

Interactive comment on “Diurnal evolution of total column and surface atmospheric ammonia in the megacity of Paris, France, during an intense springtime pollution episode” by Rebecca D. Kutzner et al.

[Response to comments #2](#)

Anonymous Referee #2

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This paper investigates ammonia diurnal variability near surface as well as in the tropospheric column over the megacity of Paris in spring 2012 during an intensive pollution episode and shows significant differences between them. The observations are analyzed in conjunction with particulate matter levels and meteorological parameters and the differences are explained by the dilution within the boundary layer and also by volatilization of ammonium nitrate particles. The study is interesting and contributes to the understanding of ammonia variability in the atmosphere. The manuscript is overall nicely written; however, some clarifications and corrections are needed before publication in ACP.

Response: First of all, we would like to thank again Referee#2 for the constructive and useful comments which served us as a guideline for compiling the second revision of the manuscript (RM revised manuscript hereafter). All comments are addressed as detailed below. We agree with his scientific comments and editing suggestions. We have corrected and added clarifications in the RM for addressing all of the remarks of the referee.

Line 382-384: This sentence is misleading. ‘higher altitudes’ needs definition because Fig 6. shows that this anti-correlation between RH and Temp is limited to the first 1000 or 1500 m.

Clarified. We have added the following clarification in Lines 409-410 of the RM: “As a conclusion, the decrease in T and the increase of RH within the boundary layer height of 1-1.5 km with respect to ground shift the equilibrium to the aerosol phase.”

Line 235: Please provide further information on the model version you are using is it running off-line and if yes with what meteorological parameters. Are the CHIMERE simulations associated with ERA-Interim meteorology mentioned in lines 238-342 to be used to analyze the meteorological conditions?

Clarified and added. The paragraph in Line 248-254 was modified to provide clearer information of the use of the ESMERALDA output. “The horizontal distribution of air pollutants at the European scale is studied with CHIMERE chemistry-transport model simulations of PM_{2.5} provided by the ESMERALDA (EtudeS Multi RégionALES De l’Atmosphère, Cortinovis et al., 2015) project (<http://www.esmeralda-web.fr/accueil/index.php>). The version 2008b of CHIMERE is run hourly and averaged at daily scale, with a horizontal resolution of 15 x 15 km² and 9 vertical levels between 20 m to 5 km. Meteorological inputs for CHIMERE come from MM5 simulations (Dudhia, 1993), using Final Analyses (FNL) data from National Centers for Environmental Prediction (NCEP) as boundary conditions.”

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.)

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>

Line 372: ISORROPIA reference is Nenes et al. 1998; ISORROPIA II reference should be Fountoukis and Nenes, ACP, 2007 please correct accordingly. Also, in which form ISORROPIA was run (forward) or backward? What input data have been used? Information on how the ISORROPIA (or ISORROPIA II) simulations are done is missing and will affect any results of the model, although in the paper such results are discussed only qualitatively.

Clarified. We use ISORROPIA II model in the forward configuration. In the RM, the reference is corrected, and we specify the type of calculation and the input species (lines 397-401) “An additional analysis was performed with the ISORROPIA II box model (Fountoukis and Nenes, 2007) to investigate the role of temperature and relative humidity in the partitioning of ammonium nitrate. The forward calculation used measurements of the SIRTa site for NH₄⁺, NO₃⁻ and NH₃ on 28 March 2012 representing the highest concentrations on the studied period, as well as the meteorological parameters. HNO₃ concentrations were set constant from values in Petetin et al., (2016) for the same period of the year.”

Line 354-355: figure 6a shows temperature profile up to 2.5km, the reference to the tropopause level is misleading, rephrasing is needed.

Corrected. We have rephrased the sentence only mentioning the altitudes shown in the figure (Page 10 Line 378-380 was rephrased). “During the pollution event on 26-30 March, temperature shows the usual steady decrease with altitude from the surface up to 2.5 km (see the median temperature profile measured by radiosoundings launched at Trappes on 26-30 March, Fig. 6a).”

Line 285: the discussed PM_{2.5} levels are the results of simulations or observations ? do in-situ observations show similar levels?

Clarified. Within the mentioned sentence the clarification that the data are simulated is provided. As well as in the following paragraph, indicating that also the in-situ measurements are similar.

Line 302-304: “On 28 March, a clear enhancement of the aerosol load over the Benelux and northern France is observed both in terms of AOD (up to 0.4) and modeled surface PM_{2.5} concentrations (up to 50 µg m⁻³).”

Line 312-313 “As polluted air masses are advected from the Benelux and west Germany on 28-29 March, PM_{2.5} levels clearly enhance (up to 80 µg m⁻³), similarly seen in daily averaged simulations.”

Line 577-580: Does your model reproduce such behavior during night? The model results need to be discussed more and valorized. Does the model reproduces observed surface and profiles of NH₃? The reader remains with the question why not comparing the model results to the observations?

Clarified. Due to shortcoming of total column data during the night, no detailed nocturnal evolution was considered. However, P2 implies gas-to-particle conversion at the surface, that could result in lower total column values of ammonia in the early morning (for 2 days).

Moreover, we use ISORROPIA as an equilibrium model to perform sensitivity tests. However, we would need a full 3D chemistry-transport model in order to compare modeled concentrations with observations. This is clarified in lines 625-626 of the RM. “This issue would be best addressed with chemistry-transport model simulations. “

Line 241-242: Provide temporal resolution. Are these data used as input to Chimere model?

Clarified. The temporal resolution of CHIMERE is one hour. This is clarified in line 251 of the RM.

Line 132: particulate matter

Done.

Line 211: the following

Done.

Line 297: homogeneous

Done.

Line 375: decreasing ammonia

Done.

Lines 507-508: 'depicts: : : depicting.. the last one: : : ' (which one?), please rephrase the sentence.

Corrected. We have avoided the repetition of the work "depict".