurement wavelengths, sampling conditions and vertical representativeness, the comparisons were performed qualitatively and should be interpreted with caution. The Mie simulations likewise suggest no consistent spectral dependence and that near-surface aerosol conditions may not be representative of those within atmospheric layers, which may partly contribute to the differences observed between surface and vertically resolved hygroscopicity retrievals.

Minor comment # 1

Line 7: The manuscript refers to "three independent approaches." Since the Raman lidar, COBALD-RS41, and tandem nephelometer measurements are based on different optical quantities and are not fully independent, the authors may consider using "three complementary approaches" instead.

Response

We appreciate the referee's suggestion. We have replaced "three independent approaches" with "three complementary approaches" (**Line 9 in the revised manuscript**).

Minor comment # 2

Lines 464-465: "The tandem nephelometers' measurements are only representative of near-surface conditions." Please revise to "the tandem nephelometer measurements are representative only of near-surface conditions."

Response

Thank you for this correction. We have revised the sentence as suggested: Note that the tandem nephelometer measurements are representative only of near-surface conditions (**Line 503 in the revised manuscript**).

Minor comment # 3

Line 639: The terminology "RALMO measurements alone enable the retrieval of aerosol hygroscopicity parameters" may be slightly too broad. Since the retrieval requires specific atmospheric conditions, particularly a well-mixed layer and suitable RH structure, I suggest changing this to

"RALMO measurements can enable the retrieval. . . " or explicitly retaining the conditions under which the retrieval is applicable.

Response

Thank you for this suggestion. We agree that the retrieval of aerosol hygroscopicity parameters depends on specific atmospheric conditions and that the wording "RALMO measurements alone enable the retrieval" is too broad. We have therefore removed this wording and revised the corresponding sentence in the **Conclusions** as follows: The continuous operation of the RAman Lidar for Meteorological Observations (RALMO), which provides measurements of temperature, water vapour mixing ratio, and aerosol backscatter during both daytime and nighttime, enabled aerosol hygroscopicity to be investigated under different atmospheric conditions over a six-month period. (**Lines 711-714 in the revised manuscript**).

Minor comment # 4

Lines 424-425: the statement "during summer, more wind profiles could be monitored due to lower cloud cover and bird migration". Can you explain bird migration? Bird migration is not obviously connected to the availability of wind lidar measurements, or is that a typo?

Response

Thank you for pointing this out. The reference to bird migration was a mistake and has been removed from the revised manuscript. (**Lines 463-464 in the revised manuscript**).

General comment # 1

The authors could consider moving some of the detailed case studies from Section 4.3 to the Supplementary Material. This could help shorten the manuscript and allow the main text to focus more clearly on the methodology and interpretation of aerosol hygroscopicity retrieval.

Response

We greatly appreciate this suggestion, which has helped us improve the manuscript's structure and readability. To shorten the manuscript and improve the focus of **Sect. 4.3**, we have moved **Figures A1, A2, 11, A3, and 13 from the original manuscript have been moved to the Supplement and renumbered as Figs. S6, S7, S8, S9, and S10, respectively.**. These figures show the in situ observations for the case studies, the temporal and vertical evolution of RALMO measurements, and profiles from COBALD-RS41 sondes where available. The main text retains the hygroscopicity retrievals and their interpretation, with references to the supplementary material where appropriate. We have also substantially revised and shortened the **Conclusions section (Lines 708–766 in the revised manuscript)** to focus more clearly on the main findings.

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