Response to Referee #2 Comments

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

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General comment

The authors characterise the aerosol hygroscopicity using several independent methods for six months in a Swiss site. A good agreement in hygroscopicity retrieval was found between Raman lidar, radiosondes and tandem nephelometer. They claim that aerosol hygroscopic growth is primarily driven by inorganic aerosol species. While the methodology is technically sound, the main message of the manuscript is not clear. The whole manuscript reads like a lengthy descriptive report rather than a research article. Key details are missing, such as back trajectory analysis. The authors need to decide what are the most important aspects and put more emphasis on them in the main text, when moving the rest to the supporting information. Therefore, the manuscript will require substantial revision for a few rounds before being considered for publication.

Response

We sincerely thank the referee for this important general assessment, which has helped us improve the structure and focus of the manuscript. We agree that, in the original version, the main scientific contribution was not sufficiently distinguished from the supporting description of the campaign observations. Although the HYGRO19 campaign provided a particularly comprehensive multi-instrument framework for investigating aerosol hygroscopicity, the hierarchy and specific roles of the different datasets were not sufficiently clear. We have therefore substantially revised the manuscript to focus more clearly on the main objective of the study and to clarify the role of the different measurements and analyses. 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).

The main objective of the revised manuscript is to investigate how complementary remote sensing and in situ techniques can be combined to characterise aerosol hygroscopicity under ambient atmospheric conditions and to assess the consistency and limitations of the resulting multi-instrument comparisons. The study brings together continuous Raman lidar observations, vertically resolved balloon-borne measurements, and ground-based tandem nephelometry. These approaches provide different perspectives on aerosol hygroscopicity, while differences in the optical properties measured, wavelengths used, sampling conditions, and vertical representativeness define the scope of the comparisons.

Within this multi-instrument framework, the RAman Lidar for Meteorological Observations (RALMO) provides the core dataset. It is one of the few instruments worldwide capable of providing high-vertical-resolution profiles of aerosol backscatter, temperature, and water vapour mixing ratio during both daytime and nighttime (Dinoev et al., 2013), making it particularly suitable for aerosol hygroscopicity studies. Its continuous operation throughout the six-month HYGRO19 campaign provided more than 200 hygroscopic aerosol layers, enabling a statistical characterisation of the temporal and vertical variability of aerosol hygroscopicity under different atmospheric conditions.

The HYGRO19 campaign also provided the opportunity to evaluate the RALMO hygroscopicity parameter retrievals using complementary approaches. Vertically resolved aerosol measurements

from the Compact Backscatter Aerosol Detector (COBALD), operated together with Vaisala RS41 radiosondes, allowed direct comparison between RALMO retrievals and balloon-borne in situ observations, making this one of the few studies in which aerosol hygroscopicity has been characterised vertically using balloon-borne measurements. Ground-based tandem nephelometer observations provided an additional comparison with near-surface aerosol hygroscopicity. These comparisons are central to the study and are interpreted in light of the different quantities and aerosol populations sampled, rather than as a quantitative validation of equivalent parameters. Additional aerosol and meteorological measurements provide supporting information for interpreting the observed hygroscopic behaviour.

We have also clarified the role of the surface chemical measurements. These observations characterise near-surface aerosol composition but cannot be assumed to represent elevated or decoupled aerosol layers. Accordingly, we have revised the discussion to avoid attributing the hygroscopicity retrieved within those layers directly to surface chemical composition.

To address the missing information on air-mass origin, we have added a backward-trajectory analysis for the investigated hygroscopic aerosol layers, with methodological details and supporting figures in the Supplement and trajectory files in a dedicated repository. This analysis is described in detail in our response to Major comment # 3. In addition, preliminary surface-based Mie simulations provide a physical baseline for assessing how differences in wavelength and optical property affect the interpretation of the multi-instrument comparisons.

To reduce the descriptive content of the main manuscript, Figs 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. The Abstract (Lines 2–18), Introduction (Lines 63–88), and Conclusions (Lines 633–699) have also been revised and shortened to focus more clearly on the main findings.

The following changes have been introduced in the manuscript:

1. We have substantially revised and shortened the **Abstract (Lines 2–18 of the original manuscript)** to focus more clearly on the main findings and better reflect the scope and conclusions of the study. The revised Abstract can be found in **Lines 2–21 of the revised manuscript**.

2. The text in **Lines 63–66 of the original manuscript** has been revised as follows (**Lines 67–71 in the revised manuscript**): 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.

3. The text in **Lines 67–80 of the original manuscript** has been revised as follows (**Lines 72–88 in the revised manuscript**): 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.

4. The text in **Lines 81–87 of the original manuscript** has been revised as follows (**Lines 89–98 in the revised manuscript**): 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.

5. The text in **Sect. 2.1: The HYGRO19 field campaign (Lines 94–98 of the original manuscript)** has been revised as follows (**Lines 105–113 in the revised manuscript**): 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.

6. We have substantially revised the **Conclusions section (Lines 633–699 of the original**

manuscript). The revised section can be found in **Lines 708–766 of the revised manuscript.**

Major comment # 1

Lines 298-300: I do not get how the claim "in agreement with AOD-AE AERONET measurements" is made. In Fig 1 b, the time dominated by fine-mode particles is June 1st – September 7th. In Fig 2 b, the time dominated by fine-mode particles is the entire period from August to January. Whether the two instruments agree or not require a robust evaluation via statistics not by eyes.

Response

Thank you for this comment. We agree that the statement "in agreement with AOD-AE AERONET measurements" was not sufficiently clear in the original text and that the corresponding statistical values should be provided to support this interpretation. Our intention was to indicate that both the scattering Ångström exponent (SAE (450–635 nm)) measured by the dry nephelometer and the extinction Ångström exponent (AOD-AE (440–870 nm)) obtained from the AERONET sun photometer generally indicate a predominance of fine particles during the campaign, although periods with a greater contribution of coarse particles were also observed. The AOD-AE values for the two campaign periods were reported in **Lines 279–286**, while the SAE values for the entire campaign and the two separate periods were provided in **Lines 294–300 and Table 1 of the original manuscript**. To clarify the comparison, we have revised the paragraph and included the median AOD-AE value for the entire campaign.

The following changes have been introduced in the manuscript:

1. The text in **Lines 298–301 of the original manuscript** has been revised as follows (**Lines 316–322 in the revised manuscript**): 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).

Major comment # 2

Lines 332-338: The definition of decoupled aerosol layers is very confused. More details and discussions are required. How about the time periods around December 1st? The β was quite strong and extended to free troposphere.

Response

We appreciate the reviewer's comment. We agree that the meaning of "decoupled aerosol layers" was not sufficiently clear in the original manuscript. We have revised the text to clarify that this

term refers here to aerosol layers observed above and decoupled from the ABL. Examples of these elevated layers were observed on 2–5 and 17–19 July. We have also revised the description of the September–December period, which was too general in the original manuscript. As indicated by the reviewer, an episode of relatively enhanced aerosol backscatter extending into the free troposphere was observed around 1 December.

The following changes have been introduced in the manuscript:

1. The text in **Lines 334–338 of the original manuscript** has been revised as follows (**Lines 359–363 in the revised manuscript**): 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.

Major comment # 3

Line 341: No significant changes in air-mass origin? Please provide the back trajectory analysis and indicate the time!

Response

Thank you for pointing this out. Following Navas-Guzmán et al. (2019), potential changes in air-mass origin with altitude should be considered in lidar-based hygroscopicity studies. We agree that our original statement was not sufficiently supported, as the backward-trajectory analysis and the corresponding trajectory times were not provided. To address this comment, 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 (version 5.4.0). The trajectories were initiated at the time of the corresponding RALMO observation and at the lower and upper boundaries of each layer. All available HYSPLIT backward-trajectory files and corresponding maps are now provided in a repository (`https://doi.org/10.5281/zenodo.23010868`). Note that some hygroscopic aerosol layers may share the same backward trajectories depending on the availability of the HYSPLIT input data.

A detailed quantitative comparison of the trajectories arriving at the lower and upper boundaries is now provided in the **Supplement (Sect. S1 and Fig. S1)**. Specifically, we evaluate the same-time horizontal separation between the two trajectories and their spatial proximity. The results show that the air masses arriving at different heights within the investigated hygroscopic aerosol layers generally follow similar paths during the first 72 h. Larger differences and greater dispersion are observed during the 72–120 h period. As discussed in the Supplement, this increasing divergence may be partly related to the increasing uncertainty of numerically calculated backward trajectories with integration time (Stohl, 1998). In addition, an air mass represents a horizontally extensive body of air rather than an individual trajectory (American Meteorological Society, 2000); therefore, spatial separation between individual trajectories may not necessarily indicate a change in air-mass origin.

We have revised the manuscript and Supplement, accordingly, as detailed below:

1. The text in **Lines 340–342 of the original manuscript** has been revised as follows (**Lines 365–371 in the revised manuscript**): 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).**

2. The following analysis has been added to **Sect. S1 of the Supplement**:

   S1 Analysis of five-day NOAA HYSPLIT backward trajectories

   To assess potential changes in air-mass origin with altitude, 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 (version 5.4.0). The trajectories were initiated at the time of the corresponding RALMO observation and at two heights corresponding to the lower and upper boundaries of each layer. For each case, the same-time separation and spatial proximity were calculated using the Haversine distance (Sinnott, 1984). The same-time separation represents the median horizontal distance between the base and top trajectories at the same backward time (e.g., base and top positions at −12 h). In contrast, the spatial proximity accounts for differences in the progression along the trajectories by determining the nearest spatial distance between the two paths, independently of backward time. This calculation was performed in both directions (base-to-top and top-to-base), and the larger of the two median distances was used as the spatial-proximity metric. A direct comparison between these metrics is shown in Fig. S1, where each point represents one investigated hygroscopic aerosol layer. The horizontal and vertical bars indicate the corresponding half interquartile range (± IQR/2).

   The results from Fig. S1 suggest that the air masses arriving at different heights within each investigated hygroscopic aerosol layer over Payerne generally follow similar paths during the first 72 h, as indicated by the relatively small same-time separation and spatial proximity values (Fig. S1a). At longer backward times (72–120 h), larger values and greater dispersion are observed (Fig. S1b). This increasing divergence may be partly related to the increasing uncertainty of numerically calculated backward trajectories with integration time (Stohl, 1998). It should also be considered that an air mass represents a horizontally extensive body of air rather than an individual trajectory (American Meteorological Society, 2000); therefore, spatial separation between individual trajectories may not indicate a change in air-mass origin. All HYSPLIT trajectory files and their corresponding maps are now available from Zenodo (`https://doi.org/10.5281/zenodo.23010868`; Díaz-Zurita et al., 2026).

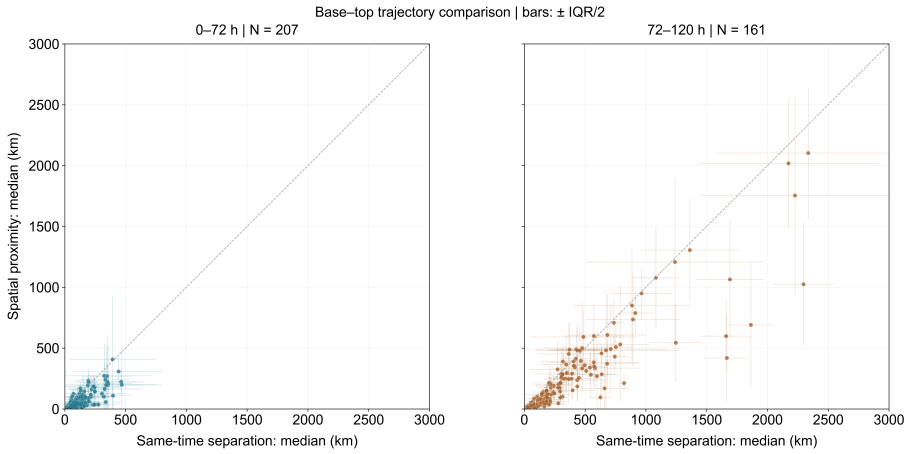


Figure S1: Comparison of five-day NOAA HYSPLIT backward trajectories for the base and top of each investigated hygroscopic aerosol layer during (a) 0–72 h and (b) 72–120 h backward time.

3. In addition, Fig. S2 and the following text have been added to Sect. S1 of the Supplement:

   The five-day HYSPLIT backward trajectories for the three case studies discussed in the main manuscript are shown in Fig. S2. For each case, the analysis shows that the air masses arriving at different heights within the hygroscopic aerosol layer generally follow similar paths.

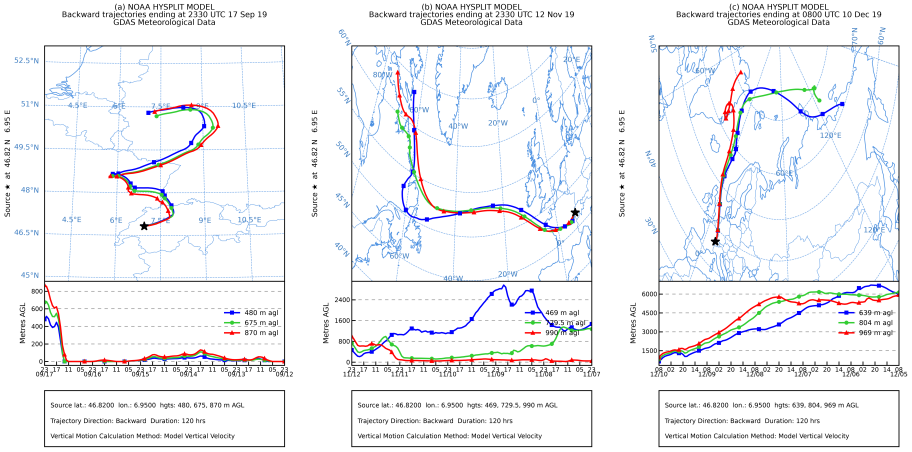


Figure S2: Five-day backward trajectories from the NOAA HYSPLIT model for (a) Case I, ending at 23:30 UTC on 17 September 2019; (b) Case II, ending at 23:30 UTC on 12 November 2019; and (c) Case III, ending at 08:00 UTC on 10 December 2019.

The corresponding references to these trajectories in the case-study sections have also been revised:

- The text in **Line 506 of the original manuscript** has been revised as follows (**Line 548 in the revised manuscript**): (backward trajectories shown in Fig. S2a in the Supplement).
- At **Line 558 of the original manuscript**, we have added the following text (**Lines 600–601 in the revised manuscript**): (backward trajectories shown in Fig. S2b in the Supplement).
- The text in **Line 596 of the original manuscript** has been revised as follows (**Line 640 in the revised manuscript**): (Fig. S2c in the Supplement).

4. To further clarify the criterion referred to in the original statement "no significant changes in air-mass origin" in **Sect. 3.4 (Lines 252–272 of the original manuscript)**, we have added the following text 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)" **(Lines 256–259 of the original manuscript)**: 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. **(Lines 275–277 in the revised manuscript)**

5. Finally, the following sentence has been added to the **Data availability section (Lines 772–773 of the original manuscript):** 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). **(Lines 772–773 in the revised manuscript)**

Minor comment # 1

Lines 36-40: Could you specify what aerosol properties can be modified during sampling? What are the large certainties under conditions close to saturation?

Response

Thank you for this comment. We have clarified the aerosol properties that can be modified during sampling, including particle number concentration, size distribution, chemical composition, scattering, and absorption. We have also quantified the uncertainties associated with f(RH) measurements at high RH. According to Titos et al. (2016), the uncertainty in $f(RH)$ is approximately 20–40 % for moderately hygroscopic aerosols. Accordingly, **Lines 36–40 of the original manuscript** have been revised as follows: 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). **(Lines 40–45 in the revised manuscript)**

Minor comment # 2

Line 50: What certain layers?

Response

Thank you for this point. We have clarified that "certain atmospheric layers" refers to well-mixed aerosol layers **(Line 50 of the original manuscript)**. Accordingly, the text has been revised as follows: 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). **(Lines 52–54 in the revised manuscript)**

Minor comment # 3

Lines 301-304: Could you please provide the HYSPLIT or any back trajectory analysis to indicate where the emissions were from? Just citing papers without showing the detail is irresponsible!

Response

We appreciate the reviewer's comment. We would like to clarify that the origin of the emissions mentioned in this paragraph was not inferred from a back-trajectory analysis performed in this study. Rather, the statement refers to the typical sources and transport pathways previously reported for Payerne in the cited literature (Minguillón et al., 2012; Dada et al., 2025). We agree that the original wording could be interpreted as an attribution based on our own analysis. Therefore, we have revised the sentence in **Lines 303–304 of the original manuscript** to explicitly indicate that this information comes from previous studies, as follows: 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. **(Lines 324–327 in the revised manuscript)**

Minor comment # 4

Lines 305-306: How do you define low and high contribution of absorbing aerosols? The range spans two orders of magnitude, from 0.06 to 26 Mm^{-1}.

Response

We appreciate the reviewer's observation. We agree that the wide range of absorption coefficients reflects substantial temporal variability and that the description "a relatively low contribution of absorbing aerosols" was insufficiently precise. Our intention was to describe the generally low absolute absorption coefficients in the context of a regional background site, rather than to infer the relative contribution of absorbing particles from the absorption coefficient alone. As reported by Rovira et al. (2025), Payerne is a regional background site and generally exhibits lower absorption coefficients than suburban, urban, and traffic sites. We have clarified this interpretation in the revised manuscript **(Lines 328–330)** and included the corresponding reference.

Revised text: 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 $\pm$ 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).

Minor comment # 5

Lines 307-309: Aerosol absorption coefficient and σ might point to the same thing, absorbing aerosols. Could you please include the range of aerosol absorption coefficient in the brackets? Otherwise, it is very confused that σ was used to indicated absorbing particles just right after aerosol absorption coefficient (in Mm^{-1}) was mentioned.

Response

Thank you for this suggestion. We clarify that σ_{ab} is the symbol used throughout the manuscript to denote the aerosol absorption coefficient and does not represent a different parameter. This notation was already used in Fig. 2c, but its definition in the text was not sufficiently explicit. We have explicitly identified the scattering and absorption coefficients as σ_{sp} and σ_{ab} in the description of Fig. 2 (Lines 305–306 of the revised manuscript). The values subsequently given in parentheses (σ_{ab}, SSA, and AAE) correspond to the optical properties used to characterise and compare the two periods.

Minor comment # 6

Line 309: What correlative analyses?

Response

Thank you for pointing this out. Here, the term "correlative" was used to refer to the interpretation of related carbonaceous aerosol measurements rather than to a statistical correlation analysis. To avoid this ambiguity and better connect this analysis with the preceding discussion, we have revised the sentence in **Lines 309–311 of the original manuscript** accordingly: The analysis of carbonaceous measurements further suggests that these changes can be associated with a higher frequency of local wood burning (Fig. 2g–h). **(Lines 333–334 in the revised manuscript)**

Minor comment # 7

Lines 314-315: Provide the range from the literature.

Response

We appreciate this comment. We have now included the range of organic aerosol contributions previously reported for Payerne. Although the reported fractions are lower than those obtained during HYGRO19, previous studies also show an important contribution of organic aerosol and a larger contribution during summer. We have revised the text in **Lines 311–315 of the original manuscript** as follows: 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) **(Lines 334–340 in the revised manuscript).**

Minor comment # 8

Line 330: How was 1.5 km defined as the cut off height for the atmospheric boundary layer height?

Response

We appreciate the reviewer's comment. The 1.5 km cut-off height was defined based on the ABL climatology at Payerne reported by Collaud Coen et al. (2014). This reference was already included in the manuscript when discussing the ABL at **Lines 339 and 368**, but we inadvertently omitted it at this point. We thank the reviewer for pointing this out. We have now added the corresponding reference.

Revised text: 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). **(Lines 355–356 in the revised manuscript)**

Minor comment # 9

Line 345: Occurrence of what?

Response

We appreciate the reviewer's comment. The "frequency of occurrence" refers to the occurrence of hygroscopic aerosol layers. To make this clearer, we have revised the sentence as follows **(Lines 374–375 in the revised manuscript): Most hygroscopic layers were observed in summer (177 hygroscopic layers, 78 %),**

Minor comment # 10

Lines 365-367: Could you provide more context about these established statistical rules?

Response

We appreciate the reviewer's suggestion and agree that providing some additional context may be helpful to the reader **(Lines 395-398 in the revised manuscript)**: ... 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.

Minor comment # 11

Line 413: I will just use winter instead of second part.

Response

Thank you for raising this point. We have retained the term "second part of the campaign", as this period covers September–December and therefore includes autumn and only one month of winter (December).

Minor comment # 12

Lines 415-420: I disagree with not showing the figure. The figure needs to be included into the supporting information. Also include a discussion on why the size dependence is weak in light of the literature.

Response

Following the reviewer's suggestion, the corresponding figure has now been included in the Supplementary Information **(Sect. 2 and Fig. S3)**. We have also expanded the discussion of the relationship between particle size and aerosol hygroscopicity. Although particle size can influence hygroscopic growth, our results suggest no clear dependence on particle size, with the differences between fine and larger particles not being statistically significant. 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., 2014; Jefferson et al., 2017; Sicard et al., 2022). The text in **Lines 414–420 of the original manuscript** has been revised accordingly: 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., 2014; Jefferson et al., 2017; Sicard et al., 2022). **(Lines 453-459 in the revised manuscript)**

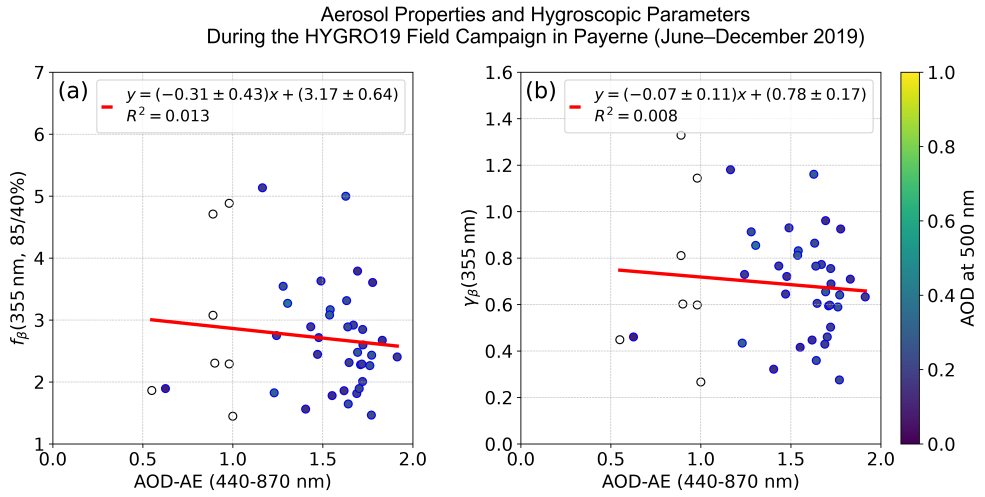


Figure S3: Relationship between the extinction Ångström exponent (AOD-AE, 440–870 nm) and the hygroscopic parameters (a) f_β(355 nm, 85/40 %) and (b) γ_β(355 nm) during the HYGRO19 campaign. The colour scale represents the Aerosol Optical Depth (AOD) at 500 nm. Red lines represent the linear regressions, with the corresponding equations and coefficients of determination (R^2) shown in each panel. White markers indicate cases for which no AOD measurements were available.

Minor comment # 13

Figure 7: Could you use direction instead of degree so that the information can be easily linked to the corresponding paragraph?

Response

Thank you very much for this suggestion, which allows the wind-direction information to be more easily interpreted and directly linked to the corresponding discussion in the manuscript. We have modified Fig. 7 in the manuscript.

Minor comment # 14

Figure 8: I am not sure if any finding can be drawn from the limited RALMO data points.

Response

Thank you for this comment. We agree that the number of RALMO observations during this period is limited. As already stated in the manuscript, only seven hygroscopic aerosol layers were identified between October and December, mainly due to the frequent presence of clouds, fog, and precipitation. Following the reviewer's comment, we have now also explicitly stated that the limited number of available RALMO cases should be considered when interpreting the comparison between both techniques. The text 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.

Minor comment # 15

Lines 477-479: Where are the HYSPLIT analysis and map of fire hotspots?

Response

Thank you for pointing this out. The HYSPLIT backward-trajectory analysis and the fire hotspot information were originally provided through their corresponding online sources rather than as figures. Following the reviewer's comment, we have now included a figure in the Supplementary Information showing the five-day HYSPLIT backward trajectories on 8 December 2019 **(Fig. S5a)**, together with the corresponding fire hotspot map **(Fig. S5b)**. The manuscript **(Lines 477–479 in the original manuscript)** has also been revised to explicitly refer to this supplementary figure as follows **(Lines 516-519 in the revised manuscript)**: 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).

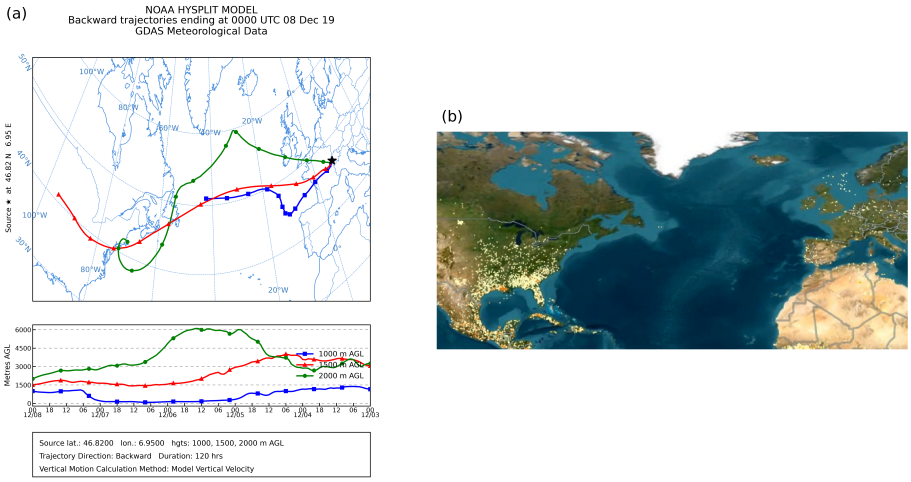


Figure S5: (a) Five-day backward trajectories from the NOAA HYSPLIT model ending at 00:00 UTC on 8 December 2019 at 1000, 1500, and 2000 m agl over Payerne. (b) Fire hotspot map from the NASA/NOAA Suomi National Polar-orbiting Partnership for the period from 4 to 8 December 2019 (`https://firms.modaps.eosdis.nasa.gov/`, last access: 29 September 2026).

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