

Interactive comment on “High resolution modelling of aerosol dispersion regimes during the CAPITOUL field experiment: from regional to local scale interactions” by B. Aouizerats et al.

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To Reviewer#2,

The authors appreciate the constructive and helpful comments provided by Reviewer#2. Its comments helped to improve our manuscript. The paper has thus been modified to take into account the recommendations given. Below, we have copied the referee comments in italics and inserted our responses in standard font where

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appropriate.

We also would like to mention that a bug has been found in the management of scalar tracers (as aerosols) by the shallow convection scheme. The aerosol concentrations are thus a little bit modified, especially for the ground concentrations of secondary species during the night, which is now lower. In that context, and in order to present a rigorous work, we redone the simulation with the bug corrected. therefore, the figures show different values, but the objectives and conclusions of this study remain the same. Moreover, we added a new figure showing the comparison of dynamical parameters between observations and model results. Finally, the fourth subfigures (previously 8d and 9d) were replaced by the horizontal cross section of the turbulent kinetic energy showing the roll structures along the whole thrid domain, and not only beginning where the aerosols are emitted.

Regards,

Benjamin Aouizerats

1 Specific comments

1. *This paper aims on reproducing the distribution of aerosol particles during different dynamical conditions by the use of high resolution simulations. Although the title and abstract give the impression that this paper is based on high resolution investigations of aerosol particle evolution in urban plumes and the impact of dynamics on the aerosol plumes, the main focus is put on the dynamical effect on the dispersion of the primary emitted aerosol component.*

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The abstract was modified to highlight that after analysing the aerosol plumes at regional scales, this study focuses on the local dynamical structures reproduced by the high resolution simulation and their impacts on the aerosol dispersion at local scales.

2. *In this study, high resolution simulations of down to 500 m are performed. For these investigations the aerosol-gas- phase thermodynamical equilibrium is treated by the EQSAM module, which was designed by Metzger et al., 2002 for the global scale. EQSAM was not developed for high resolution regional scale investigations, such as the present study, and treats the exchange between the gaseous and particulate phase simplified by assuming chemical equilibrium. The reviewer finds it contradictory to investigate the distribution of particulate matter by performing simulations with very high resolutions, but using such a simplified module as EQSAM. It is strongly suggested that the uncertainties of the assumptions made by using EQSAM are highlighted and discussed in the scope of this paper.*

We agree that the use of the EQSAM module does not consider the molecular diffusion. This is also the case for SOA that use the MPMPO solver (Griffin et al., 2005; Tulet et al., 2006). It is true to say that this choice corresponds to an assumption, where kinetic limitation of the molecular diffusion is not considered. We added a sentence to point out this limitation in the new version of the paper. However, for fine particles such as polluted aerosols, the typical timescale to reach the equilibrium between gas-aqueous phases is around several minutes. Even if the model time step is smaller than a minute, we think that the chemistry of SOA precursors and inorganic ions is stable over few minutes, and therefore that the equilibrium approximation is valid. It is much smaller than for coarser

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particles where it can take several hours (Wexler and Seinfeld, 1990). For instance, for a typical gas-phase diffusivity of $D_g = 0.1 \text{ cm}^2 \text{ s}^{-1}$ and a radius of $0.05 \text{ } \mu\text{m}$, the characteristic time for attaining a steady-state concentration profile around particles is around $0.625 \cdot 10^{-11} \text{ s}$ (Seinfeld and Pandis, 2006). At this stage, we do not have any numerical method to introduce kinetic transfer in our model and to quantify this limitation. I am not sure that rapid code exist for 3D chemical models able to solve (i) kinetic molecular transfer, (ii) the partitioning between the inorganic/organic gases and the aerosol phase, and (iii) the molecular affinity in the aerosol phase (which modified the gas-particles equilibrium).

The following three points justify our choice.

- 1) Currently most of CTM (not only for the global scale) such as CMAQ, CHIMERE, WRF-CHEM, ..., use diagnostics chemical equilibrium (ISORROPIA, ARES, ...) to solve the gas particles partitioning at the meso-scale, but also at the more local scale. To my knowledge many publications using chemical equilibrium at the mesoscale have been considered as acceptable (Jonson et al., 1998, Simpson et al., 2006 using EQSAM in HIRLAM model; Sakurai et al., 2005; Wu et al., 2005, 2007; Hu et al., 2007 using ISORROPIA in CMAQ model; or Pavlovic et al., 2006 using ISORROPIA in CAMx model). In addition, WRF and MesoNH use the same time step, so the error using chemical equilibrium are similar.

- 2) We choose EQSAM instead of ISORROPIA for numerical computation because ISORROPIA is not vectorized. Nevertheless, 0D tests between ISORROPIA and EQSAM give similar results.

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3) The focus of the paper is not to detail chemical composition. We do not have enough chemical observation for that. We also considered that the limitations given by using a chemical equilibrium are certainly less important than on surface chemical emission (particularly for VOC). The high resolution used in the paper is to model particular dynamical structure.

3. *It must be clear that the scope of this study is put on the investigation of the structure of urban plumes, under different conditions, and that the chemical evolution of the plume not is investigated.*

We emphasized in the study objectives section that the main purpose is to focus on the urban aerosol plume spatial distribution driven by the local dynamics. However, one of the objectives of this study is also to highlight that depending on the resolution used, as well as the region of interest size, the impact of local dynamics on pollutant dispersion may be considered, especially in places where the population is as concentrated as in cities. Concerning the plume chemical composition, it is true that the aerosol composition over the third domain is not shown. However, the variability between primary and secondary aerosol on a 25 by 25 km square, and over emission sources is less important than over the second domain (where the primary/secondary concentrations are shown Figure 3). This is the reason why we didn't find relevant enough to add the plume composition at the local scale. All of this may not have been clear in the previous version. We have now modified the text in order to clarify it.

4. *The following points should be clarified in the revised version of this manuscript: p. 29573 110. What input data is required for the town energy budget scheme?*

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Input for the TEB scheme are of 2 types:

- meteorological ones, that forces the schemes from above (above the urban canopy):

radiation from the sun (direct and diffuse), longwave radiation from the atmosphere, air temperature, humidity, wind, pressure, precipitation.

- urban ones, that describes the city (or part of the city, in our case, each 500m per 500m square):

All these parameters are aggregated. One does not simulate each building separately (with its own geometry and details). One simulate a "typical" road with one building on each side. For each grid mesh, one need land use parameters (mean fraction of building, mean fraction of roads), geometrical parameters (height of building, fraction of wall surface), and radiative (albedo, emissivity) and thermal (materials, insulation, heat capacity and thermal conductivity, thickness of walls and roofs) for each surface type: road, wall, roof.

The complete detailed list of these parameters are given in Masson (2000).

5. *Section 2.2. What about interaction between the aerosol particles? Is e.g., coagulation between the particles considered? Is there an interaction between primary emitted black carbon and the secondary aerosol particles.*

The dynamical processes (i.e. nucleation, coagulation, sedimentation and deposition) are resolved by ORILAM-SOA (Tulet et al., 2005; Tulet et al., 2006) as well as the growth of particles by condensation of secondary species on the primary emitted black carbon and organic carbon. We added details on the text about the aerosol evolution processes taken into account.

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6. p. 29574 | 16. *Please describe the treatment of background aerosols more detailed. This is an important factor for the investigation and the results. It is suggested to shortly describe the applied method at this place.*

For particles, the background aerosols were set up by deriving the *CO* concentration with a ratio obtained from Cachier et al., 2005 and which is similar to observations during the CAPITOUL campaign. The *CO* concentration in the atmosphere is initialised by the MOCAGE analyse fields corresponding to that period. We added details on the method to determine the background aerosol concentrations.

7. p. 29574. *The section 2.3 Emission inventory needs to be extended. Are emissions only considered from traffic sources? What about emissions from e.g., the industrial sector? Are emissions only available for the smallest domain? How are emissions for the simulations of the two other model domains treated?*

This part has been extended and more details are given in the text. Indeed it was unclear how the emissions were considered over the second and first domain. They are taken into account with nested horizontal resolutions which are the same than the simulation domain resolutions (i.e. 2.5-km and 10-km). Concerning the emission sources, we made the assumption of considering only the road transport emissions. The reasons are that firstly there are almost negligible domestic combustion in summer. Secondly the region of Toulouse is relatively clean of any major industrial sources. Moreover, industrial emissions are still very badly known, that is why we made the choice to neglect them and to be aware of the limitations we have, rather than introducing errors on emissions which would be more difficult to isolate and identify on the model results. The atmospheric concentrations being initialised by the MOCAGE fields, it appears

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reasonable to neglect the other industrial sources over France as their emissions reaching Toulouse are most likely low during those two days.

8. p. 29576. *Section 3.1.1. Cloudiness and temperature should be presented for the considered situation. These are generally important factors for the photochemistry.*

The IOP period was chosen because the meteorological situation was a summer anticyclonic case. Therefore the photochemistry was expected to be high. The cloudiness was very low the 3rd July, with the presence of some isolated cumulus and stratocumulus during few hours in the afternoon, and was nonexistent the 4th July. Concerning the temperature, we added a figure with the temperature evolution as well as other dynamical parameters, observed by several stations located over Toulouse and the larger Toulouse region, and compared with the model results. All these informations were added in the text.

9. p. 29576 | 23. *Are there only observations available in Toulouse? It would be helpful to have additional observations of the aerosol concentration in the investigated domains to evaluate the simulated horizontal distributions.*

Unfortunately aerosol observations were only available over Toulouse. However, dynamical observations were available over a greater region and are now presented in a new figure.

10. *Based on what information are you certain that pollution from Barcelona exactly is responsible for these high concentrations? Barcelona is not part of any of the model domains. Please clarify and motivate this statement.*

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We are not certain that the high concentrations at the end of the 4 July come from Barcelona, but that this air mass went over Barcelona area before entering the largest domain. The text was modified. However, model results show that the high concentrations at the southeast of Figure 2c come from outside the larger domain and are injected in the larger domain from the coupled MOCAGE field.

11. *p.29585 l8. What is the reason for this elevated aerosol layer? Which impact may this elevated layer of primary aerosol have on the SOA? What about the vertical distributions of the larger domains? Is the long range transport proven by the results of these simulations. Please comment.*

Indeed, the elevated aerosol layer is the result of the long range transport. At the end of the 4th July, a new air mass coming from the Mediterranean sea arrives above the Toulouse area. This new air mass does not include only primary aerosols, but also secondary particles. This elevated layer can be found on the larger domain several hours before reaching Toulouse at the southeast of the domain.

12. *The authors should not mix American English and British English (e.g., analyse/analyze, modeledze/initialize, modeled/modeled). The use of prepositions need to be looked over. In general, the English needs improvement and should be edited in the revised version*

The article was read and corrected by a native English speaker.