Changes introduced in the manuscript

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

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The entries below consolidate the changes described in the responses to both referees. Revised-manuscript line numbers refer to the new version of the supplied PDF. Original-manuscript line numbers should be checked against the original manuscript. Changes to the Supplement are listed separately because it has its own numbering.

The text in Lines 2–18 of the original manuscript has been revised as follows:

Abstract

Revised manuscript: Lines 2–21.

This study presents a comprehensive assessment of aerosol hygroscopicity during the HYGRO19 field campaign conducted at the MeteoSwiss station in Payerne (Switzerland) from June to December 2019. Continuous profiles of aerosol backscatter, water vapour mixing ratio, and temperature provided by the Raman Lidar for Meteorological Observations (RALMO) during both daytime and nighttime allowed aerosol hygroscopic growth to be investigated temporally and vertically under different atmospheric conditions. The campaign also provided a unique experimental framework by combining continuous Raman lidar observations with vertically resolved in situ measurements from the balloon-borne Compact Backscatter Aerosol Detector coupled with Vaisala RS41 radiosondes (COBALD-RS41), ground-based tandem nephelometers, and complementary surface aerosol chemical analyses, enabling aerosol hygroscopicity to be evaluated using three complementary approaches. Two contrasting aerosol regimes were observed: a summer period characterised by relatively high aerosol loading and frequent influence from long-range transported particles, and a second period dominated by low-pressure systems, persistent cloudiness, and reduced aerosol concentrations. Through these different atmospheric conditions, more than 200 well-mixed hygroscopic aerosol layers were identified with RALMO, with median hygroscopicity parameters of $f_\beta(355\ \text{nm}) = 2.64 \pm 0.64$ and $\gamma_\beta(355\ \text{nm}) = 0.70 \pm 0.18$. Statistical analyses revealed no systematic dependence of aerosol hygroscopicity on altitude, with comparable values in the atmospheric boundary layer and free troposphere. Multi-instrument comparisons showed generally consistent hygroscopic behaviour among RALMO, COBALD-RS41, and tandem nephelometer observations in the analysed cases, despite differences between the techniques. Surface chemical analyses showed a predominance of organic matter, with inorganic species accounting for approximately 11 % of the aerosol mass. No clear dependence of aerosol hygroscopicity on particle size was identified. These results show that continuous Raman lidar observations, complemented by vertically resolved and ground-based in situ measurements, provide a robust framework for characterising aerosol hygroscopicity under ambient atmospheric conditions.

The text in Line 27 of the original manuscript has been revised as follows:

Revised manuscript: Line 30.

to take up

The text in Lines 36–40 of the original manuscript has been revised as follows:

Revised manuscript: Lines 40–45.

Most in situ techniques alter the ambient conditions and are subject to particle losses in the sampling lines, which can modify aerosol properties relative to ambient conditions (e.g., particle number concentration, size distribution, chemical composition, scattering, and absorption; Hermann et al., 2001; Bedoya-Velásquez et al., 2018). For humidified nephelometer measurements, uncertainties in the scattering enhancement factor ($f(RH)$) can be substantial at high relative humidity (RH), with values of approximately 20–40 % reported for moderately hygroscopic aerosols (Titos et al., 2016).

The text in Lines 49-50 of the original manuscript has been revised as follows:

Revised manuscript: Lines 52–54.

In contrast, lidar measurements provide vertically resolved aerosol information, which make such measurements suitable for investigating aerosol hygroscopicity within well-mixed aerosol layers (Ferrare et al., 1998).

The text in Lines 63-66 of the original manuscript has been revised as follows:

Revised manuscript: Lines 67–71.

Despite these approaches having served as useful benchmarks, long-term vertically resolved analyses of aerosol hygroscopicity remain limited (Sicard et al., 2022; Jing et al., 2026). Indeed, most previous studies have been conducted during specific field campaigns or have focused on only a few case studies, often based on nighttime observations. Furthermore, comprehensive aerosol hygroscopicity studies require simultaneous measurements of aerosol chemical composition and particle size (Pérez-Ramírez et al., 2021; Laly et al., 2025), which are not always available.

The text in Lines 67-80 of the original manuscript has been revised as follows:

Revised manuscript: Lines 72–88.

The Raman Lidar for Meteorological Observations (RALMO) system, operating at the MeteoSwiss aerological station in Payerne, Switzerland, can overcome some of the previously mentioned limitations in aerosol hygroscopicity studies. Its capability to provide high-vertical-resolution profiles of aerosol backscatter, temperature, and water vapour mixing ratio and to operate continuously during both daytime and nighttime (Dinoev et al., 2013) makes it particularly suitable for aerosol hygroscopicity studies (Navas-Guzmán et al., 2019). In this context, the HYGRO19 field campaign, conducted from June to December 2019 in Payerne, represented a unique opportunity to combine continuous RALMO observations with complementary in situ techniques to characterise aerosol hygroscopicity under ambient atmospheric conditions. Furthermore, vertical aerosol measurements performed with the Compact Backscatter Aerosol Detector (COBALD, Brunamonti et al., 2018), alongside RH profiles from operational RS41 radiosondes, permitted the direct comparison of aerosol hygroscopicity parameters retrieved from RALMO with those derived from vertically resolved in situ observations. This makes HYGRO19 one of the few studies in which aerosol hygroscopicity has been characterised vertically using balloon-borne COBALD-RS41 measurements. 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.

The text in Lines 81-87 of the original manuscript has been revised as follows:

Revised manuscript: Lines 89–98.

This work focuses on the characterisation of aerosol hygroscopicity during the HYGRO19 field campaign using complementary remote sensing and in situ techniques, and on assessing the consistency and limitations of their comparisons. Selected multi-instrument comparisons are performed between RALMO-derived hygroscopicity parameters and those obtained from vertically resolved in situ observations from COBALD-RS41 sondes and ground-based tandem nephelometer measurements. Continuous RALMO observations additionally provide a statistical characterisation of the temporal and vertical variability of aerosol hygroscopicity within the conditions that fulfil the retrieval criteria. Complementary aerosol and meteorological observations support the interpretation of the observed hygroscopic behaviour.

The paper is structured as follows: Sect. 2 describes the experimental site and instrumentation, Sect. 3 details the methodologies used, Sect. 4 reports the main results and their discussion, and Sect. 5 summarises the conclusions and provides perspectives for future work.

The text in Lines 94-98 of the original manuscript has been revised as follows:

Revised manuscript: Lines 105–113.

The HYGRO19 campaign focused on the characterisation of hygroscopic aerosol growth under ambient atmospheric conditions using three complementary approaches: remote sensing measurements from the RALMO Raman lidar, ground-based in situ measurements from tandem nephelometers, and vertically resolved in situ measurements from COBALD sondes operated alongside RS41 radiosondes (COBALD-RS41). A central objective was to assess the consistency and limitations of the hygroscopicity parameters obtained with these techniques, accounting for their different optical properties, wavelengths, and sampling conditions. Continuous RALMO measurements of aerosol backscatter, temperature, and water vapour mixing ratio also enabled the temporal and vertical variability of aerosol hygroscopicity to be investigated within selected well-mixed layers. Additional aerosol and meteorological observations were used to characterise the atmospheric conditions and support the interpretation of the observed hygroscopic behaviour.

The text in Lines 154-155 of the original manuscript has been revised as follows:

Revised manuscript: Lines 170–171.

at three wavelengths (450, 525, and 635 nm)

The text in Lines 179 and 182 of the original manuscript has been revised as follows:

Revised manuscript: Lines 195 and 198.

The spelling "Hueglin" has been changed to "Hüglin".

The text in Section 3.4 Lines 256-259 of the original manuscript has been revised as follows:

Revised manuscript: Lines 275–277.

After the sentence: This is achieved by assuming a well-mixed atmospheric layer that typically occurs when water vapour mixing ratio and potential temperature profiles remain nearly constant with altitude (Feingold and Morley, 2003).

We have added: Additionally, following Navas-Guzmán et al. (2019), potential changes in air-mass origin with altitude should be assessed using backward trajectories as complementary information to the thermodynamic profiles.

The text in Line 287 of the original manuscript has been revised as follows:

Revised manuscript: Lines 305–306.

Figure 2 shows the temporal evolution of in situ ground-based measurements of aerosol scattering (σ_{sp}, a) and absorption (σ_{ab}, c)

The text in Lines 294–305 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Lines 312–327.

Figure 2 on page 11 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Figure 2 on page 12.

The text in Lines 298-301 of the original manuscript has been revised as follows:

Revised manuscript: Lines 316–322.

Concurrently, the analysis of the SAE (450–635 nm, Fig. 2b) suggests a predominance of fine particles (SAE = 1.64 ± 0.26), although some outliers are observed. Separately, the AERONET AOD-AE measurements (Fig. 1b) also suggest a general predominance of fine particles in the atmospheric column, with a median AOD-AE (440–870 nm) value of 1.46 ± 0.21 for the entire campaign. These observations refer to different temporal coverage and sampling conditions and should not be interpreted as a quantitative agreement between the two datasets. The predominance of fine particles near the surface is also supported by the small difference observed between PM_{10} and $PM_{2.5}$ ($PM_{10-2.5}$ = 3.0 ± 1.6 $\mu g/m^3$, Fig. 2f).

The text in Line 302 of the original manuscript has been revised as follows:

Revised manuscript: Line 323.

The spelling "Hueglin" has been changed to "Hüglin".

The text in Lines 303-304 of the original manuscript has been revised as follows:

Revised manuscript: Lines 324–327.

Previous studies have reported that Payerne is usually influenced by local and Swiss Plateau emissions, as well as by the transport of anthropogenic pollution from mainland Europe (Minguillón

et al., 2012; Dada et al., 2025). This description provides background information on the site and does not constitute a source attribution for individual episodes during HYGRO19.

The text in Lines 305-306 of the original manuscript has been revised as follows:

Revised manuscript: Lines 328–330.

The analysis of the aerosol absorption coefficient (Fig. 2c) exhibits values ranging from 0.06 to 26 Mm^{-1}, with a median of 3.4 ± 1.9 Mm^{-1} at 520 nm, indicating substantial temporal variability but generally low absorption levels compared with values typically reported for suburban, urban, and traffic sites (Rovira et al., 2025).

The text in Lines 309-311 of the original manuscript has been revised as follows:

Revised manuscript: Lines 333–334.

The analysis of carbonaceous measurements further suggests that these changes can be associated with a higher frequency of local wood burning (Fig. 2g-h).

The text in Lines 311-315 of the original manuscript has been revised as follows:

Revised manuscript: Lines 334–340.

Regarding chemical composition (Fig. 2h), inorganic species (sulphate, ammonium, and nitrate) show an important contribution to particulate matter, accounting for 11 %. However, the TC values compared with measured inorganic species and $PM_{2.5}$ support that organic matter is the main fraction (83 %), which is typically the largest fraction in summer (OM = 87 %), compared with the second part of the campaign (OM = 81 %). The predominance of organic aerosol and its larger contribution during summer (55–60 %) than in winter (30–40 %) have also been reported at Payerne (Aksoyoglu et al., 2011; Minguillón et al., 2012; Bozzetti et al., 2016; Hüglin and Grange, 2021; Nowak et al., 2023–2025; Dada et al., 2025).

The text in Lines 321–331 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Lines 346–356.

Figure 3 on page 13 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Figure 3 on page 14.

The text in Lines 330-331 of the original manuscript has been revised as follows:

Revised manuscript: Lines 355–356.

reveal the highest values within the first 1.5 km agl corresponding approximately to the mean atmospheric boundary layer (ABL) height (Collaud Coen et al., 2014).

The text in Lines 334-338 of the original manuscript has been revised as follows:

Revised manuscript: Lines 359–363.

During the first period, aerosol is frequently present throughout the entire ABL and even within the free troposphere (FT). However, on some days, aerosol layers are observed above and decoupled

from the ABL (e.g. 2–5 and 17–19 July). In the second period, aerosol backscatter is generally lower (Table 2), and aerosol is typically confined below 1.0 km agl, consistent with a lower ABL height (Collaud Coen et al., 2014). However, an episode of relatively enhanced aerosol backscatter extending into the FT is observed around 1 December.

The text in Section 4.2, Lines 340-342 of the original manuscript has been revised as follows:

Revised manuscript: Lines 365–371.

The backscatter enhancement factor retrieved with RALMO ($f_\beta(355\ \text{nm})$) during the campaign was computed within the layers fulfilling the criteria described in Sect. 3.4. To assess potential changes in air-mass origin with altitude, as reported by Navas-Guzmán et al. (2019), five-day (120 h) backward trajectories arriving over Payerne were calculated for each investigated hygroscopic aerosol layer using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT model 5.4.0; Stein et al., 2015; Rolph et al., 2017). The trajectory analysis considered arrival times corresponding to the RALMO observations and arrival heights at the lower and upper boundaries of the investigated layers. A detailed comparison of the trajectories arriving at both boundaries is provided in the Supplement (Sect.S1 and Fig. S1). For comparison

The text in Line 345 of the original manuscript has been revised as follows:

Revised manuscript: Lines 374-375.

Most hygroscopic layers were observed in summer (177 hygroscopic layers, 78 %),

The text in Lines 365-367 of the original manuscript has been revised as follows:

Revised manuscript: Lines 395–398.

...using established statistical rules for histogram bin selection (Doane's rule (Doane, 1976), Rice's rule (Rubia, 2024), and Scott's normal reference rule (Scott, 2010)). These methods account for different characteristics of the data distribution, including the number of observations, variability, and skewness.

The text in Lines 389–393 of the original manuscript has been revised as follows:

Revised manuscript: Lines 420–430.

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.

The text in Lines 402–406 of the original manuscript has been revised as follows:

Revised manuscript: Lines 439–451.

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. 1 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.

The text in Lines 414-420 of the original manuscript has been revised as follows:

Revised manuscript: Lines 453–459.

The relationship between particle size and the enhancement factor shows no clear dependence on particle size, with differences not being statistically significant. Fine particles yielded $f_\beta(355\ \text{nm}) = 2.60 \pm 0.49$ and $\gamma_\beta(355\ \text{nm}) = 0.70 \pm 0.14$, compared to $f_\beta(355\ \text{nm}) = 2.30 \pm 0.80$ and $\gamma_\beta(355\ \text{nm}) = 0.60 \pm 0.22$ for larger particles. The corresponding analysis is shown in Fig. S3 in the Supplement. Previous studies have also reported no clear dependence of aerosol hygroscopicity on particle size, with different size-dependent hygroscopic behaviours observed depending on aerosol properties and atmospheric conditions (Kammermann et al., 2010; Zieger et al., 2013; Titos et al., 2014b; Jefferson et al., 2017; Sicard et al., 2022).

The text in Line 425 of the original manuscript has been revised as follows:

Revised manuscript: Lines 463–464.

The reference and bird migration was removed from the manuscript.

The text in Fig. 7 (before original Line 440) of the original manuscript has been revised as follows:

Figure 7 has been updated by replacing wind-direction labels in degrees with compass directions (e.g. N, NE, E) to facilitate interpretation and comparison with the manuscript discussion.

The text in Lines 464–465 of the original manuscript has been revised as follows:

Revised manuscript: Line 503.

Note that the tandem nephelometer measurements are representative only of near-surface conditions.

The text in Lines 476 of the original manuscript has been revised as follows:

Revised manuscript: Line 515.

We have changed the date from December 9 to December 8.

The text in Lines 477-479 of the original manuscript has been revised as follows:

Revised manuscript: Lines 516–519.

For this day, HYSPLIT backward trajectories showed long-range transport from the North Atlantic and North America (Fig. S5a in the Supplement), where numerous fire hotspots were detected by the NASA/NOAA Suomi National Polar-orbiting Partnership satellite (`https://firms.modaps.eosdis.nasa.gov/`, Fig. S5b in the Supplement).

The text in Lines 490–492 of the original manuscript has been revised as follows:

Revised manuscript: Lines 530–534.

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.

The text in Line 506 of the original manuscript has been revised as follows:

Revised manuscript: Line 548.

(backward trajectories shown in Fig. S2a in the Supplement)

The text in Line 508 of the original manuscript has been revised as follows:

Revised manuscript: Line 550.

(graphs shown in Fig. S6 in the Supplement)

The text in Line 510 of the original manuscript has been revised as follows:

Revised manuscript: Line 552.

(Fig. S6a-d)

The text in Line 512 of the original manuscript has been revised as follows:

Revised manuscript: Line 554.

(Fig. S6e)

The text in Line 512 of the original manuscript has been revised as follows:

Revised manuscript: Lines 554–555.

daily surface aerosol chemical composition

The text in Line 515 of the original manuscript has been revised as follows:

Revised manuscript: Line 557.

(Fig. S6, right panels)

The text in Line 516 of the original manuscript has been revised as follows:

Revised manuscript: Line 559.

(Fig. S6h)

The text in Line 519 of the original manuscript has been revised as follows:

Revised manuscript: Line 562.

(Fig. S6i)

The text in Line 551 of the original manuscript has been revised as follows:

Revised manuscript: Line 593.

The ceilometer retrieval, however, yielded lower hygroscopicity parameters, with

The text in Line 558 of the original manuscript has been revised as follows:

Revised manuscript: Lines 600–601.

We have added (backward trajectories shown in Fig. S2b in the Supplement)

The text in Line 560 of the original manuscript has been revised as follows:

Revised manuscript: Lines 602-603.

(graphs shown in Fig. S7 in the Supplement)

The text in Line 563 of the original manuscript has been revised as follows:

Revised manuscript: Line 606.

(Fig. S7b)

The text in Lines 564-565 of the original manuscript has been revised as follows:

Revised manuscript: Line 607.

(Fig. S7c)

The text in Line 566 of the original manuscript has been revised as follows:

Revised manuscript: Line 609.

Daily surface aerosol chemical measurements

The text in Line 569 of the original manuscript has been revised as follows:

Revised manuscript: Lines 612-613.

(Fig. S8 in the Supplement, left panel)

The text in Line 571 of the original manuscript has been revised as follows:

Revised manuscript: Lines 614-615.

(Fig. S8 in the Supplement, right panel)

The text in Line 572 of the original manuscript has been revised as follows:

Revised manuscript: Line 615.

(Fig. S8d) and in RH from 65 % to 90 % (Fig. S8e).

The text in Line 573 of the original manuscript has been revised as follows:

Revised manuscript: Line 616.

(Fig. S8f)

The text in Line 584 of the original manuscript has been revised as follows:

Revised manuscript: Line 627.

(Fig. 11a)

The text in Lines 586 and 589 of the original manuscript has been revised as follows:

Revised manuscript: Lines 629 and 632.

(Fig. 11b)

The text in Line 613 of the original manuscript has been revised as follows:

Revised manuscript: Line 657.

(Fig. 12a)

The text in Lines 585–593 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Lines 629–637.

Figure 12 on page 28 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Figure 11 on page 28.

The text in Line 596 of the original manuscript has been revised as follows:

Revised manuscript: Line 640.

(Fig. S2c in the Supplement)

The text in Line 599 of the original manuscript has been revised as follows:

Revised manuscript: Line 643.

(shown in Fig. S9 in the Supplement)

The text in Line 602 of the original manuscript has been revised as follows:

Revised manuscript: Line 646.

(Fig. S9d)

The text in Lines 602-603 of the original manuscript has been revised as follows:

Revised manuscript: Line 647.

daily surface aerosol chemical composition

The text in Line 606 of the original manuscript has been revised as follows:

Revised manuscript: Line 650.

Figure S10a-c (in the Supplement) shows

The text in Line 607 of the original manuscript has been revised as follows:

Revised manuscript: Line 652.

Fig. S10a

The text in Line 610 of the original manuscript has been revised as follows:

Revised manuscript: Line 654.

Fig. S10

The text in Line 610 of the original manuscript has been revised as follows:

Revised manuscript: Line 654.

Fig. S10d

The text in Line 611 of the original manuscript has been revised as follows:

Revised manuscript: Line 655.

Fig. S10e

The text in Line 612 of the original manuscript has been revised as follows:

Revised manuscript: Line 656.

Fig. S10f

The text in Line 613 of the original manuscript has been revised as follows:

Revised manuscript: Line 657.

(Fig. 12a)

The text in Line 614 of the original manuscript has been revised as follows:

Revised manuscript: Line 658.

(Fig. 12b)

The text in Line 620 of the original manuscript has been revised as follows:

Revised manuscript: Line 664.

(Fig. 12b)

The text in Lines 622–632 of the original manuscript has been revised as follows:

Revised manuscript: Lines 666–674.

Figure 14 on page 30 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: Figure 12 on page 30.

Table 7 on page 30 of the original manuscript has been repositioned to improve the layout and reduce blank space.

Revised manuscript: page 30.

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 ($f_\beta(355\ \text{nm}) = 6.98 \pm 0.37$). Actually, previous studies have also reported that particles with high EC concentrations tend to exhibit lower hygroscopicity, while those primarily composed of sulphuric acid and ammonia show higher hygroscopicity (Zieger et al., 2014; Jefferson et al., 2017; Bedoya-Velásquez et al., 2019; Chen et al., 2019; Pérez-Ramírez et al., 2021; Cedeño et al., 2023; Deshmukh et al., 2025; Li et al., 2025; Lorenzo et al., 2025). But the key aspect here is the close proximity to clouds, suggesting how aerosol hygroscopicity can facilitate particle activation as cloud condensation nuclei under favourable thermodynamic and dynamical conditions (Pérez-Ramírez et al., 2021).

The text in After Lines 622–626; before the original Conclusions of the original manuscript has been revised as follows:

A new Sect. 4.3.5 has been added after Line 631 of the original manuscript, before the Conclusions: Assessment of aerosol hygroscopicity using Mie simulations

Revised manuscript: Lines 675–707; Fig. 13; Table 8.

To assess the effects of wavelength and optical property on the retrieved hygroscopicity parameters, a quantitative comparison was performed using preliminary, surface-based Mie simulations (Bohren and Huffman, 2007). Aerosol backscatter coefficients at the RALMO and COBALD wavelengths (355 and 455 nm, respectively) and aerosol scattering coefficients at the tandem nephelometer wavelength (550 nm) were simulated using the Mie code (Prahl, 2026). For these simulations, daily surface aerosol chemical composition measurements (Sect. 2.5) were used to estimate the effective dry-particle refractive index (m_{dry}) using a volume-weighted mixing rule (Liu and Daum, 2008), as well as the hygroscopicity parameter (κ; Petters and Kreidenweis, 2007) The refractive indices of the individual dry aerosol components used as inputs to the Mie simulations are provided in Table S1 of the Supplement. Hygroscopic particle growth and the corresponding wet particle size distributions were then calculated from the measured dry SMPS size distributions using the κ-Köhler theory (Petters and Kreidenweis, 2007). Finally, the change in refractive index due to

water vapour uptake was calculated by volume-weighting m_{dry} with the refractive index of water (m_{H_2O}; Hale and Querry, 1973).

Based on these calculations, the dependence of $f_\zeta(\lambda)$ on RH (Fig. 13) was obtained for the study cases with available SMPS size distributions. The simulated hygroscopicity parameters (Table 8) suggest that differences in wavelength and optical property alone may not explain the differences observed between the RALMO and tandem-nephelometer hygroscopicity retrievals (Table 7). Under the assumption of the same aerosol population, surface chemical composition, and hygroscopic growth, the Mie simulations yielded similar or larger hygroscopicity parameters for scattering at 550 nm than for backscatter at 355 nm, with γ_σ(550 nm) ranging from 0.31 to 0.41 and γ_β(355 nm) from 0.30 to 0.32. Differences among the dry size distributions (Fig. S4 in the Supplement) could partly account for the range of values obtained for the simulated hygroscopicity parameters. Since the Mie size parameter depends on both particle size and wavelength, hygroscopic growth can modify the relative scattering and backscatter behaviour at the different wavelengths. Therefore, the larger γ_σ(550 nm) values should not be interpreted as a general increase in aerosol hygroscopicity with wavelength. Conversely, the simulations yielded greater γ_β values at 355 than at 455 nm, in qualitative agreement with the RALMO and COBALD-RS41 retrievals (Table 7).

Results from Table 8 also show that the simulated hygroscopicity parameters are generally closer to those obtained with the surface tandem 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. This subsection clarifies the surface-based nature of the simulations and the limitations of their comparison with vertically resolved retrievals. 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. These results are consistent with previous Mie studies (e.g., Zieger et al., 2013; Bedoya-Velásquez et al., 2018; Navas-Guzmán et al., 2019), which showed that aerosol hygroscopicity derived from scattering and backscatter can depend on particle size, chemical composition, and wavelength.

The text in Lines 633–699 of the original manuscript has been revised as follows:

Revised manuscript: Lines 708–766.

The Conclusions have been substantially revised and shortened.

Conclusions

This study has focused on aerosol hygroscopicity during the intercomparison campaign of hygroscopic aerosol properties using remote sensing and in situ techniques (HYGRO19 campaign), which was carried out at the MeteoSwiss aerological station in Payerne, Switzerland (46.82° N, 6.95° E, 491 m asl) from June to December 2019. 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. More than 200 hygroscopic aerosol layers were identified, constituting one of the largest databases available for Raman lidar aerosol hygroscopicity studies. The use of balloon-borne Compact

Backscatter Aerosol Detectors coupled with Vaisala RS41 radiosondes (COBALD-RS41) provided unique vertically resolved in situ observations for direct comparison with RALMO retrievals. Additionally, tandem nephelometer measurements allowed aerosol hygroscopicity to be evaluated under well-mixed near-surface conditions. Complementary in situ and remote sensing observations were also used to provide a comprehensive characterisation of aerosol and atmospheric conditions.

Two contrasting aerosol regimes were observed during the campaign. The first, from June to August 2019, was characterised by relatively high aerosol loading and frequent clear-sky conditions, whereas the second, from September to December, was dominated by low-pressure systems, persistent cloudiness, and reduced aerosol concentrations. The median values of Aerosol Optical Depth (AOD) at 500 nm and extinction Ångström exponent (AOD-AE) between 440–870 nm were 0.24 ± 0.09 and 1.50 ± 0.19, respectively, during the first period, representing a considerable aerosol load for a rural background site such as Payerne. Backward-trajectory analyses indicated the influence of air masses from mainland Europe, which typically act as a source of anthropogenic pollution. However, some cases of air masses originating from Southwestern Europe and North Africa were also detected, showing higher aerosol loads (AOD (500 nm) approximately 0.3) and low AOD-AE values (approximately 0.4), suggesting the presence of dust particles. During the second period, only a few AERONET measurements were available, with median AOD (500 nm) and AOD-AE (440–870 nm) values of 0.07 ± 0.02 and 1.15 ± 0.13, respectively, more typical of a rural background site. These meteorological conditions largely limited the number of cases available for aerosol hygroscopicity studies using RALMO.

Aerosol hygroscopicity retrieved with RALMO showed high hygroscopic growth, with f_β(355 nm, 85/40 %) values ranging from 1.2 to 7.2 and a median of 2.64 ± 0.64, corresponding to a median γ_β(355 nm) of 0.70 ± 0.18. The highest occurrence was in summer (177 layers, 78 % of all cases), with aerosols being more hygroscopic in June (f_β(355 nm) = 3.65 ± 0.70 and γ_β(355 nm) = 0.94 ± 0.15). Fewer cases were detected during October–December, when lower hygroscopicity was observed (f_β(355 nm) = 2.36 ± 0.26 and γ_β(355 nm) = 0.60 ± 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. Regarding the vertical variability of aerosol hygroscopicity, no statistically significant differences were observed between layers below and above 1.5 km agl, used as a threshold to distinguish the Atmospheric Boundary Layer (ABL) from the Free Troposphere (FT). A more detailed classification distinguishing the Convective Boundary Layer (CBL), Residual Layer (RL), and FT showed no statistically significant differences.

Multi-instrument comparisons showed generally consistent hygroscopic behaviour between RALMO, COBALD-RS41, and tandem nephelometer observations in the analysed case studies. RALMO and tandem nephelometer retrievals showed good agreement for aerosol layers within the residual layer, when well-mixed conditions extended up to the RL. For example, on 10 December 2019, γ_β(355 nm) = 0.46 ± 0.09 was obtained from RALMO and γ_σ(550 nm) = 0.48 ± 0.05 from the tandem nephelometers. However, some discrepancies were found for some layers identified within the free troposphere, which may be due to the presence of decoupled layers, which can exhibit different optical and microphysical properties compared to those near the surface. The results obtained confirmed the capability of these techniques to robustly

characterise aerosol hygroscopicity, as indicated by the close agreement between the retrieved hygroscopicity parameters and their associated uncertainties. 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.

Further investigation of the relationship between aerosol chemical composition and hygroscopicity would require the combination of remote sensing observations with real-time, vertically resolved aerosol chemical composition measurements. Furthermore, Raman lidar measurements revealed high aerosol hygroscopicity in the vicinity of clouds, suggesting that aerosol hygroscopicity may facilitate particle activation as cloud condensation nuclei under favourable thermodynamic and dynamical conditions. Additional studies using numerical weather prediction models capable of accurately reproducing both clouds and the evolution of aerosol hygroscopicity are needed.

The text in Lines 700–705: Data availability of the original manuscript has been revised as follows:

Revised manuscript: Lines 767–773.

RALMO data can be accessed through the EARLINET network (`https://data.earlinet.org/earlinet/`), while ceilometer and Doppler wind lidar data can be obtained via the E-PROFILE network (`https://data.ceda.ac.uk/badc/eprofile/data/switzerland/payerne`). Co-author Alexander Haefele can be contacted for access to raw RALMO and radiosonde measurements (alexander.haefele@meteoswiss.ch). Sun photometer data can be retrieved from the AERONET network (`https://aeronet.gsfc.nasa.gov/`). In situ aerosol data can be requested from co-author Benjamin T. Brem (benjamin.brem@psi.ch), while chemical composition data from the NABEL station can be accessed at `https://www.bafu.admin.ch/en/data-query-nabel`. The HYSPLIT backward-trajectory files and corresponding maps are available for download from Zenodo (`https://doi.org/10.5281/zenodo.23010868`; Díaz-Zurita et al., 2026).

The text in Line 715: Acknowledgements of the original manuscript has been revised as follows:

Revised manuscript: Lines 778 and 780.

The order of the funding projects in the Acknowledgements section has been revised. Grant PID2024-162154OB-I00 is now listed on line 778, while Grant PID2021-128008OB-I00 has been moved to line 780.

Revised manuscript: Line 782.

The grant number has been added: COST Action EARLICOST (CA24135).

References

The following references have been added to the revised manuscript:

Revised manuscript: Lines 787–789.

Aksoyoglu, S., Keller, J., Barmpadimos, I., Oderbolz, D., Lanz, V. A., Prévôt, A. S., and Baltensperger, U.: Aerosol modelling in Europe with a focus on Switzerland during summer and winter episodes, Atmos Chem Phys, 11, https://doi.org/10.5194/acp-11-7355-2011, 2011.

Revised manuscript: Lines 814–815.

Bohren, C. F. and Huffman, D. R.: Absorption and Scattering of Light by Small Particles, Wiley, https://doi.org/10.1002/9783527618156, 2007.

Revised manuscript: Lines 816–819.

Bozzetti, C., Daellenbach, K. R., Hüglin, C., Fermo, P., Sciare, J., Kasper-Giebl, A., Mazar, Y., Abbaszade, G., Kazzi, M. E., Gonzalez, R., Shuster-Meiseles, T., Flasch, M., Wolf, R., Křepelová, A., Canonaco, F., Schnelle-Kreis, J., Slowik, J. G., Zimmermann, R., Rudich, Y., Baltensperger, U., Haddad, I. E., and Prévôt, A. S.: Size-Resolved Identification, Characterization, and Quantification of Primary Biological Organic Aerosol at a European Rural Site, Environ Sci Technol, 50, https://doi.org/10.1021/acs.est.5b05960, 2016.

Revised manuscript: Lines 869–870.

Díaz-Zurita, A., Navas-Guzmán, F., and Pérez-Ramírez, D.: Five-day NOAA HYSPLIT backward trajectories for hygroscopic aerosol layers over Payerne, https://doi.org/10.5281/zenodo.23010868, 2026.

Revised manuscript: Lines 918–919.

Hale, G. M. and Querry, M. R.: Optical Constants of Water in the 200-nm to 200-μm Wavelength Region, Appl Opt, 12, 555–563, https://doi.org/10.1364/AO.12.000555, 1973.

Revised manuscript: Lines 920–921.

Hermann, M., Stratmann, F., Wilck, M., and Wiedensohler, A.: Sampling characteristics of an aircraft-borne aerosol inlet system, J Atmos Ocean Tech, 18, https://doi.org/10.1175/1520-0426(2001)018<0007:SCOAAB>2.0.CO;2, 2001.

Revised manuscript: Lines 961–962.

Liu, Y. and Daum, P. H.: Relationship of refractive index to mass density and self-consistency of mixing rules for multicomponent mixtures like ambient aerosols, J Aerosol Sci, 39, https://doi.org/10.1016/j.jaerosci.2008.06.006, 2008.

Revised manuscript: Lines 1007–1008.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos Chem Phys, 7, https://doi.org/10.5194/acp-7-1961-2007, 2007.

Revised manuscript: Line 1012.

Prahl, S.: miepython: Pure Python calculation of Mie scattering, https://doi.org/10.5281/zenodo.7949264, 2026.

Revised manuscript: Lines 1016–1017.

Rolph, G., Stein, A., and Stunder, B.: Real-time Environmental Applications and Display sYstem: READY, Environ Model Softw, 95, https://doi.org/10.1016/j.envsoft.2017.06.025, 2017.

Revised manuscript: Lines 1020–1028.

Rovira, J., Savadkoohi, M., Chen, G. I., Močnik, G., Aas, W., Alados-Arboledas, L., Artiñano, B., Aurela, M., Backman, J., Banerji, S., Beddow, D., Brem, B., Chazeau, B., Coen, M. C., Colombi, C., Conil, S., Costabile, F., Coz, E., de Brito, J. F., Eleftheriadis, K., Favez, O., Flentje, H., Freney, E., Gregorič, A., Gysel-Beer, M., Harrison, R., Hüglin, C., Hyvärinen, A., Ivančič, M., Kalogridis, A. C., Keernik, H., Konstantinos, G., Laj, P., Liakakou, E., Lin, C., Listrani, S., Luoma, K., Maasikmets, M., Manninen, H. E., Marchand, N., dos Santos, S. M., Mbengue, S., Mihalopoulos, N., Nicolae, D., Niemi, J. V., Norman, M., Ovadnevaite, J., Petit, J. E., Platt, S., Prévôt, A. S., Pujadas, M., Putaud, J. P., Riffault, V., Rigler, M., Rinaldi, M., Schwarz, J., Silvergren, S., Teinemaa, E., Teinilä, K., Timonen, H., Titos, G., Tobler, A., Vasilescu, J., Vratolis, S., Yttri, K. E., Yubero, E., Zíková, N., Alastuey, A., Petäjä, T., Querol, X., Yus-Díez, J., and Pandolfi, M.: A European aerosol phenomenology – 9: Light absorption properties of carbonaceous aerosol particles across surface Europe, Environ Int, 195, https://doi.org/10.1016/j.envint.2024.109185, 2025.

Changes to the Supplement

1. Sect. S1: campaign backward trajectories are presented in Fig. S1. Fig. S2 and explanatory text have been added to document the air-mass origins for the three case studies. The revised manuscript now refers to Fig. S2a, S2b, and S2c in Lines 548, 600–601, and 640, respectively.

2. Sect. S2: Fig. S3 provides the particle-size comparison discussed in Lines 452–459 of the revised manuscript.

3. Sect. S3: Table S1 provides the dry refractive indices used in the Mie calculations; Fig. S4 provides the corresponding SMPS size distributions.

4. Sect. S4: Fig. S5 presents the backward trajectories and satellite fire detections for the 8 December 2019 case, supporting the discussion in Lines 516–519 of the revised manuscript.

5. 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 supplementary figures provide the additional case-study observations and vertical distributions.