Answers to Referees

We would like to thank all the referees for their constructive suggestions and comments. We believe that the manuscript has improved thanks to their recommendations. In the following text we report the referees' comments in gray and the answer to the referees in black. Modified manuscript text is reported in blue.

Referee #1

The paper presents an interesting analysis of aerosol properties measured at a high elevation site in the Alps. The analysis is sound and interesting, and the aerosol data collected at the site can certainly add novel and unique information for the complex Alpine environment. The hope is that these aerosol measurements will continue and be augmented in the future. Overall, I think the paper can be published with rather minor revisions.

We would like to thank the referee for the positive comments.

General comments

- Briefly describe the inlet and potential losses for the aethalometer

We agree with the referee about the need to add information about the AE33 sampling line. We added a description of the inlet used for the AE33 measurements and derived the inlet efficiency based on von der Wedien et al., 2009. The manuscript was modified as follows:

Aerosol is sampled through a heated sampling head with no size cut. The stainless-steel sampling line has a half-inch diameter, resulting in an inlet efficiency greater than 99% for particles smaller than 3 μm, which decreases to 90% at 8 μm (van der Weiden et al., 2009), i.e. well above the mode of long-range transported dust.

- I would guess that during the sampling, there might have been periods with low-level clouds. Did this affect the measurements? The authors screened the data by the RH, so I would think eventual cloudy periods were removed, but some mention would still be useful.

We would like to thank the referee for this comment. We screened the data based on the relative humidity recorded by the OPC inside the measuring device (internal RH), to remove interferences due to the detection of particles that were not completely dried before measurements. The manuscript has been revised to clarify this point as follows:

Relative humidity (RH) is continuously monitored inside the sampling line and measurement corresponding to RH larger than 40% were discarded from analysis (1.8% of datapoints) to prevent inaccuracy caused by the detection of wet particles. Measurements were not filtered for low-level clouds, so during cloudy periods the measured aerosol included both interstitial particles and cloud residuals from the evaporation of small cloud droplets.

- The hypothesis of mixing between black carbon and dust particles is interesting. However, that begs the question of how the mixture was formed. Is it reasonable to believe that the mixture would form in the atmosphere through collision during transport or near the site, even if the particle concentration is low? Or are these mixtures forming closer to the source, like dust emitted very close to combustion sources (either human in nature or due to biomass burning)?

We thank the referee for this comment. Measurements suggest that long-range transported dust at Testa Grigia was mixed with black carbon. However, we lack evidence to determine whether dust and BC were internally or externally mixed or where this mixture formed. We are working to implement mixing state measurements at the observatory and to integrate these data with LAGRANTO back-trajectory analysis and CAMS dust and black carbon models. This will enable testing of the internal mixture hypothesis and identification of mixing regions. The paragraph concerning dust and BC mixing in the conclusion section was modified as follows:

Aerosol absorption coefficient measurements, available for only part of the observational period, clearly indicate that the dust reaching the observatory is mixed with black carbon. Nevertheless, evidence is insufficient to conclude whether dust is internally or externally mixed with black carbon and the regions where this mixture formed.

- This is not necessarily a request for changes, but the paper focuses a lot on dust, while the title does not mention dust. I wonder if a specific mention of dust in the title would make the link more immediate.

We agree with the referee that mentioning dust in the title of the manuscript would make the link more immediate, thus the title was modified as:

First Continuous Aerosol Measurements at Testa Grigia at 3480 m asl: Aerosol Populations and Dust Transport Dynamics in the Southern European Alps

Specific comments

In the last sentence of the abstract: "The seasonal variability of PM10 concentration associated to SDE is explained by the variability to dust emission regions, dust mobilization over source region, and efficiency of dust transport mechanisms." I wonder if the authors meant "sensitivity to dust emissions…" instead of "variability to…". If they indeed meant "variability" then it should read "variability of"?

We replaced variability with "sensitivity to"

Opening sentence of the introduction. I believe that some aerosols can also have radiative effects from interactions with thermal radiation.

The sentence is corrected and now reads:

"Atmospheric aerosols play a crucial role in the climate system due to their ability to absorb and scatter shortwave and longwave radiation"

Line 96: How were these potential contamination periods identified?

The approach used to remove data points corresponding to local pollution is described in section 2.2, so we added a reference to this section on line 96.

Line 97: How representative is a locally measured windrose of the main air transport path?

Local windrose is representative exclusively of local wind circulation, so it does not represent long-range air transport path. The sentence was modified to be clearer:

"Windrose from 1-minute time resolution data (Fig. S1b) indicates that the prevailing local wind circulation is from south-west and from north-east, with no specific day/night pattern."

Lines 101-103: Are the numbers after the plus/minus sign standard deviations?

The referee is correct. The plus/minus sign indicates the standard deviation. To avoid confusion, we specify it adding the "SD" annotation:

"Temperature at Testa Grigia is characterized by a clear seasonality, with higher values in summer (June - August average was equal to 3.3°C plus/minus 3.3°C SD) and lower in winter (December - February average was equal to -9.8°C plus/minus 5.0°C SD), while the diurnal temperature range does not show significant seasonal differences, with annual average equals to 6.6°C plus/minus 2.3°C SD(Fig. S1c)."

Line 103: "equals" should be "equal".

Corrected

Line 110: Have inlet losses been calculated? If so, what is the upper size cut-off?

We estimated the inlet efficiency based on van der Weiden et al. 2009. This information is now added to the text:

"The inlet efficiency is estimated to be larger than 99% up 9 μm and to decrease to 92% at 10 μm (Von Der Weiden et al., 2009)."

Line 113: Maybe clarify why they were discarded, for clarity.

We discarded data potentially affected by local contamination to be sure to describe the background atmospheric composition. This is now clearly reported in the manuscript:

"To ensure that measurements were representative of background atmospheric composition, we excluded potential contamination from local sources in the OPC data at 1-minute time resolution according to the algorithm developed by Beck et al. (2022)."

The specific reference to parametrization employed has now been removed because it is specified in the manuscript, as explained in the following response.

Line 114: Briefly explain Beck's correction scheme here to avoid the need for reading the cited paper to have a rough idea of the method.

We added the following description to the text:

Briefly, local pollution was identified by an increase in the first derivative of the data series that exceeded 1.7 times the running interquartile range calculated over 24 hours. Following, a filter on the median was applied to flag measurements that were 1.4 times higher than the running median calculated over one hour.

Line 143: Please provide a URL.

Reference is now corrected and reports the website address

"Database Centro Funzionale Regione Autonoma Valle d'Aosta: https://presidi2.regione.vda.it/str_dataview_station/1720, last access: May 2023."

Line 151: How is the "predominantly aerosol-free atmosphere" being identified? In other words, what's the threshold?

We thank the reviewer for the opportunity to clarify this point. The manuscript now explicitly defines the threshold used: the "predominantly aerosol-free atmosphere" is identified as the region above the Continuous Aerosol Layer (CAL). The CAL is defined as the layer where the total backscatter signal, retrieved from ALC measurements as described in Bellini et al. (2024), exceeds the molecular-only backscatter for at least 98% of its vertical extension (Bellini et al., 2025). Above this altitude, the signal drops to molecular levels, effectively separating the boundary layer from the free troposphere. The manuscript has been modified as follows:

"The CAL is operationally identified as the layer where the total backscatter, retrieved from ALC measurements as described by Bellini et al. (2024), exceeds the molecular-only backscatter for at least 98% of its vertical extension (Bellini et al., 2025). Above this altitude, the signal drops to molecular levels, enabling the separation of boundary layer aerosols from free tropospheric conditions."

Line 154: How good is the spherical approximation, especially for dust?

Volume calculation using the spherical assumption is incorrect for a dust-dominated aerosol population, due to the irregular shape of dust particles. Nevertheless, this is a common assumption in aerosol models to simplify volume and mass calculation. We modified the text to explicitly acknowledge this source of uncertainty.

"The average particle volume size distribution was calculated on an hourly basis assuming spherical shape for computational simplicity. We acknowledge that this assumption may introduce uncertainties in dust-dominated populations where irregular particle shapes prevail."

Line 161-162: Why is that? Why was that exact size cut chosen?

This size cut was selected because it most closely approximates a 10 μm diameter. Additionally, the instrument's inlet efficiency drops below 90% for larger particles, and the low abundance of these particles makes their counting not representative of real ambient

conditions when measurements are performed at high-time resolution. The text was modified accordingly. The text was modified accordingly:

"For cluster analysis, we focused solely on particle numbers measured in the first 22 size bins, which correspond to optical diameters smaller than 9.4 μm. This size cut was selected because it most closely approximates a 10 μm diameter. Additionally, the instrument's inlet efficiency drops below 90% for larger particles, and the low abundance of these particles makes their counting not representative of real ambient conditions, especially when measurements are performed at high-time resolution".

Line 168: How is the "how low as possible" number being determined?

Ideally, to maximize the similarity of the objects within each cluster might lead to have clusters composed by one single object. We understand that this sentence can be misleading, and it has been removed. The text now reads:

"The optimal number of clusters was determined by maximizing the similarity among the elements within each cluster"

Line 257: What is the reason for the low capture rate?

We thank the referee for this comment. We realized that absorption coefficient data capture in Figure 1 is an underestimation of real values because it is calculated based on the time when both particle number size distribution and aerosol absorption coefficient measurements are available. We corrected the figure, and the new data capture is larger for the months when the instrument was operating. We modified the text as follows:

As black carbon time series is characterized by several gaps due to instrumental failure, seasonal variability is not discussed.

Figure 2: Is there a reason for using a 0 to 10 vertical scale on panel c. Panel a. ranges from 0 to 30, while panel b. ranges from 0 to 10, so I don't see a reason to keep the same scale in panel c., and zooming in would allow for a better view of the details.

Figure 2 has been modified according to the referee's suggestion.

Line 308: It would be nice to add some of these webcam pictures, at least in the SI.

A new figure has been added to the supplementary material and supplementary figure numbering has been modified accordingly.

Line 322: Are the AAE estimates precise enough to allow differentiating between 1.56 and 1.48? Also, why would a lower AAE indicate slightly darker particles? Is the assumption that a slightly lower AAE would indicate a larger contribution from black carbon?

We agree with the referee that the precision of the measurements, especially at low absorption coefficients, is not enough to differentiate between 1.56 and 1.48. We rounded the AAE values to the first decimal digit throughout the manuscript and in Table 1. The comment about the lower AAE refers to the fact that we expect this aerosol to absorb more uniformly across the

visible spectra. Nevertheless, the difference between the AAE values is within the standard deviations, and the comment about the difference has been removed. Now the text reads:

"…which are indicative of slightly higher absorbing particles enriched in black carbon"

Line 327: This share is different from that provided in line 325; is that because this refers to winter only?

The referee is correct, this share refers to the winter months. We modified that text as follows to make it clearer:

"Cluster 3 is observed mainly in winter months (December – February), when its occurrence frequency was about 50%."

Line 330: Does the low absorption necessarily indicate the negligible impact of absorbing aerosol, or just reflecting low concentrations?

We agree with the referee that this sentence is inaccurate. It has been modified as follows:

"Cluster 3 is the one characterized by the lowest average aerosol absorption coefficient ($Babs_{880}$ = 0.35 plus/minus 0.37 Mm^{-1}), in agreement with the lowest aerosol loading."

Figure 5: It might be useful to add a title on top of each column (cluster 1, 2, and 3), respectively, for more immediate readability.

Figure 5 has been modified accordingly.

Line 501: For direct comparability, provide the actual number from the Jungfraujoch

The number of events has been reported in the text.

Line 543: An "a" is missing after "19" and before "severe".

Corrected

Line 590: Please provide a range of values from the published work from the Jungfraujoch you are referring to here for more direct comparability.

The following sentence has been added to the text:

"From 2017 to 2023 this site experienced on average more than 100 SDE hours in February, March, April and June and more than 75 hours in October (Collaud Coen et al., 2025)"

Line 639: The lower wet removal in the upper troposphere is reasonable and expected, but why would the gravitational settling be affected by the elevation? Is it just because being higher up, the particles take a longer time to settle to the ground, or are there some other processes at play (such as effects of shape or orientation) implied here?

Gravitational deposition has been removed.

Line 632: High-resolution, in what sense? I guess in time?

We now specify “high-time-resolution”.

Lines 644-659: I am confused by the two sentences that are connected by “on the contrary”. Why “on the contrary”, if both are larger in summer?

We agree with the referee. “On the contrary” has been removed.

Line 663: Here, it is a bit less critical, but I find the “on the contrary” to be potentially confusing here as well, considering that both cluster 1 and 2 seem to have in common at least the large-scale transport (although from different regions).

“On the contrary” has been removed.

Line 669: Type 1 is the same as cluster 1? If so, I would stay consistent with the rest of the paper.

“Type” has been replaced by “cluster”

Lines 680-681: See one of my previous comments on this topic. Can the authors comment on how reasonable it is (in terms of the probability of collision between dust and BC particles, and given the particle concentrations) that the dust-BC mixture would “form” at the receptor site?

Available measurements do not enable us to draw conclusions about the type of mixing state (internal or external mixture) or the region where the mixture formed. This topic will be the subject of a future study. The text was modified as follows:

“Nevertheless, evidence is insufficient to conclude whether dust is internally or externally mixed with BC and the regions where this mixture formed.”

Referee #2

General comments:

The paper presents high-time-resolution measurements of aerosol size distribution and absorption at in Italian Alps, over the period September 2021–May 2023. The author applied a clustering method based on the aerosol volume size distribution shape to identify three main aerosol types. This, together with trajectory analysis and comparison with CAMS, has been shown to capture the influence of boundary layer dynamic, long range transport and Saharan dust event.

I found the manuscript to be well written and clearly structured. The methodology is presented in a clear way, and the figures are generally well designed. While the study does not introduce fundamentally new concepts, it provides a solid and well-supported analysis of aerosol transport mechanisms based on a robust dataset.

We would like to thank the referee for the general positive comments.

Technical comments:

Line 91: Typo in “measurements”.

The typo has been corrected

Equation 2: It seems that a factor Dp^3 could be missing.

We would like to thank the referee for pointing out this mistake. The equation 2 has now been corrected:

$$n_V(D_p) = \frac{\pi}{6} D_{p\,bin}^3 \, n_N(D_p) \quad (2)$$

Lines 55–57: It is not entirely clear how this statement connects to the results presented later in the manuscript.

We agree with the referee that this statement does not relate to the results presented later in the paper. It is meant to underline the relevance of high elevation measurements to capture changes in background atmospheric composition. We modified the paragraph as follows:

Mountaintop observatories represent an essential tool for the continuous monitoring of aerosol properties and composition variability at high elevations (Andrews et al., 2011; Collaud Coen et al., 2020), enabling the analysis of daily (Nyeki et al., 1998a; Shaw, 2007), seasonal (Nicolás et al., 2018; Gallagher et al., 2011; Singh et al., 2020; Sellegri et al., 2010), and long-term atmospheric composition changes (Collaud Coen et al., 2013). They also allow the detection of long-term trends in natural and anthropogenic emissions at the hemispheric scale (Collaud Coen et al. 2020).

Lines 50–68: This part of the introduction would benefit from a clearer structure. It appears that the intention is to highlight the relevance of mountain sites for studying aerosol transport, but the cited conclusions seem somewhat disconnected from the rest of the paper. Can you clarifying the main message of this section to strengthen the introduction?

We modified the two paragraphs indicated by the referee to make them clearer. The first paragraph has been modified as described in the previous comment and the second paragraph has been changed as follows:

“Furthermore, observatories located at high elevations provide a unique opportunity to sample the free troposphere, allowing for the investigation of long-range transport dynamics, frequency, and potential trends. For instance, measurements of particle number size distribution at Monte Cimone, in the northern Apennines, allowed the analysis of long-term impact of long-range Saharan dust transport events over distances greater than 1000 km and revealed that the frequency of dust transport days in the study region ranged from 15% to 20% over the past 20 years, with no significant trend observed over time (Vogel et al., 2025; Duchi et al., 2016). Similarly, aerosol optical properties measured at Jungfraujoch, in the western Alps, indicated that Sahara dust transport episodes reached the Alps between 30 and 150 times per year during the last 23 years (2001-2024) (Collaud Coen et al., 2025). In addition to dust, high-

elevation observatories allowed the study of transport of wildfire plumes in the upper troposphere and lower stratosphere at hemispheric scale (Masoom et al., 2025; Dzepina et al., 2015; Laj et al., 2009)."

Line 99: For clarity, does this refer to the integrated frequency of winds exceeding 8 m/s, from North (90°) to East (0°)?

We agree with the referee that the sentence is ambiguous. We modified the text to make it clearer.

Wind speed higher than 8 m/s was recorded for 35% of the measurement period; within this subset, the prevailing direction was from North to East.

Line 175: It is not entirely clear how the additional pressure levels (10, 30, and 50 hPa below surface pressure) are used. Are these additional trajectory releases? If so, how are the resulting trajectories combined for interpretations, is it an integration of them all?

The analysis integrates all the trajectories. This is now specified in the manuscript.

The analysis integrates all the calculated trajectories to mitigate the uncertainty derived from the relatively coarse resolution of the model.

Line 220: Missing parenthesis.

Parenthesis added

Line 224: Duplicate citation of Nyeki et al. (inside and outside parentheses).

The format of citations has been corrected.

Line 225: The definition of "coarse particles" is repeated. Unless intentional for some reason, it would be sufficient to define it once.

Removed

Figure 5: I would recommend using a different colormap. The current one is not perceptually uniform and may introduce visual artifacts in the interpretation. A sequential colormap (e.g. viridis or plasma) may be more appropriate.

The maps have been re-plotted using the "viridis" color scheme.

Line 467: The plots also suggest that, for Cluster 3, a larger fraction of air masses originates from higher altitudes compared to the other clusters.

The referee is correct. This is clearly indicated in the following paragraph, where we specified that back trajectories associated to cluster 3 spent 63% of the time in the free troposphere.

Figure S2: A wind rose representation may be more appropriate for this type of data

Figure S2 has been modified according to the referee's suggestion.

Line 489: Typo in "Saharan dust".

Corrected

Line 498: The approach combining air mass history and aerosol size distribution to identify Saharan dust events is well established in the literature. The use of cluster analysis instead of fixed thresholds is a useful refinement, but it may be more appropriate to present it as an alternative implementation rather than a fully novel methodology.

We replaced “innovative” with “alternative”

Line 505: The comparison between observations and CAMS is interesting, but it relies on different definitions of dust, which may partly explain the discrepancies. For example, it is stated “In contrast, hourly data points classified as SDE in the observations but associated with lower PM10 were not correctly identified by CAMS (green markers in Fig. 6a)”, but you also used a threshold definition of those CAMS dust events which may also cause part of the mismatch.

We thank the referee for this comment. In this paragraph we intended to underline that the comparison strongly rely on the definition of dust events. We agree that the sentence can be confusing, and the paragraph has been modified as follows:

Hourly data points classified as SDE in the observations but associated with lower PM10 were not correctly identified by CAMS (green markers in Fig. 6a), due to the limitation of the criteria used to identify CAMS dust events.

Figure 7 (panels b and c): The colorbar values are difficult to read. It would be better to increase their size and also include units directly on the colorbar rather than only in the caption.

Panel b and c of Fig.7 have been replaced with new figures with a larger font size.

Figure S3: Why does the log10 of the frequency reach values up to 3?

Figure S3 (now Figure S4) reports counts and not frequency values. The legend of the color scale has now been corrected.

Line 671: Typo (“This means…”).

Corrected

In addition to the referees’ suggestions, the following corrections have been implemented:

Line 102: “north-east” instead of “north to east” (north to east could be misinterpreted)

Line 112: “0.25 μm and 1 μm” instead of “0.3 μm and 1 μm” (the measurement range starts at 250 nm and not 300 nm)

Line 126: “has been” instead of “is”

Line 143: “rates” instead of “rate”

Line 185: “served” instead of “serves”

Line 271: “February” instead of “March”

Line350. Removed “particularly” to avoid repetition

Line 553: “Alps” instead of “alps”

Line 664: “500 nm” instead of “300 nm”

Replaced PM10 with PM_{10} throughout the manuscript

Label position in Figure 4 was modified to make it consistent with other figures