Answer to the comments of Anonymous Referee #2

We would like to thank the reviewer for their comments (marked blue in the following text). Here are our answers:

- please double check the reference list: I checked the first three citations, and they are all missing in the list at the end of the manuscript.

Thanks to the reviewer for noticing this, all references from the first few paragraphs had indeed disappeared from the reference list. The reference list has been fixed.

- I find too much overlap and redundancy in the introductory part on model-to-obs uncertainties (page 2-4) and the discussion (section 4). My impression is that a quite long list of possible reasons for model-to-obs disagreement is presented in both parts, but never really demonstrate them for the specific simulations presented here. I thus suggest to shorten both the introduction and the discussion, and possibly move all the model-to-obs issue directly into the discussion section.

The model-observation comparison issues will be moved to discussion.

- page 4, lines 7-10: I do not completely agree with this final statement. From my understanding, the "main reasons behind model-measurements" are not clearly identified in the study. I would better state that the model error regarding the PM simulation is characterized against available measurements.

The sentence will be restated according to the reviewer's suggestion.

- Figure S4: please define PPMr in the caption.

Done

- page 8, lines 9-13: there seems to be some inconsistency between the major components illustrated in figures S5-S8 and values given in Table 6. In particular, from Table 6 one would say that carbonaceous aerosol are the major fraction of PM, not secondary organic aerosol. Please clarify.

There are several reasons for the noticed discrepancies. Firstly, the concentrations are given in different units. Table 6 reports the annual mean concentrations observed by the EMEP network, averaged over all the stations. They are given in the units the observations are provided – micrograms of nitrogen, sulphur, sodium, calcium or carbon. The model median maps on the figures S5-S8 follow the speciation in the models and are in the total mass of the component (NO_3, NH_4, SO_4, sea salt, mineral dust, organic aerosol, fire-emitted PM). Converting the observed values to the total mass of the components, we get the sum of the secondary inorganic species very close to the carbonaceous ones in $PM_{2.5}$. Taking into account that the models underestimate substantially the carbonaceous part, the model maps probably overestimate the SIA fraction in PM. Additional differences are introduced by sampling the map only at the station locations. For the SIA species the station network is well covering and representative, while the carbonaceous components were measured in only four stations.

Clarifications will be added.

- page 8, line 31: could a poor correlation coefficient for NO3- be related to a pulsed behaviour of the aerosol nitrate production in the PBL, as recently described in this paper: Curci, G., Ferrero, L., Tuccella, P., Barnaba, F., Angelini, F., Bolzacchini, E., Carbone, C., Denier van der Gon, H. A. C., Facchini, M. C., Gobbi, G. P., Kuenen, J. P. P., Landi, T. C., Perrino, C., Perrone, M. G., Sangiorgi, G., and Stocchi, P.: How much is particulate matter near the ground influenced by upper-level processes within and above the PBL? A summertime case study in Milan (Italy) evidences the distinctive role of nitrate, Atmos. Chem. Phys., 15, 2629-2649, doi:10.5194/acp-15-2629-2015, 2015.

The paper of Curci et al presents a detailed study on nitrate formation in Po Valley and how its concentration is influenced by the boundary layer processes and temperature and humidity vertical profiles. It is indeed an interesting question, how well these effects are taken into account in the regional models. The models use a range of different algorithms for SIA creation, such as thermodynamic equilibrium model ISORROPIA2 in LOTOS-EUROS or separate equilibrium computations for NH_4NO_3 and a parameterization for coarse NO_3 production on sea salt particles in SILAM and EMEP. Generally these should be capable of simulating the profile of NO_3 creation and brake up, provided that the model vertical resolves the temperature and relative humidity profiles. However, this can easily not be the case - for instance LOTOS-EUROS has a single layer representing the whole boundary layer and SILAM layers at boundary layer top can easily be 500-1000 meters thick.

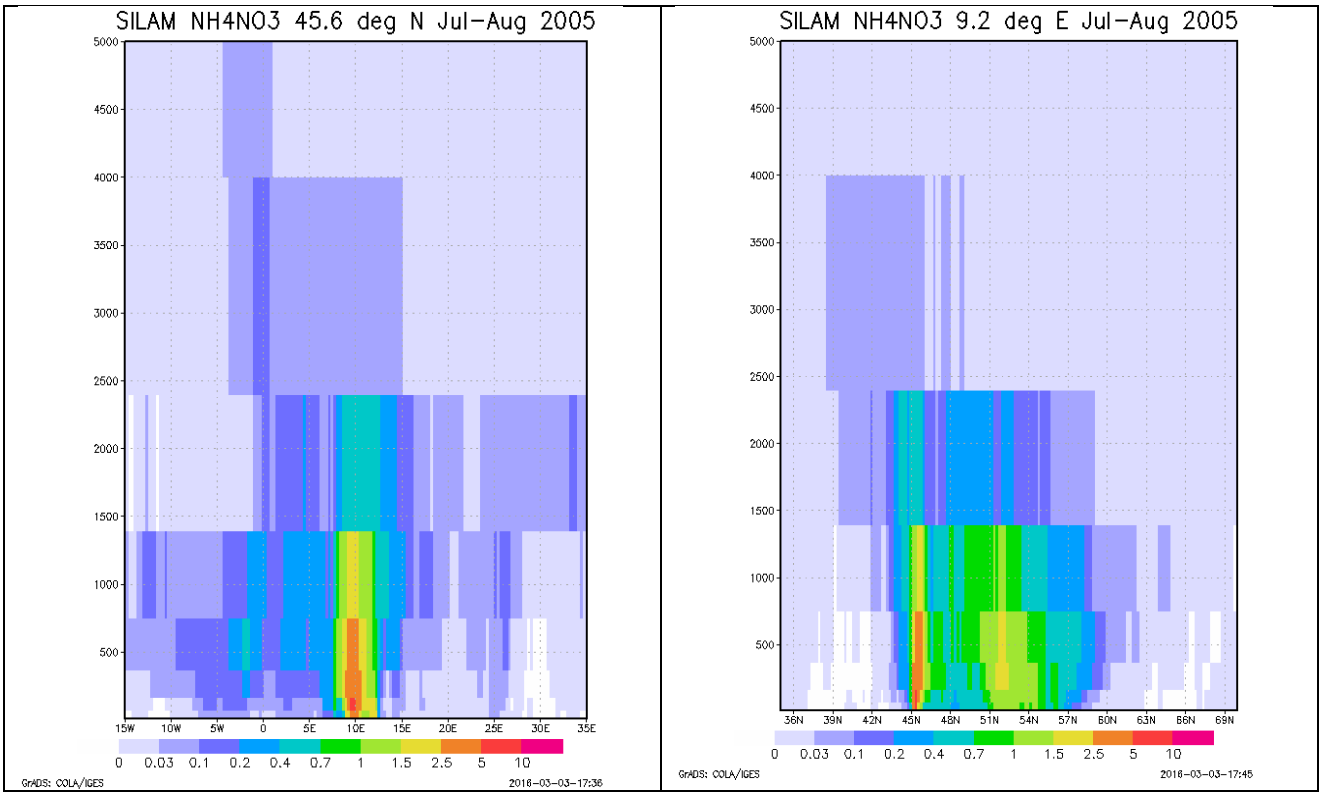

Figure 1 Summer time concentration profiles of NH_4NO_3 from SILAM model, west-east and south-north cross-sections of Europe, selected to cut through Po Valley.

While the low vertical resolution of the gathered model data does not allow for a detailed analysis of this effect in the whole dataset, elevated plumes of NH_4NO_3 are clearly visible in SILAM model output (Figure 1), mainly because the efficient dry deposition of HNO_3 depletes its concentration in the near-surface layers making NH_4NO_3 break to the gaseous compounds. Thus, the formation of the residual layers and mixing them down in the morning and the ability of the models to reproduce these effects can have an impact on the simulation quality. However, as the EMEP aerosol composition observations have daily resolution, it's

hard to tell, how much these processes influence the model-measurement correlation compared with all the other uncertainties related to NH_4NO_3 formation, such as the temperature dependence of NH_3 emission.

The discussion of the uncertainties in NO_3 modelling will be extended in the manuscript.

- page 9, lines 7-8: the discussion on HNO3 bias is made difficult by the fact that HNO3 is not shown alone in the figure. Could this be shown, or the results commented on the NO3+HNO3 concentration that the reader may actually directly see in the plots?

The reason for selecting HNO_3+NO_3 as the presented variable was, that there are more stations in EMEP network that measure the sum than there are those that measure HNO_3 alone, also the sum is measured with higher accuracy, while the HNO3 concentration is more prone to observation artefacts .

Figure 2 shows HNO_3+NO_3 together with the contribution from HNO_3 (shaded). On annual average level all models overestimate the sum by 5-35 %. EMEP and LOTOS-EUROS accurately predict the HNO_3 fraction in the sum, while SILAM underestimates and CMAQ overestimates it by 10%. However, the models do not reproduce well the seasonal variations in HNO_3 concentration.

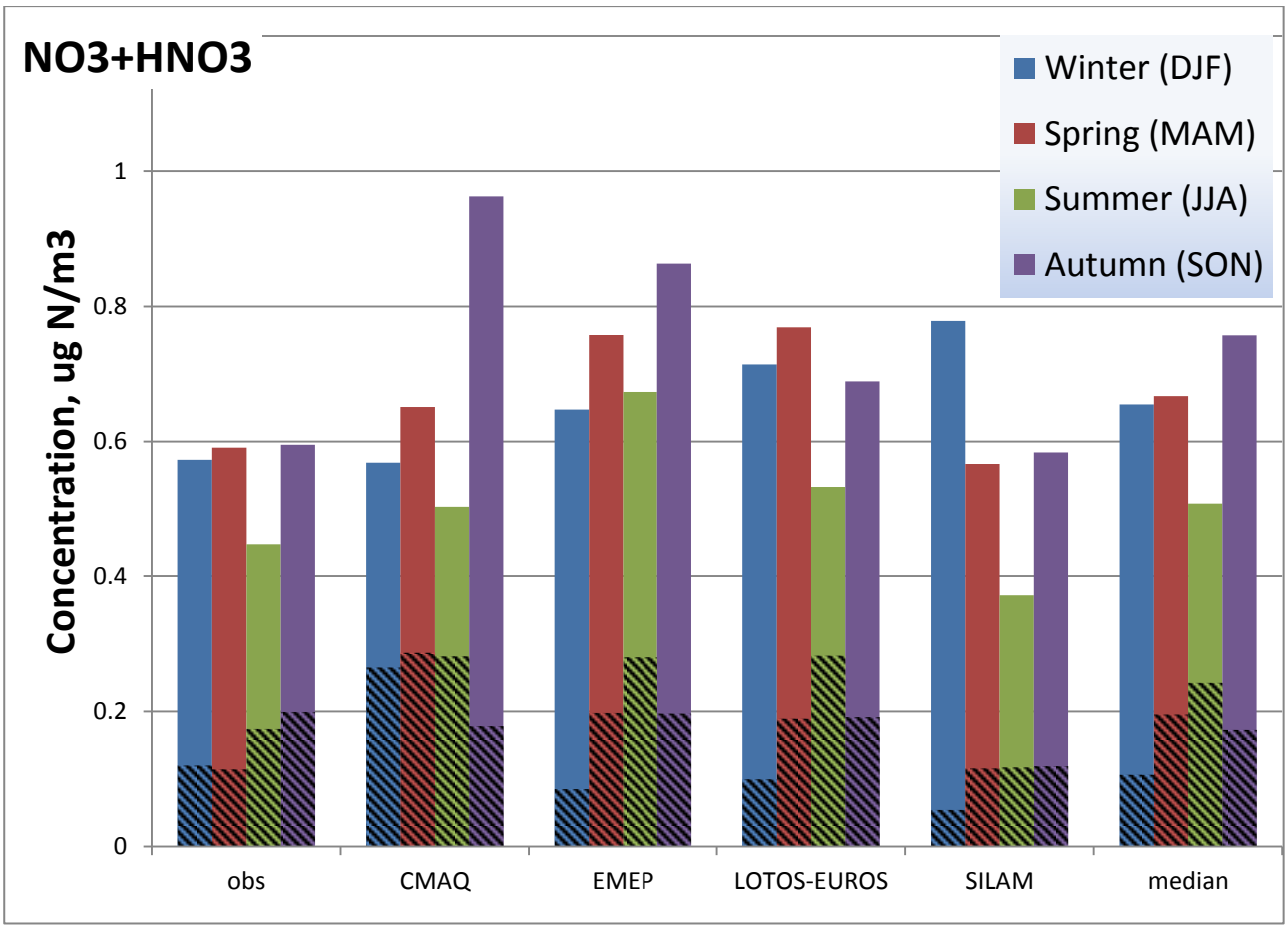

Figure 2 Observed and predicted seasonal concentrations of HNO_3+NO_3, mean over the EMEP stations [µg N m^{-3}]. Shaded part shows the concentration of HNO_3. Only the stations where at least two of HNO_3, NO_3 and HNO_3+NO_3 were observed, so that the HNO_3 fraction could be estimated, are included in the averaging.

The figure and related discussion will be updated in the paper.

- page 9, line 10: it is not completely true that the seasonal cycle is not reproduced by all SIA, e.g. NO3 and NH4 are reproduced quite well.

Will be restated

- page 9, line 16: "... overestimate the temperature dependence ..." suggest to rephrase with "... have an exaggerated temperature dependece ..." to avoid confusion with overestimated/underestimated resulting PM values

Done

. - page 9: in general, natural PM seems to be a major factor contributing to the spring PM maximum: may you confirm that (or not)?

Of the natural aerosols considered (Figure 5 in the manuscript), sea salt exhibits no spring peak - elevated Na^+ concentrations are observed only in winter and the models reproduce this behaviour. Dust, on the other hand, seems to be contributing – observed nss-Ca concentrations peak in spring and so do the modelled Saharan dust concentrations. Previous studies about Saharan dust have found the emissions peaking at spring (Fiedler et al., 2013; Laurent et al., 2008). Additionally to desert dust there are other possible reasons for elevated crustal aerosol concentration in spring, such as agricultural activities and vehicle caused erosion of roads - in colder regions where the roads are sanded against slipperiness high dust emissions occur when the road conditions get dry in spring.

Considering the biogenic primary particles, pollens are abundant in air in spring - maximum concentrations can reach a few tens of thousands of pollen/m3, equivalent to tens of µg/m3. However, their size is mostly too large to make them relevant for PM_{10} (~20 µm for abundant early spring flowering species like birch). Evidence exist, that pollen grains can break in atmosphere and produce particles in size range relevant for even $PM_{2.5}$, however, such particles are unlikely to be abundant enough to be noticeable in the PM budget. Also the fungal spores become abundant only in summer – high concentrations can be observed from June to October.

Regarding the anthropogenic contribution, all observed secondary inorganic species show spring maxima.

- page 10, lines 16-17: perhaps could be useful the discussion on EC lifetimes presented in this paper: Wang, X., Heald, C. L., Ridley, D. A., Schwarz, J. P., Spackman, J. R., Perring, A. E., Coe, H., Liu, D., and Clarke, A. D.: Exploiting simultaneous observational constraints on mass and absorption to estimate the global direct radiative forcing of black carbon and brown carbon, Atmos. Chem. Phys., 14, 10989-11010, doi:10.5194/acp- 14-10989-2014, 2014.

The paper of Wang et al reports shorter EC lifetimes to be more consistent with observations and a similar conclusion was reached by Samset et al., (2014) - both studies found that shorter lifetime was necessary for reproducing the EC vertical profiles and low concentrations in remote regions. This result somewhat contradicts with the current model intercomparison, where SILAM was found to best reproduce the observed EC concentrations, and longer EC lifetime due to slower deposition in that model was assumed as the main reason for the model-to-model differences. Also the temporal correlation with 2005 observations

and spatial correlation between the models 2005 average EC and EC observed during the 2002-2003 EMEP campaign is no worse for SILAM than it is for the other models, so there is no clear indication that the slower deposition would not be consistent with the surface EC observations in European scale. However, as indications were found of strong underestimation of EC emission, the slower deposition in SILAM is likely to be compensating for the missing emissions. Observations of vertical profiles and concentrations in more remote locations would be necessary for investigating this issue; unfortunately such were not available in Europe in 2005.

Discussion of EC lifetimes will be added to the manuscript.

Fiedler, S., Schepanski, K., Heinold, B., Knippertz, P., Tegen, I., 2013. Climatology of nocturnal low-level jets over North Africa and implications for modeling mineral dust emission. J. Geophys. Res. Atmos. 118, 6100–6121. doi:10.1002/jgrd.50394

Laurent, B., Marticorena, B., Bergametti, G., Leon, J.F., Mahowald, N.M., 2008. Modeling mineral dust emissions from the Sahara desert using new surface properties and soil database. J. Geophys. Res. Atmos. 113, 1–20. doi:10.1029/2007JD009484

Samset, B.H., Myhre, G., Herber, A., Kondo, Y., Li, S.M., Moteki, N., Koike, M., Oshima, N., Schwarz, J.P., Balkanski, Y., Bauer, S.E., Bellouin, N., Berntsen, T.K., Bian, H., Chin, M., Diehl, T., Easter, R.C., Ghan, S.J., Iversen, T., Kirkevag, A., Lamarque, J.F., Lin, G., Liu, X., Penner, J.E., Schulz, M., Seland, Skeie, R.B., Stier, P., Takemura, T., Tsigaridis, K., Zhang, K., 2014. Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations. Atmos. Chem. Phys. 14, 12465–12477. doi:10.5194/acp-14-12465-2014