Response to Anonymous Referee #4

General comments: The paper analyzes the impact of pollutants transport from the Po Valley to the air quality on the Alpine region. Three selected cases studies of transport over the Aosta Valley are selected among a 3 years period and analyzed in detail by means of a wide- ranging set of observations and numerical simulations. Moreover, the observational data are used to evaluate the performances of the FARM chemical transport model. These results provide an interesting and comprehensive description of the complex phenomena of mountain-valley exchange and deserve therefore to be published.

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. Even if the paper is well written, though, the large amount of data presented in both the main paper and the support material makes the manuscript dispersive and fragmented. I would suggest to further select the figures that are useful to support their message and eliminate redundancies. For example, I invite the authors to carefully evaluate if the section 4.1.5 is bringing any relevant information: the analysis seems not fully convincing, is limited to a single case and the information that intends to bring is already conveyed by the FARM model. Similarly, some figures can be just substituted by a short mention in the text (I would reconsider the relevance of fig. 3, S2c, S4e, S5, S12).

Author's response 1. We understand the referee's objection. At the same time, we believe that the evaluation of the spatial extent of the phenomenon under investigation is important for its understanding. In this respect, Sect. 4.1.5 is the only one in which the phenomenon, which is carefully evaluated locally (Aosta region) with a large set of in situ and ground based measurements, is **observed** (not simulated) over the wider Northern Italy domain using space-based data. We therefore believe that this section, although with some limitations, is an important **observation-based** support to the FARM **model** results. For this reason, we would prefer to keep it.

We are also aware that the manuscript has a high number of figures to show results and support the relevant discussion, including several points raised by the reviewers. That's why we made the choice to leave additional, supporting material out of the main text (Supplement). We believe this choice still allows a straightforward reading of the main text, while providing additional details to those readers interested in more specific aspects of the study.

Still, following the reviewer's advice, we removed Fig. 3 from the text as most information was already

available in Fig. 4a.

RC2. Page 2, line 19–22: Basing on the results of the FARM comparison I would rather highlight that the paper aimed at an evaluation of the model and then mentioning that, despite its limitations, it still brought insights on the cases, supporting the observations. Similarly, I would rephrase the paragraph in the conclusions, accordingly (Page 35, line 14 onward)

AR2. Both the abstract and the conclusions were rephrased following the reviewer's advice.

Abstract: Results show that the simulations are important to the understanding of the phenomenon under investigation. However, in quantitative terms, modelled PM_{10} concentrations are 4–5 times lower than the ones retrieved from the ALC and maxima are anticipated in time by 6–7 hours. Underestimated concentrations are likely mainly due to deficiencies in the emission inventory and to water uptake of the advected particle not fully reproduced by FARM, while timing mismatches are likely an effect of suboptimal simulation of up-valley and down-valley winds by COSMO.

Conclusions: 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 (thermallydriven) 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.

RC3. Page 4, line 3: Do you really mean that is the partition between local and non-local sources that help to assess the impact of air pollutants on health, climate and ecosystem?

AR3. No, we meant to say that in order to set up mitigation actions to limit the impact of air pollutants on health, climate and ecosystems it is first necessary to understand where these are originated. Thank you for pointing out that the sentence was misleading, we rephrased it as follows:

Over the impacted areas, a correct partitioning between local and non-local sources is therefore necessary to 1) correctly interpret the exceedances of air quality limits; 2) develop joint efforts and large-scale mitigation strategies (WMO, 2012) to reduce the frequency and impact of pollution episodes on citizen health (Straif et al., 2013; WHO, 2016; Zhang et al., 2017), climate (Clerici and Mélin, 2008; Lau et al., 2010; Zeng et al., 2015) and ecosystems (Carslaw et al., 2010; Bourgeois et al., 2018; Burkhardt et al., 2018).

RC4. Page 14, line 21. "Therefore, to reduce errors in trajectories with increasing running time, we limit the computation to this duration". The sentence between commas, in this context, is mislead-ing. Rewrite as "Therefore, we limit the computation to this duration"

RC5. Page 18, Figure 6. It would be more meaningful to show the trajectories at 18:00 UTC for this case, since at this time you observe the start of the intense layer arrival and the trajectories are also showing a larger impact from the Po Valley.

AR5. The figure (Fig. 5, with the new numbering) was modified according to the referee's comment. It is shown here below (cf. also AR7 for further updates to the figure):



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).

RC6. Page 16, line 14: Mention here that you see these surface impacts in your measurements (Fig. 8). Also, what will happen if you average the hourly measurements of TEOM on a daily interval? Will it be consistent with the results of figure 8? Please note that the time interval shown in figure 4 seems to be, compared to figure 8, in the middle of the event (Starting from 26 August and ending at the end of 3 September), so that is not possible to identify any difference with the previous or following phase.

AR6. The discussion about the TEOM has been refined in the updated manuscript, also following the

comment by referee #3 (RC11).

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 µgm⁻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 $\rho = 0.84$).

Regarding the second comment by the referee about the time interval shown in former Fig. 4, we followed the reviewer's suggestion and similar remarks from referee #2 (RC2) and referee #3 (RC1 and RC10). Thus, 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).

RC7. Page 17, line 33: Do you mean here that the transport from the Po Valley persists at every hour along the following days? Indeed, considering the evolution of the layer as seen from the measurements, it would be useful to show which is the typical transport during the whole event at different hours, for example adding the average position of the trajectories (or the PDF of the position in case of large variability) on the considered period. If the authors want to still keep the trajectory of the first day of the episode as a reference for the layer arrival, I would suggest, to avoid burden the paper with additional plots, showing just the synoptic hours instead of a 3-hour time step; it should still be enough to demonstrate the transport evolution. This would also allow comparing the first and third transport type with the mean transport behaviour of the second case study, when the layer persists for the whole duration of the event.

AR7. The sentence at line 33 was modified as follows: *The analysis of the corresponding back-trajectories confirms that transport of polluted air masses from the Po basin also occurs in the afternoons of the other days of this episode, until the flux changes again to a north-western configuration (Fig. S3c and f).*

Following the comment by the reviewer (and similar remarks from referees #1 (RC3) and #3 (RC15)), the back-trajectories figures were modified in this way: we plotted in separate panels trajectories ending at altitudes < 2000 and > 2500 m a.s.l. over Aosta–Saint Christophe. Also, to further simplify the figures, we only show back-trajectories for specific times corresponding to the most significant variations of circulation patterns. Finally, according to referee #3 (RC15), for each time selected we added a bottom panel with the trajectory altitude along their journey. An example of the new plots is provided in Fig. 5, see AR5.

RC8. Page 21, line 15: Can you add some reference for the typical Angstrom exponent for the Aosta valley in comparison?

AR8. Done, we added the following sentence: *These values should be compared to the lower Ångström exponents typically measured in the Aosta Valley, i.e.* ~1.1 *on average (Diémoz et al., 2014, 2019).*

RC9. Page 25, line 18 (but same for page 30, line 6): How do you estimate this residence time, is it an average of the time spent by each trajectory over the Po Valley at all levels? From the shown figure (even if it is difficult to distinguish), the transport, including the part outside the PBL, seems faster (around 20 hours or even less). Can you be more specific?

AR9. This was simply computed considering that each dots along each trajectory mark a 1-hour step (as stated in caption of Fig. 5). Given this comment we clarified this point in the text as follows: *The trajectory altitude tends to decrease in the afternoon reaching the elevations of the polluted boundary layer* (Fig. S6b), *thus leading to effective aerosol transport to the Aosta Valley. In fact, considering that each dot in Fig. S6b represents a 1-hour step, we estimate a mean air masses residence time in the Po Valley PBL of 30–35 hours before arriving over the observing site. Finally, trajectories turn westerly on 29 January, in agreement with the removal of the layer over Aosta (Fig. S6c and f).*

RC10. Page 25, line 33–35: How do you justify these affirmations?

AR10. The sentence was partially rephrased: Taking into consideration these different daily evolution patterns and the sources included in the emission inventory, the most likely reasons for the differences between the model and the measurements at the surface appear to be an underestimation of the residential heating (actually switched on all day during these very cold days) and an overestimation of the traffic road contribution, together with an overestimation of the mixing layer height growth at midday by the NWP model.

RC11. Page 32, line 29: It would be interesting to see this comparison indeed since, from figure S1, it seems that the model has the tendency to see easterly winds more often and with higher intensity

respect to the observations. Is this comparison limited just to the surface measurements? May it be that there are problems in the higher layers (where most of the pollution layer transport is coming from)? For example, in the case study 2, when the winds were weaker both at the ground and at higher layers, the time evolution of the FARM simulation of Figure 10d was in better agreement with the observations. Is also interesting to note that the time evolution of the vertical distribution of PM10 from FARM is in better agreement (especially the local contribution) with the surface TEOM measurements rather than with the elevated layers observations.

AR11. The reviewer raised some important issues, also mentioned by referee #3 (RC3). We therefore decided to better investigate the capability of COSMO in reproducing the measured wind fields (unfortunately, our instrumentation allows validation only at the surface) and we updated Sect. 4.4 accordingly:

As already mentioned in the description of the three cases, the model [...] anticipates the peak concentrations compared to the profiles from the ALC [...], i.e. [it shows] 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, as already mentioned in AR2: *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.*

Finally, please note that an in-depth examination of FARM capabilities to reproduce the observed PM concentrations for event and non-event days was done in the companion paper based on the long-term dataset (2015–2017). We anticipate here Fig. 17 (companion paper), showing that the discrepancies between simulated and observed PM_{10} concentrations at the surface are minimum in cases of non-event days, which agrees with the referee's remark.



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.

RC12. Page 4, line 10: "will be quantified in a companion ..."

AR12. Done.

RC13. Page 4, line 28: For easier reading, specify which valley are you talking about. In the same paragraph, you are referring to both the Po valley and the Alpine valleys.

AR13. Done. The sentence now reads: [...] the heaviest burden of particles did not come from the largest urban settlement in the Aosta Valley, but rather from outside the region, namely from the Po basin.

RC14. Figure 2: The dashed lines are not needed.

AR14. Removed.

RC15. Page 9, line 21: "... altitude of the extinction coefficient."

AR15. Done.

RC16. Page 21, line 22: Refer also to the figure S8.

AR16. Reference to the figure was added.

RC17. Page 34, line 6: Add a ":" after "(see Introduction)"

AR17. Done.

RC18. Page 35, line 19: "... these issues may be partly attributed to ..."

AR18. Done.

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