Response to Anonymous Referee #1

General Comment. The paper evaluates the impact of trans-regional transport of aerosol from the Po Plain to the Aosta Valley (north-western Italian Alps), by means of both the analysis of experimental data and numerical simulations. The paper is complete, well-written and can be of interest for the community. Therefore in my opinion it is worth publishing in Atmospheric Chemistry and Physics, after a few minor comments are addressed.

We thank the reviewer for taking the time to revise our manuscript and for his/her pertinent comments. Our point-to-point reply 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. Meteorological model: the Authors say (pag. 8, line 7) that "we used a nudged, high-resolution variant, called COSMO-I2, covering Italy". However, in my opinion 2.8 km is not high-resolution in complex terrain. The floor of the Aosta Valley is 2-3 km wide, so meteorological phenomena triggered by the orography may be not well resolved by the model at this resolution, also considering the fact that the actual model resolution is 6-8 times the grid cell (Skamarock 2004). In fact authors say that the model surface altitude of Aosta urban area is 900 m a.s.l., whereas the actual altitude is 600 m a.s.l. For example Schmidli et al. (2018) showed that at 2.2-km resolution the COSMO model poorly simulates valley winds, while at 1.1-km resolution the diurnal cycle of the valley winds is well represented. Similarly, Giovannini et al. (2014) showed that 2-km resolution can be considered as the limit for a good representation of valley winds in narrow Alpine valleys.

Author's response 1. We understand the point raised by the reviewer. In the submitted version, we used the terminology "high resolution", as this appears in the COSMO web page (http://www.cosmo-model.org/content/tasks/operational/default.htm), where COSMO is indeed defined as a "high-resolution" model.

However, we accept the reviewer's remark and thus removed the term "high-resolution" in the revised text. Also, in Sect. 3.1 we now discuss the advantages of COSMO-I2 version over COSMO-ME, with reference to the high*er* resolution of the former (but not "high" in absolute terms). The revised text reads as follows:

The forecasts, inclusive of the complete set of parameters (such as the 3-D wind velocity used here) for eight time steps (from 00 to 21 UTC), are disseminated daily in two different configurations by the meteo-rological operative centre – air force meteorological service (COMET): a lower-resolution version (COSMO-ME, 7 km horizontal grid and 45 levels vertical grid, 72 hours integration), covering central and southern

Europe, and a nudged, higher-resolution version (COSMO-I2 or COSMO-IT, 2.8 km, 65 vertical levels, 2 runs/day), covering Italy (orange rectangle in Fig. 1a). Owing to the complex topography of the Aosta Valley, and the consequent need to resolve as much as possible the atmospheric circulation at small spatial scales, we used the latter version in the present work (cf. Sect. 4.5 for a discussion about possible effects of the finite model resolution in complex terrain).

Furthermore, following the reviewer's comment, we now preferably use the term "grid spacing/step" instead of "resolution" throughout the revised text.

We also added the suggested references in Sect. 4.5:

Although the COSMO-I2 grid spacing is certainly in line with that of other state-of-the-art operational limited-area models, in our complex terrain it could be insufficient to appropriately resolve local meteorological phenomena triggered by the valley orography (Wagner et al., 2014), also considering that the actual model resolution is 6–8 times the grid cell (Skamarock, 2004). For example, Schmidli et al. (2018) show that at 2.2-km grid step the COSMO model poorly simulates valley winds, while at 1.1-km grid step the diurnal cycle of the valley winds is well represented. Similarly, Giovannini et al. (2014) show that 2km step can be considered as the limit for a good representation of valley winds in narrow Alpine valleys. Smoothed digital elevation model (DEM) used within COSMO and FARM could also play a direct role in the detected underestimation. In fact, as mentioned in Sect.3.2, the model surface altitude of the Aosta urban area is 900 m a.s.l., whereas the actual altitude is about 580 m a.s.l. (Table 1). The adjacent cells are given an even higher altitude, owing to the fact that the valley floor and the neighbouring mountain slopes are not properly resolved at the current resolution. This results in an apparent lower elevation of the sites located in flatter and wider areas, such as Aosta, while the real topography profile at the bottom of the valley presents a monotonic increase from the Po plain to Aosta. Therefore, it is expected that, just by better reproducing the orography, a higher resolution would allow to better resolve the horizontal advection of aerosol-laden air from much lower altitudes on the plain.

Most of these issues were extensively addressed in the companion paper (Diémoz et al., 2019). In that study, COSMO-I2 was shown to be capable of reproducing the mountain-plain wind patterns observed at the surface both on average (cf. Fig. S1 in the companion paper) and in specific case studies (Fig. S13 therein). Nevertheless, it was also found to slightly anticipate in time and overestimate the easterly diurnal winds in the first hours of the afternoon and to overestimate the nighttime drainage winds (katabatic winds, ventilating the urban atmosphere and reducing pollutant loads). These limitations were mostly attributed to the finite resolution of the model.

RC2. Building on the previous consideration, it is not easy to evaluate the performance of the chemical model without a validation of the meteorological model. The Authors attribute most of the discrepancies with respect to measurements to deficiencies in the boundary conditions of the chemical model. However, this statement is difficult to be demonstrated without a complete validation of the modelling chain. Moreover, increasing the boundary conditions by a factor 4 reduces the mean bias, but does not reduce the RMSE (Table 3). So it seems that there is still a compensation of underestimations and overestimations.

AR2. One of the most relevant updates in the revised manuscript is the study of the chemical transport model performances in a flatter terrain compared to the complex orography of the mountain valley. This is accomplished by including the PM_{10} dataset measured in a station at the boundary of the domain, Ivrea. This addition clearly showed that the same PM_{10} underestimation of the model is also obtained over flat terrain, demonstrating this is not driven by inaccuracies of the simulated wind fields over the more complex terrain of Aosta. Therefore, Sect. 4.5 has been modified to better explain this point and investigate the relative importance of both emissions (inventories/unaccounted physical processes) and transport (performances of the meteorological model) in generating the model-measurements differences:

The observed model-measurement discrepancies might originate from (1) an incomplete representation of the inventory sources (emission component), (2) inaccurate NWP modelling of the meteorological fields, and notably the wind (transport component), or a combination of (1) and (2). Some details on these aspects are provided in the following.

1. Emissions. Inaccuracies in the (local and national) emission inventories could degrade the comparison between simulations and observations. Based on results shown in Fig. 17(a,b), we decided to investigate the sensitivity of our simulations in Aosta-Downtown to the magnitude of the external contributions (boundary conditions). In particular, we used a simplified approach to speed up the calculations: assuming that the contribution from outside the regional domain and the local emissions add up without interacting, we simulated the surface PM_{10} concentrations turning on/off the boundary conditions (dark- and light-blue lines in Fig. 16) to roughly estimate the only contribution from sources outside the regional domain. Interestingly, the difference between the two FARM runs correlates well with the advection classes observed by the ALC (Fig. 18). This clear correlation between the simulations and the experimentally-determined atmospheric conditions is a first good indication that the NWP model used as input to the CTM yields reasonable meteorological inputs to FARM. Then, to further explore the sensitivity to the boundary conditions, we gradually increased, by a weighting factor W, the PM_{10} concentration from outside the boundaries of the domain trying to match the observed values. In Figs. 17(c, d) we show the results obtained with W=4. This exercise show that the overall mean bias error is much reduced compared to the original simulations (W=1, Figs. 17(a, b)), especially for the winter and autumn seasons, while slight overestimations are now visible for summer and spring. Also the annually-averaged MBE, NMBE and NMSD improve (Table 3), whilst the other statistical indicators remain stable or even slightly worsen.

Clearly, our simplified test has major limitations. (...)

2. Although the COSMO-I2 grid spacing is certainly in line with that of other state-of-the-art operational limited-area models, in our complex terrain it could be insufficient to appropriately resolve local meteorological phenomena triggered by the valley orography (Wagner et al., 2014), also considering that the actual model resolution is 6–8 times the grid cell (Skamarock, 2004). (...)

Also, to disentangle the role of factors (1) and (2) discussed above, we evaluated the model performances in a flatter area of the domain, where simulations are expected to be less affected by the complex orography of the mountains. We therefore compared PM_{10} simulations and measurements in Ivrea (Fig. 16b). This test is also useful to operate the CTM model at the boundaries of the domain, with special focus on the emissions from the "boundary conditions". Model underestimation in both the warm and cold seasons is evident. Incidentally, it can be noticed that concentrations simulated without "boundary conditions" are nearly zero, since the considered cell is far from the strongest local sources and most part of the aerosol comes from outside the regional domain. As done for Aosta (...), Fig. 19(a,b) presents the absolute and relative differences between the model and the observations in Ivrea. The overall NMBE is -0.73, which rather well corresponds to the underestimation factor W=4 (or NMBE=-0.75) found for Aosta–Downtown and other sites in the valley. Since it is expected that in this flat area the NWP model is able to better resolve the circulation compared to the mountain valley, this result suggests poor CTM performances to be mostly related to underestimated emissions at the boundaries rather than to incorrect transport. In addition to this general underestimation, a large scatter between observations and simulations can be noticed in Fig. 16, the linear correlation index between simulations and observation in Ivrea being rather low (ρ =0.54). If such a large scatter, due to the erroneous boundary conditions, is already detectable at the border of the domain, it is likely that at least part of the RMSE reported in Table 3 for Aosta–Downtown can be attributed to inaccuracies in the national inventory.

We report the new figures 16 and 19, previously mentioned in the text, here below. Fig. 17 is shown in AR3.



Figure 16: Long-term (1 year) comparison between PM₁₀ surface measurements and simulations (FARM) in Aosta–Downtown (a) and Ivrea (b).



Figure 19: Absolute (a) and relative (b) differences between simulated and observed PM_{10} concentrations at the surface in Ivrea.

Note that, in accordance with the inclusion of the additional measurement site of Ivrea, Sect. 2 has been integrated as follows:

We also consider in our analysis the PM_{10} records collected in the city of Ivrea, a site in the Po Plain (31 µg m^{-3} average PM_{10} in 2017) located just outside the Aosta Valley, in the Italian Piedmont region (...). This was done to check if and how much inaccuracies of the model in reproducing the aerosol loads over the Aosta Valley are due to difficulties in simulating the wind field in such a complex terrain. In fact, the city centre of Ivrea (24000 inhabitants) is approximately in the middle between the measurement site (south of the city) and the nearest cell of our model domain (north of the city), the distance between these two points being only 2 km. The altitude of the cell (from the digital elevation model used in our simulations) is 241 m a.s.l., i.e. approximately the real one (i.e., 243 m a.s.l). (...) The station of Ivrea features, among other instruments, a TCR Tecora Charlie/Sentinel PM_{10} sampler. PM_{10} concentrations are then determined by a gravimetric technique.

Finally, following the reviewer's comment, we also mention in the revised manuscript that *an additional contribution to the observed RMSE could still be given by errors in modelling transport due to the coarse resolution of the NWP model, although we are not able to quantify the relative role of the two factors. To unravel this issue, high-resolution models (grid step 0.5 km, e.g. Golzio and Pelfini, 2018; Golzio et al., 2019) are being tested on the investigated area and their results will be addressed in future studies.*

RC3. In Figure 17 it would be interesting to see also a normalized mean bias (i.e. a mean bias normalized by the average concentration measured for each class).

AR3. Figure 17 has been modified taking the reviewer's advice into account (see panels b and d). References to the Donnas station have been removed following comment RC20 by Referee#2. The new figure is reported below.



Figure 17: Absolute (a,c) and relative (b,d) differences between simulated and observed PM_{10} concentrations at the surface in Aosta–Downtown. The mean bias error (MBE) and the normalised mean bias error (NMBE) for each case are reported in the plot titles. First row: FARM simulations as currently performed by ARPA. Second row: the PM_{10} concentrations from outside the boundaries of the domain were multiplied by a factor W=4.

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RC4. Page 1, line 4: ... 3-year period ... Section 4.3: The two final sentences begin with "Finally". Page 36, line 18: ... this regular air mass transport ...

AR4. Thank you for these technical remarks. All suggested changes have been accepted in the revised manuscript.

RC5. References [...]

AR5. All recommended references have been included in the revised manuscript.

References

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