Anonymous Referee #2 - Comments on REVISED SUBMISSION

The authors have addressed all comments by both reviewers and the presentation of the results and the discussion have been improved. To my opinion, a few additional improvements are needed before publication of the paper in ACP.

1- Abstract: line 44-46 :"These differences are mainly explained by vertical mixing within the boundary layer, as suggested by ground-based measurements of vertical profiles of aerosol backscatter, used as tracer of the vertical distribution of pollutants in the atmospheric boundary layer"

New Comment: From your new analysis (new figure 10), i.e. calculation of the boundary layer column of NH3 as suggested by reviewer Benjamin Loubet, I understand that the difference in the diurnal patterns between surface concentrations and total column measurements can be mostly attributed to the changes in the mixing layer height, provided that boundary layer is considered well mixed and therefore homogeneous in ammonia concentrations (new Figure 10). This finding has to reflect in the abstract.

Done. We have added this aspect in lines 45-46 of the revised manuscript: "*These differences are mainly explained by vertical mixing within the boundary layer, provided that this last one is considered well mixed and therefore homogeneous in ammonia concentrations. This is suggested by ground-based measurements of vertical profiles of aerosol backscatter, used as tracer of the vertical distribution of pollutants.*"

2- Earlier reviewer comment: Line 237: The reference you provide is an entire textbook. Please be specific. Which thermodynamic model is used in that simulation. (Unfortunately, the web site provided for the model in line 234 requires password, so it seems to be useless for the reader. I suggest removing it.)
Author reply: Clarified and added new information. We specify the thermodynamic model input information in Lines 254-255. "Chemical reactions are simulated using the MELCHIOR2 mechanisms scheme and tabulations from ISORROPIA model for thermodynamic equilibrium calculations of the species." And changed the web site provided to one accessible without password: http://www.esmeralda-web.fr/accueil/index.php

New comment: I am afraid it remains unclear to me: Do I understand correct that ISORROPIA model has been used to produce tables that have been then inserted in the CHIMERE model? Or is only that the equilibrium coefficients and other thermodynamic input data from ISORROPIA have been used in some module of Chimere to calculate the equilibrium? Please further clarify.

Clarified. ISORROPIA is used to produce tables that are inserted in the chemistry-transport model for calculating the thermodynamic equilibrium of the species.

This is stated in the manuscript as (lines 254-257): *"Chemical reactions are simulated using the MELCHIOR2 mechanisms scheme and the ISORROPIA model (Nenes et al., 1998). This last one has been used to produce tables that are inserted in the CHIMERE model for calculating the thermodynamic equilibrium of the species."*

3- New Comment: Line 313: PM2.5 levels ARE clearly enhancED (up to 80 ug m-3), as also seen in the daily averageD results of the simulations.

Done.

4- New Comment: Line 323: Reading the manuscript I understand that PM1 was measured at SIRTA while PM2.5 at other location in the Ile-de-France region. Thus, PM1 and PM2.5 measurements are not co-located. These two types of measurements could be compared when the measurements are co-located, which -to my understanding - is not the case, or when the PM2.5 measurements could be considered as characteristic of the region. It is true that the 3 PM2.5 sites provide very similar results, with the exception of the night of the 26-27 March but SIRTA is not included in the triangle defined by the PM2.5 sites. I do not see how the authors exclude the fact that there might be differences in the background levels at SIRTA and the other stations as well as different sources and sinks. Do the authors have another argument/evidence that the 3 PM2.5 stations are representative of the entire area including SIRTA? As mentioned only with co-located observations one can relate PM1 and PM2.5 observation.

Clarified and agreed. These two aspects are clarified in the following:

The SIRTA site measurements of PM1 are statistically consistent over a long period with respect to PM2.5 data from Gennevilliers, Bobigny and Vitry-sur-Seine. This is shown by the ancillary figure 17 from Petit (2014) shown below. We observe that the statistical distribution over 2 years of the measurements of PM2.5 at the urban background sites Gennevilliers, Bobigny and Vitry-sur-Seine of AIRPARIF (the ones from Figure 4 of the manuscript Kutzner et al.) are very similar to that of PM1 at the SIRTA site. This is clearly not the case of traffic (A1, AUT, BP_EST) nor rural stations (RUR_S).

We agree that PM1 measurements at SIRTA might occasionally differ in terms of background levels with respect to PM2.5 at this site itself and with respect to the other 3 sites. However, most (larger than 80 %) of the variability of PM2.5 during pollution events are measured at SIRTA in terms of PM1. This is illustrated in the Figure 18 from Petit (2014) showing the difference between PM1 and PM2.5 at the SIRTA site itself for one month in 2013. We clearly remark that the largest variability is observed for the PM1 fraction, which represents in average 85% of PM2.5, over the period of comparison. In cases of relatively higher pollution level (PM1 larger than 20 µg/m3), PM1 is larger than 80% of PM2.5 while in is about 50% for low pollution levels.

These aspects are clarified in the revised manuscript as (lines 325-329) "*In the Paris region, the PM1 generally represent 90% of PM2.5 (Petit et al., 2017), particularly when PM1 is larger than 20 µg m-3 (although for lower levels, PM1 may represent around 50% of PM2.5, Petit (2014)). Occasionally, some background levels of PM might not be accounted in PM1 that are measured as PM2.5 (Petit, 2014). Moreover, comparisons made by Petit (2014) show a very similar statistical distribution for both PM1 at SIRTA and PM2.5 at the urban background stations at Paris suburbs mentioned in Fig. 4.*"

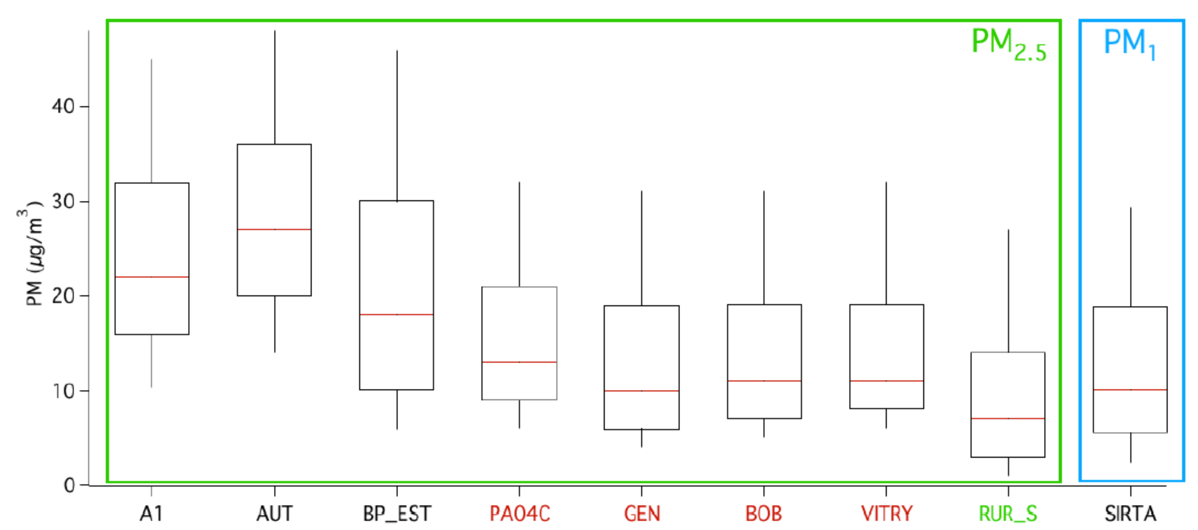

Figure 17 of Petit (2014): Variability in terms of box plots of PM observations from 01/04/2012 to 01/05/2014 from traffic stations (A1, AUT, BP_EST), urban background station (PA04C Paris downtown, GEN: Gennevilliers, BOB: Bobigny and VITRY: Vitry sur Seine), a rural station (Bois-Herpin) and the SIRTA site (only this one is PM1 while others are PM2.5).

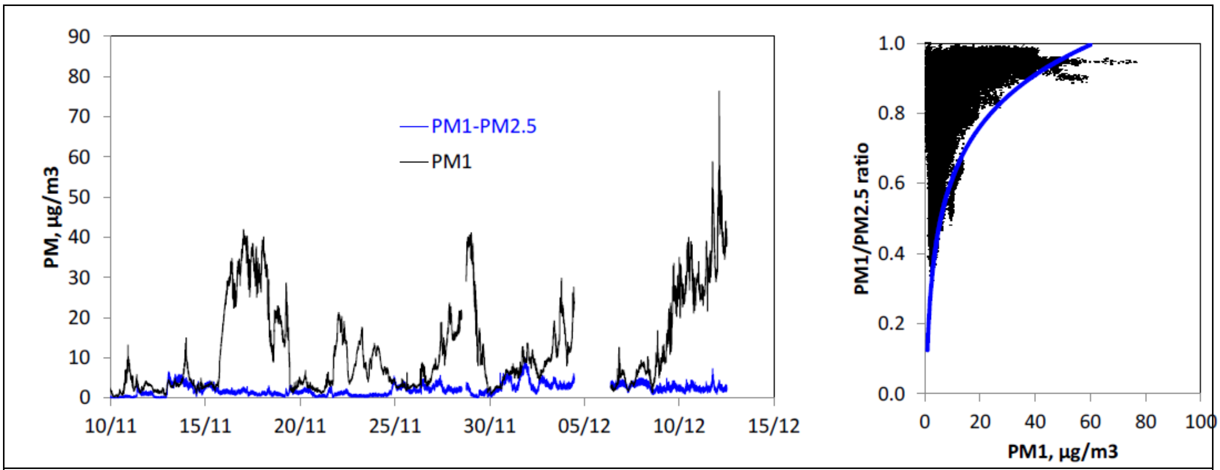

Figure 18 of Petit (2014): (Left) time evolution of PM1 (diameter smaller than 1 µm) and PM2.5-PM1 (diameter between 1 and 2.5 µm from November to December 2013 measured at SIRTA. (Right) scatterplot of the ratio PM1/PM2.5 with respect to PM1 (44032 minute-resolution points)

Petit, J.-E. : Compréhension des sources et des processus de formation de la pollution particulaire en région Ile-de-France, PhD manuscript of the University of Versailles Saint-Quentin-en-Yvelines, France, 2014

5- Line 473: act AS local sinks

Done.

6- Author reply: This is clarified in lines 625-626 of the RM. “This issue would be best addressed with chemistry-transport model simulations. “

New Comment: This sentence is misleading because ISORROPIA II is a thermodynamic equilibrium model and CHIMERE is a chemistry transport model. What is needed is coupling the CTM with ISORROPIA, and the reader is wondering why these two have not been coupled for this study to fully address the gas-to-particle conversion issue of ammonia. In addition, CHIMERE model output could have been easily used off line in the ISORROPIA II model. A better argument is needed here.

Clarified. It is true that ISORROPIA II is a module integrated (as tables in the version used in the paper) within the chemistry transport model CHIMERE. However, we would need to parametrize and validate it to use it to describe particle-gas partitioning for the specific case analyzed in the paper and the region of the Paris megacity. The current low accuracy for simulating ammonia/ammonium nitrite gas-to-particle conversion with a chemistry transport model is illustrated by Petetin et al. (2016). A dedicated study of parametrization and validation of the model would need co-located in situ measurements of particulate nitrate, ammonia, and nitric acid. Therefore, such study is out the scope of the present paper.

We have added the following comment to clarify this aspect (lines 629-631) “*This issue would be best addressed with chemistry-transport model simulations and dedicated in situ measurements (including nitric acid, particulate nitrate and ammonia) for the parametrization and validation of the model. “*

7- Author reply lines 611-614: Other processes such as surface and canopy uptake from surrounding ecosystems, depending on pH and total nitrogen input, may also explain surface concentration reductions (Massad et al., 2010, Flechard et al., 2013, Personne et al., 2015).

New Comment: Please also add light and temperature.

Done.