Response to Anonymous Referee #3

The paper analyses 3 cases of transport from the Po Valley to NW Alps with several in-situ (4 sites in Aosta and in pristine environment) and REM instruments from ground and space. All these measurements are extensively documented in the paper and in the supplement, allowing to have a broad view of the pollution events. After having validated the meteorological model COSMO, a chemical transport model (FARM) is evaluated. This paper is a clear documentation of middle range pollution events affecting the Alps and the evaluation of model's performances under these conditions is very interesting.

We thank the reviewer for taking the time to revise our manuscript and for his/her pertinent comments. Our reply to these is given hereafter (the text in italics represents a citation of the revised manuscript and the figure references follow the updated numbering).

Referee's comment 1. Figures do not always allow to verify the descriptions or conclusions of the study. For example, higher aerosol concentrations are measured during the case studies (Fig. 4e, Fig. 8, Fig. 13e Fig. S8, ...), but the figures do allow to be sure that the increase in aerosol load is really specific and cannot be attributed to usual/local fluctuations. In other words, it is not possible to see the differences between the period of the case studies and the period without influence from the Po Valley. I do understand that figures similar to Fig. 4 cannot display longer period of time, but another solution should be found! The time period covered by Fig. 12 is fine.

Author's response 1. Following this comment, and similar remarks from referee #2 (RC2) and referee #4 (RC6), we expanded (and homogenised) former panels 4, 10 and 13 (old numbering) to include the same number of days. At the same time, we also paid attention at introducing into the sequence a "clear" day in order to better show the effect of the advections. We thus opted to show one week of measurements in each of these figures, as a compromise between completeness and detail (e.g., of the wind velocity field). The new plots extend from 25 to 31 August 2015 (case 1), from 24 to 30 January 2017 (case 2) and from 24 to 30 May 2017 (case 3), respectively. This allows to better appreciate the difference between event- (clear) and non-event (polluted) days.

Moreover, for each episode, the information on the respective seasonal average concentrations were added to the text to provide reference values. Also note that a rigorous assessment of the long-term impact of the phenomenon presented in this part 1 is indeed the purpose of our companion paper (Diémoz et al., 2019) based on a statistical analysis of the complete dataset (2015–2017).

RC2. FARM leads to too low PM10 concentration in summer and in spring, but seems to work better (even if it does not reproduce the diurnal cycle correctly) in winter (case study 2). Is there an explanation for this difference between summer and winter?

AR2. We recognise that the results of case study 2 could give the mistaken impression that FARM works better in winter. However, this episode is not representative of a more general behaviour, as thoroughly explained in our companion paper. The latter specifically address the long-term evaluation of the phenomenon described in this "part 1" of the study. We anticipate here Fig. 17 of the companion paper, showing that the discrepancies between simulated and observed PM_{10} concentrations at the surface are larger in winter compared to the other seasons. This is to ascribe to an underestimation of the PM_{10} emission sources from outside the boundaries of the domain (in the second row, the comparison looks better if this external contribution is multiplied by a factor W=4).

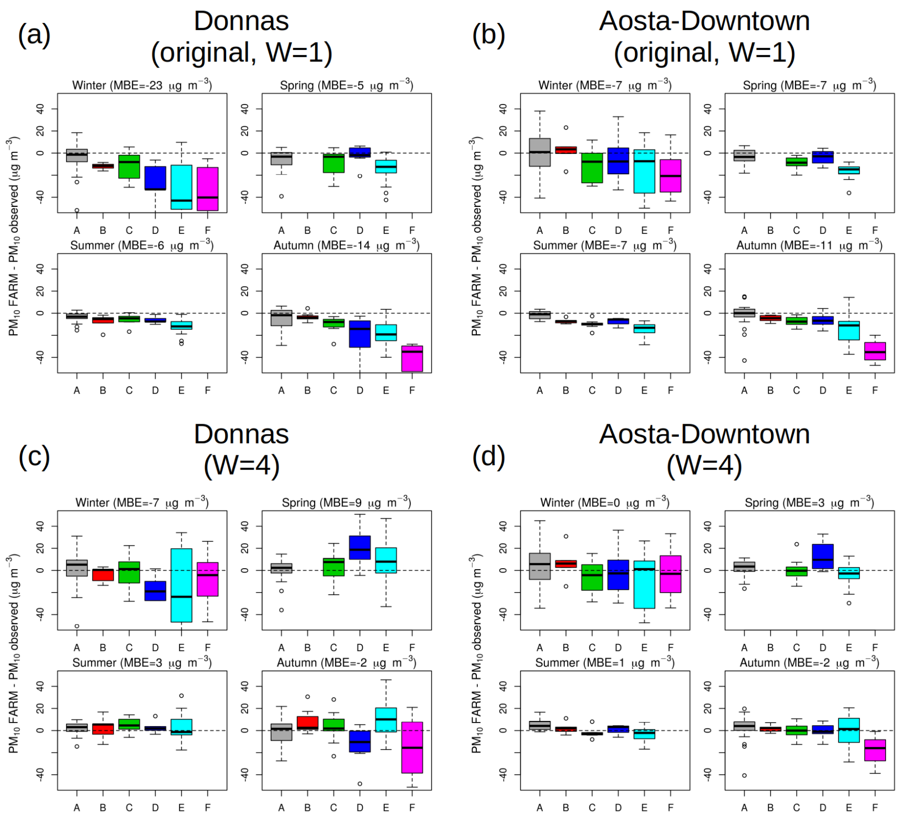

Figure 17 (companion paper): Differences between simulated and observed PM_{10} concentrations at the surface. The mean bias error (MBE) for each case is reported in the plot titles. First row: FARM simulations as currently performed in ARPA for the Donnas (a) and Aosta–Downtown (b) stations. Second row: the PM_{10} concentrations from outside the boundaries of the domain were multiplied by a factor W=4.

Section 4.4 of the revised manuscript was slightly modified to anticipate this issue: *The emission inventory used within FARM likely underestimates the real emissions, as also reported in other cases, and for different models, in the scientific literature (e.g., EMEP, 2016; Uchino et al., 2017). In particular, the boundary conditions could not be accurate enough for our aims owing to the abrupt change of the national emission inventory grid resolution (12 km) to the local scale (1 km). This issue can affect the comparison between the model (dry* PM_{10}*) and surface measurements, especially in winter, as discussed more extensively in the companion paper (Diémoz et al., 2019).*

RC3. Why FARM regularly has time shift in its estimation (for example in Fig. 13 for case study 3)? Fig. S10 shows that modification of the PM concentration from the boundary conditions and the modification of the hygroscopic growth have no impact on the time of the aerosol increase.

AR3. The reviewer raised an important issue, also mentioned by referee #4 (RC11). We therefore further investigated this aspect, added a figure (S13 in the Supplementary) and updated Sect. 4.4 to include a discussion of this phenomenon:

As already mentioned in the description of the three cases, the model: a) underestimates the PM mass both in the layer aloft and at the surface, and b) anticipates the peak concentrations compared to the profiles from the ALC. A variety of (possibly concurrent) reasons can explain the observed underestimation [...] The previous considerations, however, fail at comprehensively explaining the time shifts sometimes noticeable between the model and the measurements, i.e. anticipation of the advection arrival time (even in "dry" conditions in the afternoon on the first day of each sequence) and of the layer disappearance in the morning (where hygroscopicity may have an important role). Although an accurate assessment would require a more sophisticated set of instruments to characterise the vertical profile of the wind velocity, here we formulate some hypotheses:

1. *the NWP model likely anticipates and overestimates the easterly thermally-driven winds in the first hours of the afternoon. This is noticeable, for example, in Fig. S13, where the zonal component of the wind from both COSMO and the surface measurements for case study 1 (August 2015) is plotted, and, on a longer statistical basis, in Fig. S1(b,c), showing that the model has the tendency to see easterly winds more often and with higher intensity compared to the observations. A possible reason for that is the smoothed valley orography used in the NWP model compared to the real one. This is displayed in Fig. S14, showing the difference of the Digital Elevation Model (DEM) used within COSMO and a more realistic DEM (10 m resolution): both valleys and mountain crests are clearly smoothed out by COSMO, with absolute differences well* > 500 *m (and up to 1000 m). This difference could additionally explain why the altitude of the entrainment zone (i.e., the boundary between the free atmosphere and the boundary layer where the thermally-driven circulation develops) is underestimated by COSMO compared to the height of the aerosol layer detected by the ALC;*
2. *COSMO overestimates the nighttime drainage winds (katabatic winds), as noticeable, again, from Fig. S13 for case study 1. This might trigger enhanced cleansing of the lower atmospheric layers during the night as simulated by FARM (see, e.g., the supplementary video file,* `https://doi.org/10.5446/38391`*), but undetected by the ALC. An overestimation of the drainage winds would also explain the differences between the simulated and measured daily cycle of specific humidity [...]*

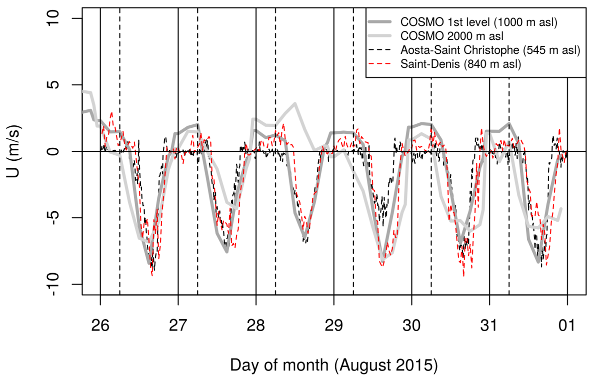

Figure S13: Zonal component of wind velocity during episode 1 (August 2015) from COSMO (1000 and 2000 m a.s.l.) and two surface stations (Aosta–Saint Christophe and Saint-Denis). Positive U represent wind from the west, negative U wind from the east.

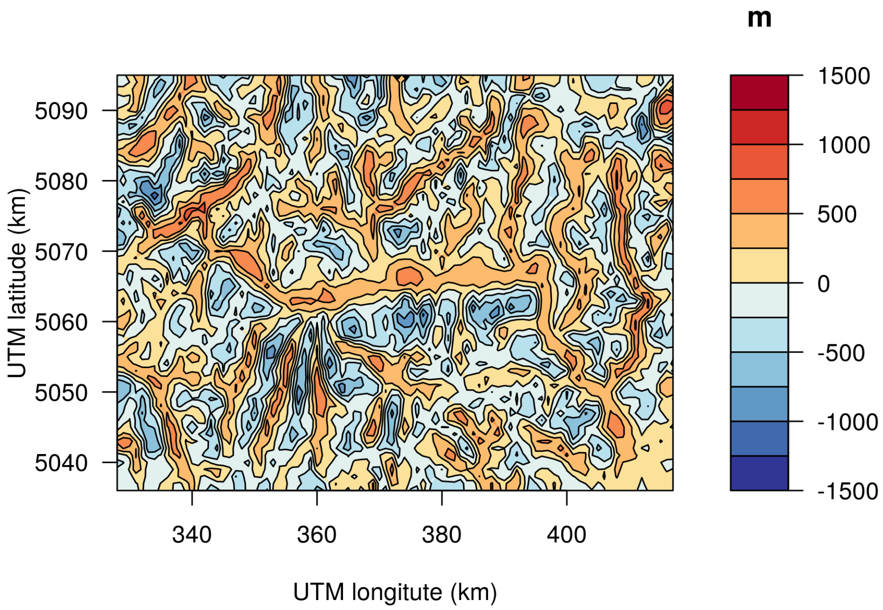

Figure S14: Difference between the (smoothed) Digital Elevation Model (DEM) used by COSMO-I2 (2.8 km resolution) and a higher-resolution DEM ("real topography", 10 m resolution).

Conclusions were also updated to underline this issue:

Our investigation allowed an evaluation of the FARM model. Notably, FARM could reproduce the observed arrival of elevated aerosol layers and it correctly attributed them to sources external to the Aosta Valley. However, absolute values of PM concentrations and the timing of the advections were poorly reproduced, with underestimations of aerosol concentrations and time anticipations compared to the measurements. On the basis of a sensitivity study, the former issue may be partly attributed to both water uptake by highly-hygroscopic particles, not fully taken into account in the model, and deficiencies in the emission inventories, especially owing to the coarse resolution of the national one (12 km). As for the timing discrepancies, suboptimal performances of the NWP model to simulate daytime (thermally-driven) and *nighttime (katabatic) winds are the most likely sources of error. Despite these limitations, FARM brought insights on the phenomenology addressed, supporting the observations and helping to interpret them. On the other hand, the observation-based results of this work could drive the improvement of the emission inventories, thus enhancing the reliability of the CTM (e.g., Diémoz et al., 2019). In turn, this could allow extending the findings of this work to a wider domain, not covered (or not fully covered) by observations.*

RC4. P. 1 line 14: "and hygroscopic": the begin of the sentence is a comparison, so that the meaning of this last 2 words is not obvious.

AR4. Thank you for pointing this out. The sentences now reads: *Results also indicate that the aerosol advected from the Po Valley is hygroscopic, smaller in size and less light-absorbing compared to the aerosol type locally-emitted in the northwestern Italian Alps.*

RC5. P. 1–2, lines 22–1: for clarity purpose I would add a TO: "likely owing to deficiencies in the emission inventory and TO particle water uptake not fully taken into account"

AR5. Done.

RC6. Figure 4d + p. 19 lines 2–7: 1) "PM concentration from non-local sources is represented by the coloured background and the effect of local sources by the contour line, at logarithmic steps;" The logarithmic steps are hardly visible on Fig. 4d (I had to discover that they exist on Fig. 10 to find them on Fig. 4). Could you perhaps use a color scheme with a legend? Without this, it is not evident that the non-local sources exceed the local ones.

AR6. To address this problem highlighted by the referee, we modified the figures in the following way: a) we made the lines thicker; b) we used different line styles (rather than an additional colour legend) and reported the corresponding concentrations in a new legend. An example is provided below, in Fig. 9 (panel d).

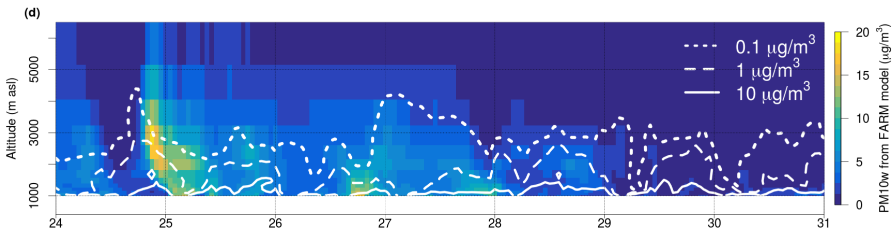

Figure 9 (panel d): Case study of 24–30 January 2017. (d) Mass concentration (PM_{10w}) from FARM. PM concentration from non-local sources is represented by the coloured background and the effect of local sources by the contour line, at logarithmic steps (dotted: 0.1 $\mu g\,m^{-3}$; dashed: 1 $\mu g\,m^{-3}$; continuous, near the surface: 10 $\mu g\,m^{-3}$).

RC7. P. 16 line 11: delete the "," after "scattering ratio" + give some indication on the altitude of the described layer.

AR7. Done, the sentence was modified as follows: *The appearance of the PBL layer is clearly noticeable on 26 August as an increase in the backscatter coefficient up to an altitude of 3 km a.s.l., with scattering ratios SR≃* 4 *at midday (light-blue area in the figure) almost doubling (SR>* 8*, orange-yellow) in a few hours.*

RC8. P. 16 lines 12-19: I do not really understand your description: from the end of 26th of September, the lowest layer measured by ALC already shows a SR of about 6, so that the "lower levels, with potential consequences on the surface air quality" are already impacted before sunrise on the 27th of September. Your description does not really describes why the SR decrease in the middle of the days. Is the aerosol rich layer entering the ML and contributes then to higher aerosol load in this ML (not observed in Fig. 4a), is it dispersed to higher altitude due to the thermal convection or dispersed horizontally? Please clarify.

AR8. We believe that the reviewer is referring here to the case of August 26-27 (not September). Indeed, we clearly stated in the text that *a thick aerosol layer is detected by the ALC over the Aosta–Saint Christophe observatory from the afternoon of 26 August*, thus well before the sunrise of August 27, and further specified that *the appearance of the PBL layer is clearly noticeable on 26 August as an increase in the backscatter coefficient up to an altitude of 3 km a.s.l., with scattering ratios SR≃* 4 *at midday (light-blue area in the figure) almost doubling (SR>* 8*, orange-yellow) in a few hours.*

With respect to the missing explanation of the SR decrease in the middle of the day, this was because the interpretation of this ALC-observed behaviour was left to the following paragraphs. However, following the referee's comment, we modified the relevant sentence as follows:

On 27 August, the ALC backscatter is then observed to decrease in the central part of the day and to increase again in the afternoon. This behaviour keeps very regular for almost a week, with the aerosol-rich layer extending from ground up to 3–3.5 km. As further discussed in the next paragraphs, we anticipate here that the main factor driving this cycle is likely an enhanced hygroscopic growth of aerosol advected from the Po Valley from the afternoon to the early morning, this effect also leading to formation of low clouds within the aerosol layer at night (screened out as white areas in the figure). In fact, the transition from aerosol to the cloud phase is very sharp, as also noticeable from the sudden increase of more than 40 Wm^{-2} *of the downward infrared irradiance monitored at the same site.*

RC9. P. 19: is it possible that the observed shift (from some hours to 1/4 of a day) of the maximum concentration between FARM and ALC can be due to an overestimation of the local effects in comparison to the non-local ones?

AR9. Local effects can indeed impact on the daily evolution of the concentrations. However, we believe that the shift observed in this case is mainly triggered by suboptimal simulations of the wind fields by the NWP model, as explained at point AR3 of the present document.

RC10. Figure 8 and p. 6 lines 18–20 + p. 20 lines 13–15: the measured PM2.5 and PM10 during the case study are similar to the annual mean concentration (p.6). An increase of PM2.5 and PM10 is clearly visible from the 26th of August to the beginning of September (at least at Aosta), but the readers cannot be sure that this does not correspond to a usual fluctuations of the aerosol load. Similarly, it is not really visible that FARM predicts an increase of the PM10 concentration at Aosta during the case study.

AR10. Following this comment, we modified the relevant sentence as follows: *An increase in daily concentrations (up to maximum values of 10* $\mu g m^{-3}$ *for* $PM_{2.5}$ *and 16–22* $\mu g m^{-3}$ *for* PM_{10}*) can be clearly noticed at both sites, leading to concentrations slightly higher than average for the same period (7* $\mu g m^{-3}$ *for* $PM_{2.5}$ *in Aosta–Downtown and 12* $\mu g m^{-3}$ *for* PM_{10} *at both sites, considering the 2015–2017 series). The statistical significance of such PM increases during these transport episodes, compared to the natural variability of the aerosol load in non-advection conditions, is assessed in the companion paper using the full long-term dataset.*

Moreover, as already mentioned in AR1, the seasonal average concentrations were added to the text to provide reference values.

RC11. Fig. 4e + page 20 lines 8–9: the PM10 daily cycle does not correspond to the Po Valley air masses transport seen by the ALC. The given explanation relates to the mass losses occurring in TEOM due to secondary aerosol volatility. Is it expected that this volatility should be different between nighttime and daytime (i.e. larger during the night) in order to explain the different diurnal cycles?

AR11. This is an important remark. We therefore decided to update the manuscript to discuss this issue in further detail.

Section 3.1.5: *Two Tapered Element Oscillating Microbalance (TEOM) 1400a monitors (Patashnick and Rupprecht, 1991) are used for continuous measurements of* PM_{10} *hourly concentrations at the stations of Aosta–Downtown and La Thuile. These instruments are not compensated for mass loss of semi-volatile compounds (Green et al., 2009) and could be insensitive to specific compounds, such as ammonium nitrate (e.g., Charron et al., 2004), which leads to underestimations, especially in the cold season, compared to the SM200. Conversely, overestimations by the TEOM compared to daily averages from the SM200 reference instrument are found in summer and are not fully understood at present. Therefore, TEOM monitors are only employed here for qualitative estimates of short-term variations of the aerosol burden while daily-averaged concentrations will be only taken from the SM200 instruments.*

Section 4.1.3: *To evaluate the impacts on surface air quality parameters during the episode, hourly* PM_{10} *concentrations at the surface as measured in Aosta–Downtown and simulated by FARM in Aosta–Saint Christophe are presented in Fig.3e (*PM_{10} *monitoring at La Thuile was not yet operational, at that time). Apart two spikes (80* $\mu g m^{-3}$*) on 25 and 26 August (presumably of local origin), the concentrations measured in Aosta–Downtown by the TEOM show a slight increase after the arrival of the layer, but without sudden jumps. Also, PM concentrations are generally higher during daytime compared to the night, according to the expected cycle of the summertime local sources (e.g., traffic, resuspension, etc.). This features, however, can be connected to the fact that mass loss occurs in TEOM due to secondary aerosol volatility, as better discussed in the companion paper by comparing the daily* PM_{10} *cycle from this instrument and the Fidas OPC in Aosta–Saint Christophe. Moreover, this volatility could be different between nighttime and daytime, which would also contribute to the observed daily behaviour. Besides, FARM estimates at the surface are again lower than measurements (-60%, on average). Daily* PM_{10} *concentrations observed by Opsis SM200 instruments during the case study in Aosta–Downtown and Donnas are shown in Fig. 7, which includes the whole episode (correlation index with TEOM measurements* $\rho = 0.84$*).*

RC12. Fig. S5 is cited before Fig. S4

AR12. Corrected, thank you.

RC13. P. 25 line 2: it has however to be mentioned (and perhaps explained) that the wind direction at Saint-Denis (Fig. S4e), after turning to the east during the afternoon of the 26th of January, turns again to west on the 27th in the morning and stay globally at west during the rest of the case study.

AR13. To better show the different flow regimes during this episode, we made some integrations to the analysis. First, wind direction was plotted in Fig. S4 only for cases when wind speeds exceeds 1 $m\,s^{-1}$ to exclude calm wind conditions. Secondly, we further considered the wind measurements in Donnas, where the wind regime change is much clearer, owing to the weaker temperature inversion. Table 1 (used instruments) was modified accordingly.

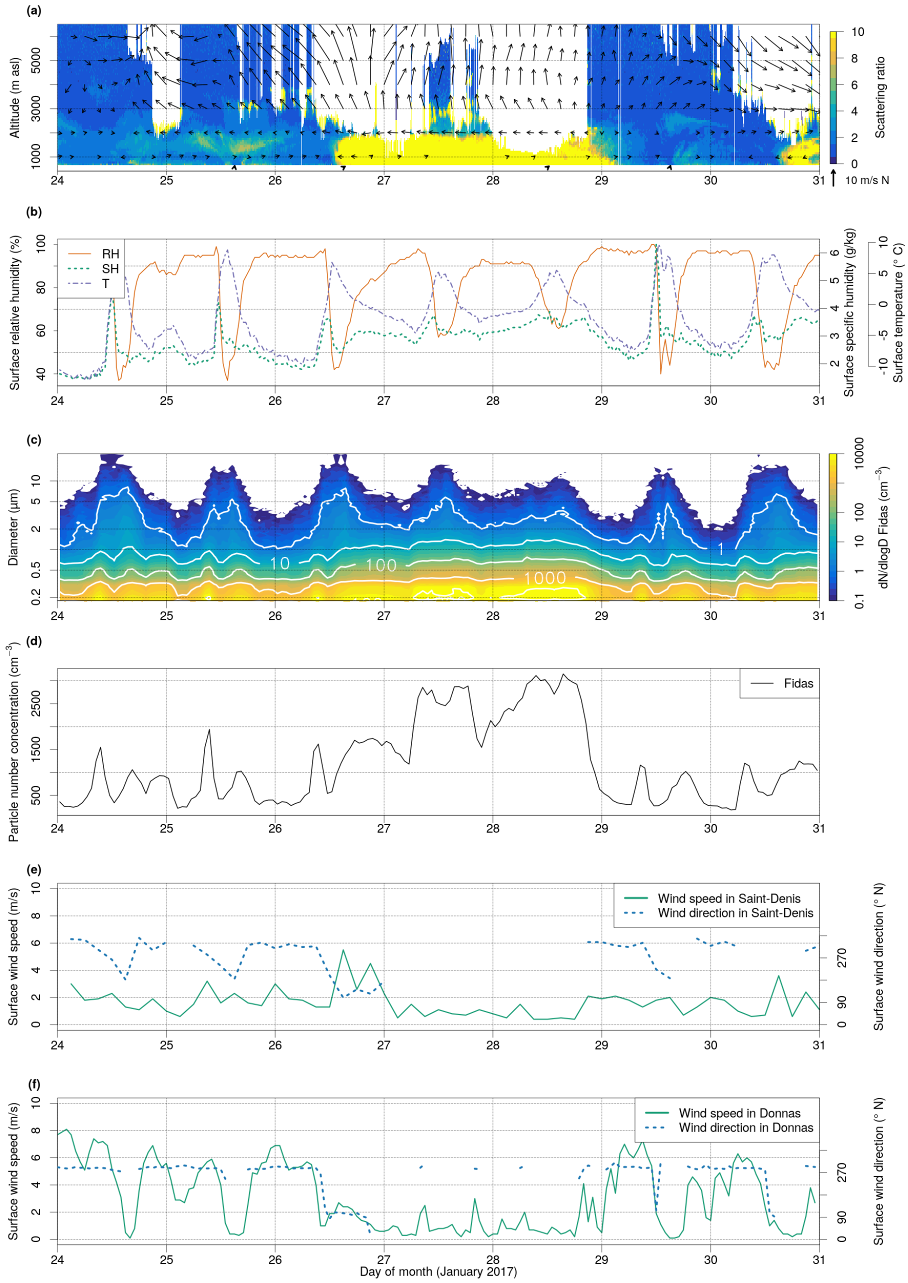

Figure S4: Case study of 26–29 January 2017. (a) Same as in Fig. 9 in the main paper; (b) Surface relative humidity, specific humidity and temperature measured at the Aosta–Saint Christophe weather station; (c) Particle number distribution from the Palas optical counter; (d) Particle number concentration (sum of all channels) from the Palas optical counter; (e) Wind speed and direction at the Saint-Denis station (800 m a.s.l); (f) Wind speed and direction at the Donnas station (316 m a.s.l). No data from the photometer are available for the selected period, since the instrument was not operating.

Given these changes, Sect. 4.2.2 now reads:

The wind field over Aosta–Saint Christophe, depicted in Fig. 9a, presents a very different pattern compared to the first case addressed (Fig. 3a). Firstly, calm wind is measured for the whole period at the bottom of the valley. This is due to a shallow temperature inversion in the lower atmospheric layers in the main valley. Conversely, at the Saint-Denis station, located above the inversion layer, and at the Donnas station, where the temperature inversion is weaker, the wind pattern is more representative of the wider circulation: for example, the average wind speed in Saint-Denis is about 4 $m\,s^{-1}$ on 26 January afternoon (Fig. S4e) and *the wind clearly turns from west (morning) to east (afternoon), simultaneously with the appearance of the layer. The same wind change is detected in Donnas on the same day (Fig. S4f), with easterly wind speeds* $> 1\ ms^{-1}$ *for several hours in the afternoon. As a further difference with the first case, the forecasted wind at 1000–2000 m a.s.l. does not show any change in direction typical of the thermal winds. For example, at 2000 m a.s.l. the circulation is continuous, and vigorous (up to 6* ms^{-1}*), from the afternoon of 26 to the beginning of 29 January. Indeed, this winter case study interestingly shows that thermally-driven winds are not the only mechanism, especially in winter, driving the advection of air masses from the Po Valley to the Alps. Rather, the synoptical circulation can push the air masses towards the Alpine valleys, as in this case. In fact, the flow clearly reveals its southern origin at elevations above the mountain crest (e.g., 3000 m a.s.l.), where the wind is not channelled within the main valley. At that altitude, the wind speed is even greater than 20* ms^{-1}*. Finally, on 29 January, the measurements in Saint-Denis (gradual increase of the speed of westerly wind) and in Donnas (even stronger wind, again from the west), and COSMO simulations (wind reversal at 1000–2000 m a.s.l) correlate with the disappearance of the layer better than observations performed at the bottom of the valley (calm wind). [...]*

RC14. P. 25 lines 10–12: the increase of the wind speed at Saint-Denis on the 29th is not very large (about 2 m/s on the 26th in the morning, increase to 4 m/s on the 26th in the afternoon, about 1 m/s until the end of the 28th, then 2 m/s), so that the correlation with the disappearance of the layer is not really explicit.

AR14. Please, refer to AR13.

RC15. Fig. S6 + p. 25 lines 13–15: the described transition (for the lowest levels I suppose) is quite difficult to see: too much backtrajectories, the lowest ones being represented under the highest ones. A figure allowing to see not only the trajectories but also the change of the altitude over the Po Valley (lines 15–29) would perhaps be more interesting).

AR15. Following this comment, and the ones by referee #1 (RC3) and #4 (RC7), we modified the back-trajectory figures in the following way: a) we plotted in separate panels trajectories ending at altitudes < 2000 and > 2500 m a.s.l. over Aosta–Saint Christophe; b) to further simplify the figures, we only show back-trajectories for specific times corresponding to the most significant variations of circulation patterns; c) for each time selected we added a bottom panel with the trajectory altitude along their journey. An example of the new plots now included in the paper is provided in Fig. 5.

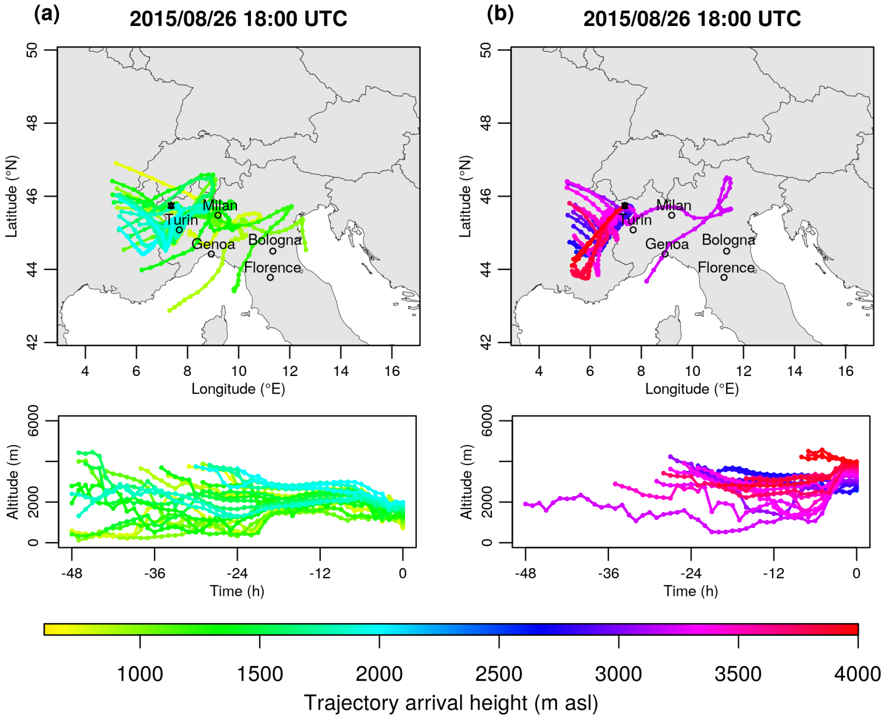

Figure 5: 48-hours back-trajectories ending at Aosta–Saint Christophe on 26 August 2015 at 18 UTC at altitudes lower than 2000 m a.s.l. (a) and higher than 2500 m a.s.l. (b). The trajectories are cut at the border of the COSMO model. The colour scale represents the back-trajectory arrival height. Corresponding altitudes of the backtrajectories vs time are reported in the bottom panels. The dots along each trajectory mark a 1-hour step and the black star indicates the trajectory arrival point (Aosta–Saint Christophe).

RC16. Fig. S4d: the particle number concentration increase during the case study is clearly visible. It is however not possible to determine if this is a peculiar or a normal event occurring regularly during winter.

AR16. As explained in AR1, the duration of the phenomenon represented in the figure was extended to one week in order to include more "non-event" days and better show the effect of the advections on the "standard" conditions in Aosta. The text in Sect. 4.2.3 was also updated in the following way: *The instrument reveals a notable increase in the number concentration for particles smaller than 0.5* µ*m (Fig. S4c) in coincidence to the arrival of the aerosol layer. The total number concentration (Fig. S4d) gradually increases from few hundreds particles* cm^{-3} *up to 3000 particles* cm^{-3} *and decreases again on 29 January (the average value in winter 2016–2017 in conditions of local pollution being 650 particles* cm^{-3}*).*

RC17. Fig. S9 is cited before fig. S8.

AR17. Corrected.

RC18. P. 32–33, lines 31–6:The hygroscopic growth of the aerosol can clearly be a cause of the discrepancy between FARM and ALC, even if FARM is also not able to reproduce PM measurements and their diurnal cycles. The difference of the measured size fraction (TSP for ALC and PM10 for FARM does however not seem to be really relevant since: 1) Aerosol from the Po Valley are described as small ones in the paper, see for ex. Case study 2, where the increase in PM10 is similar to the one of PM2.5 (Fig. 12), 2) the size distribution measured by the OPC and sun photometer peak in the accumulation mode and do not show big particles.

AR18. The referee is right, we modified the relevant sentence as follows:

A variety of (possibly concurrent) reasons can explain the observed underestimation, mainly related to:

1. *inaccuracies in retrieving the PM concentration from the ALC backscatter. As mentioned, ALC measures aerosol backscatter, so that specific tools were developed (Dionisi et al., 2018) and are used here to associate a PM value to it. Still, the expected error associated to these estimates is of the order of 30–40%. In addition, the ALC retrieval is based on functional relationships derived assuming maximum RH of 95%. Higher FARM-ALC discrepancies can be expected when RH>DRH, and particularly at RH>95%;*
2. *inaccuracies in the CTM simulations [...]*

RC19. P. 33 lines 7–13: How good is the estimation of the PBL height in the CTM simulations? Could a bad estimation of the PBL height be also a source of inaccuracies?

AR19. This could be a further, interesting evaluation of the model output to be performed by comparing the mixing layer height (Hmix) of the model pre-processor (SURFPRO) to the ALC measurements. In fact, these instruments are generally quite powerful in showing the temporal evolution of the mixing layer using the aerosol as tracer (e.g., Angelini et al., 2009). According to this comment/suggestion we modified Sect. 4.4 as follows:

Finally, some underestimation of PM values could also be due to overestimation of the FARM simulated PBL height. An evaluation of this kind of effect is in principle possible by comparing the simulations with the ALC-derived PBL height (e.g., Angelini et al., 2009; Haeffelin et al., 2012). However, this should be performed only selecting non-advection conditions, i.e., those in which the ceilometer signal is only affected by local aerosols and is thus able to follow the daily evolution of the local PBL using local particles as tracers. This kind of investigation was, however, beyond the scope of the present work. Challenges and recent efforts to define a PBL in mountainous areas ("Mountain Boundary Layer", MBL) and discrepancies between the MBL and the aerosol layer are more extensively described by Lehner and Rotach (2018).

RC20. P. 34 line 30–32: as explained previously, I do not see any evidence of a residual layer sinking towards the surface in this study.

AR20. Following the referee's comment, all references about sinking residual layers were removed from the text.

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