

*We are thankful to the reviewer for her/his help in improving the quality of the manuscript. Our detailed responses to the comments are given below in italic*

**The authors have included an interesting update to the chemical transport model CHIMERE, involving an online coupling between aerosols and radiative effects on photolysis. This interaction has been shown before to be important in extremely polluted locations like Beijing but its importance in cleaner parts of the world is not clear. They focus on a wildfire event in Russia in order to address this importance for the model's performance against evaluation data in Moscow when the plume passes over. The difference between the model predictions with and without the new coupling are modest for ozone predictions. The major weakness of the paper is that the discussion sidesteps the comprehensive impacts of the wildfire, and instead only mentions the model's performance against itself (specifics described below). I would recommend the authors add some of this to the manuscript or perhaps submit to a more appropriate journal like Geoscientific Model Development.**

*Thanks to the reviewer's remarks, the aerosol radiative feedbacks on photochemistry have been now discussed in a more comprehensive way in the revised manuscript (see our detailed responses below). Moreover, as suggested by the reviewer 2, the evaluation of model results has been also improved by additional statistical comparisons for PM10 components (ammonium, nitrates and sulphates) in the section 3.2 of the revised manuscript:*

	$NH_4^+$				$NO_3^-$				$SO_4^{2-}$			
	Mod.	Obs.	Corr.	RMSE	Mod.	Obs.	Corr.	RMSE	Mod.	Obs.	Corr.	RMSE
<i>with</i>	1.09	0.86	0.48	1.03	0.14	0.12	0.20	0.15	2.24	0.48	0.23	1.80
<i>without</i>	1.16	0.86	0.42	1.04	0.19	0.12	0.26	0.23	2.33	0.48	0.45	1.88

*Statistical comparisons between the near-surface concentrations of  $NH_4^+$ ,  $NO_3^-$  and  $SO_4^{2-}$  simulated with and without aerosol radiative feedbacks and measured at Moscow by an air quality station. Mod. and Obs. are the period-averaged modelled and observed concentration. Corr. and RMSE are the temporal correlation and the root mean square error.*

*As for  $NO_2$  and  $O_3$ , including the optical effect of aerosols in the photolysis calculations slightly improves the formation of secondary inorganic species in the CHIMERE model with a RMSE systematically reduced. The overestimation of sulphates levels is decreased and the simulated concentrations of ammonium and nitrates get close to the observed one. This result suggests that taking into account the aerosol solar extinction in the photolysis calculation gives an added value in the capacity of the model to reproduce the photochemistry under polluted environments.*

### **Major Issues:**

**1) The introduction is brief, and focuses mostly on the problem from the perspective of model development. Yet the larger environmental impact perspective is somewhat lost. In other words, the authors present the fire episode as an example where the model might screw up if it doesn't have correct optical feedbacks. But what is the nature of this model problem in the context of the effects of the fire on the total environment, or model domain? Is it emitting so many BC particles that the effects of photolysis on secondary particles are kind of negligible? Are the uncertainties from photochemical reaction rates swamped by uncertainties in precursor emission rates? A reader could leave this paper thinking that fire plumes act to decrease  $O_3$  concentrations, increase  $NO_2$  and slightly decrease PM10, and that all of these effects are mild. Of course the whole story involves massive emissions of a host of pollutants. I think this piece of the story needs to be emphasized more**

As suggested, questions relatives to fire events in the context of atmospheric composition and impacts have been better put into perspective in the Introduction of the revised manuscript by adding the following sentences :

« Also, the study of Chubarova et al. 2012 clearly shows that, during this specific wildfire episode, the aerosol optical thickness over the Moscow region was more than three times larger than the one observed during typical August conditions over the period 2001-2010. This suggests that, even if anthropogenic aerosols are present over the studied region, the contribution of smoke aerosols during this specific event is very large. Then, this case study represents an excellent opportunity to discuss how aerosol solar extinction, especially biomass burning particles, can affect photochemistry. Fires can affect atmospheric chemistry in several ways. They emit primary gaseous pollutants (such as CO, OH, NO, NO<sub>2</sub> and volatile organic compounds) that can react in the atmosphere to form ozone and other pollutants (Turquety, 2013). They also released aerosols that can directly affect air quality or indirectly by acting as a medium in complex heterogeneous reactions (Slade and Knopf 2013, Nie et al. 2015). Finally, they can affect the intensity of solar radiation, which in turn could affect photochemistry of the atmosphere. The latter impact is the subject of the present study. »

References quoted in the response and added in the revised manuscript :

- Chubarova, N., Nezval, Y., Sviridenkov, I., Smirnov, A., and Slutsker, I.: Smoke aerosol and its radiative effects during extreme fire event over Central Russia in summer 2010, *Atmos. Meas. Tech.*, 5, 557–568, 2012.

- Nie, W., Ding, A. J., Xie, Y. N., Xu, Z., Mao, H., Kerminen, V. M., Zheng, L. F., Qi, X. M., Huang, X., Yang, X. Q., Sun, J. N., Herrmann, E., Petäjä, T., Kulmala, M., and Fu, C. B.: Influence of biomass burning plumes on HONO chemistry in eastern China, *Atmospheric Chemistry and Physics*, 15, 1147–1159, 2015.

- Slade, J. H. and Knopf, D. A.: Heterogeneous OH oxidation of biomass burning organic aerosol surrogate compounds: assessment of volatilisation products and the role of OH concentration on the reactive uptake kinetics, *Phys. Chem. Chem. Phys.*, 15, 5898–5915, 2013.

- Turquety, S.: *The Atmospheric Impact of Wildfires. Fire Phenomena and the Earth System: An Interdisciplinary Guide to Fire Science*, Wiley Blackwell, 2013.

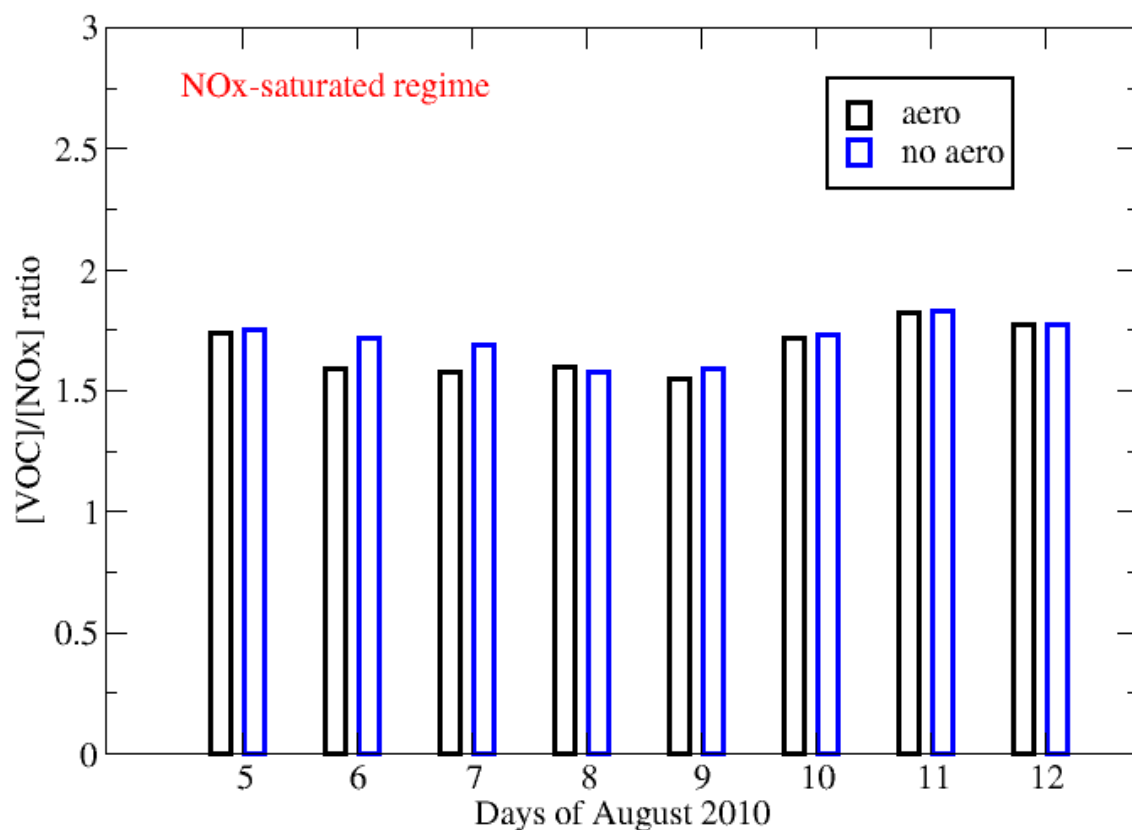
**2) Figure 5 simplifies quite a lot of information and is a useful graphic. It also puzzles one at first glance. The JNO<sub>2</sub> values have changed by about half of the change in JO<sub>3</sub> as AOT increases. Yet the total change in O<sub>3</sub> barely shows up (at about -3% at most severe). Clearly, the concentrations of NO<sub>2</sub> and O<sub>3</sub> are playing important roles here. I think it would be interesting to probe the model a little further and look at the actual rates of formation and destruction of O<sub>3</sub> to get a more intuitive feeling of how the balance is affected by the aerosol direct effect.**

Ozone formation is driven by two major precursors: nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) and volatile organic compounds (VOC) in a complex photochemistry. However, it is possible to identify two regimes of ozone formation by looking at the ratio between the concentrations of VOC and NO<sub>x</sub>: A NO<sub>x</sub>-limited and a NO<sub>x</sub>-saturated regime (Seinfeld and Pandis 1998). The Figure below indicates that the simulated photochemical regime was characterized by a NO<sub>x</sub>-saturated situation over Moscow during the studied period. In this case, inclusion of the aerosol radiative impact on photochemistry leads to two antagonists responses : (1) Increase of NO<sub>x</sub> concentration trough the reduction of their photolysis is unfavorable to ozone formation in a NO<sub>x</sub>-saturated environment. In parallel, (2) reduction of the ozone photolysis is favorable to its accumulation. The overall impact

of aerosols on the ozone concentration is then small due to these two antagonist responses.

This point has been now clarified in the revised manuscript by adding the figure below and corresponding comments in the Section 3.2 of the revised manuscript :

« Response of the ozone concentration under the aerosol radiative influence is more complex. Indeed, ozone formation is driven by two major precursors: nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) and volatile organic compounds (VOC) in a complex photochemistry. However, it is possible to identify two regimes of ozone formation by looking at the ratio between the concentrations of VOC and  $\text{NO}_x$  : A  $\text{NO}_x$ -limited and a  $\text{NO}_x$ -saturated regime (Seinfeld and Pandis 1998). Figure 6 indicates that the simulated photochemical regime was characterized by a  $\text{NO}_x$ -saturated situation over Moscow during the studied period. In this case, inclusion of the aerosol radiative impact on photochemistry leads to two antagonist responses : (1) Increase of  $\text{NO}_x$  concentration trough the reduction of their photolysis is unfavourable to ozone formation in a  $\text{NO}_x$ -saturated environment. In parallel, (2) reduction of the ozone photolysis is favourable to its accumulation. The overall impact of aerosols on the ozone concentration is then small due to these two antagonist responses, about 1 % per unit of AOT (see Figure 5). »



*Mean VOC to  $\text{NO}_x$  ratio simulated over Moscow during the studied period*

Reference quoted in the response and added in the revised manuscript :

- Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics. From air pollution to climate change, Wiley-Interscience publication, 1998.

**Also, why were the values here plotted for midday? Was it because it makes it easier to calculate AOT through a single column. As the authors say in the text, it's the time of day with the least atmospheric path.**

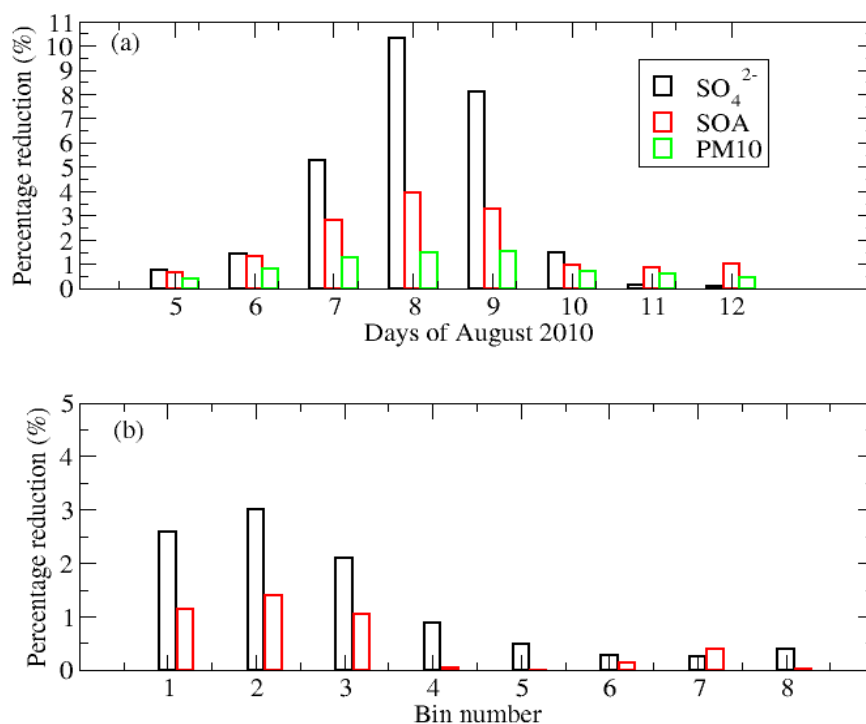
*There is no specific reason for this choice. To keep coherence with previous discussions, Figure 5 has been rebuilt with daytime average values in the revised manuscript.*

**3) Page 7070-7071, lines 24-4: I'm uncomfortable with this discussion and the conclusion that taking the photolysis impacts into account could have a non-negligible effect on the air quality prediction. It is clear from figure 7 that the model is quite wrong on August 8. Yes, the model with aerosol feedbacks does very slightly better, but I think the authors are overstating the importance of the model improvement when they imply that it can make the difference between exceedance and attainment in air quality models. It seems like a relative coincidence after all, that the model without aerosol feedbacks falls above the threshold and the one with feedbacks falls below**

*We agree with the reviewer. This conclusion has been now removed.*

**4) The authors focus almost exclusively on O3 and include one plot at the end of the manuscript addressing PM mass. However, just by looking at the substantial reductions in sulfate and OA, I would expect there to be a significant effect on aerosol growth and the size distribution in general. Can the authors discuss this? How are there 8 aerosol bins affected?**

*This is a very interesting suggestion. Figure 8 has been modified to include some discussions on the aerosol size distribution in the Section 3.2 of the revised manuscript:*



a) Daytime average percentage reduction of the near-surface concentration of sulphates, secondary organic aerosols and PM10 over Moscow due to the aerosol feedback. b) Repartition of this sulphate and SOA mass

*reduction between the 8 aerosol size bins for the 8 August*

« As illustrated in figure 9a, the maximum reduction in the near-surface concentrations of sulphates (oxidation product of SO<sub>2</sub>) and SOA (oxidation product of COV) occurs on 8 August with daytime average values of 10 % and 4 %, respectively. For this day, figure 9b shows that these changes are mainly due to a reduced formation of very fine particles, i.e with a diameter comprised between 40 nm (bin 1) and 150 nm (bin 3). The overall impact is then a slightly reduction of the total aerosol mass concentration (PM10) comprised between 1 and 2 % over the entire period (Figure 9a) »

**5) Since most of the discussion involves model development, I was surprised to not see a description of the computation time increase associated with incorporating the online coupling with the aerosols (including the core-shell calculations). This would help model developers decide whether or not they would like to implement the method in their own models (i.e. are the changes in O3 worth the time spent computing?)**

The following sentence has been added in the Section 2.3 of the revised manuscript :

« It should be noted that adding the aerosol impact on solar extinction in simulation (2) induces a computation time increase of 50 % compared to the simulation (1).

#### **Minor Issues:**

**1) The boundary conditions are not exactly consistent in time with the episode. Do the authors have an estimate for the influence of boundary conditions on their model domain? When predicting a very specific episode like this one, are they convinced that the lack of time dependence at the boundaries is acceptable?**

*The evaluation study of Péré et al. 2014 showed that the fire inventory of Kaiser et al. 2012 used in this work combined with the CHIMERE model have been shown to well capture the evolution of the Russian fire plume during the studied period, suggesting a low influence of these boundary conditions.*

*This point has been clarified by adding the following sentence in the section 2.1.1 of the revised manuscript :*

« *The evaluation study of Péré et al. 2014 showed that the fire inventory of Kaiser et al. 2012 used in this work combined with the CHIMERE model have been shown to well capture the evolution of the Russian fire plume during the studied period, suggesting a low influence of these boundary conditions.* »

*References quoted in the response and included in the revised manuscript :*

*- Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J. J., Razinger, M., Schultz, M. G., Suttie, M., and van der Werf, G. R.: Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, Biogeosciences, 9, 527–554, 2012.*

*- Péré, J. C., Bessagnet, B., Mallet, M., Waquet, F., Chiapello, I., Minvielle, F., Pont, V., and Menut, L.: Direct radiative effect of the Russian wildfires and its impact on air temperature and atmospheric dynamics during August 2010, Atmospheric Chemistry and Physics, 14, 1999–2013, 2014.*

**2) What version of WRF is being run offline? A little more detail on some of the weather selections for WRF would not be inappropriate. After all, the indirect effect calculations and the convective transport modules are relevant to this study.**

The configuration of WRF used in this study has been now better described by adding the following sentences in the section 2.1.1 of the revised manuscript :

*«The version 3.1 is used in this study with the same configuration as in Péré et al. (2011). It has 27 vertical levels from 40 m to about 20 km and includes the following parameterizations: the WRF single-moment five-class scheme of Hong et al. (2006) for the microphysics module, the Kain-Fritsch cumulus parametrization (Kain, 2004), the NOAA land surface module of Chen and Dudhia (2001) and the Yonsei University planetary boundary layer scheme (Hong et al., 2006; Hong, 2007).»*

References quoted in the response and added in the revised manuscript :

- Chen, F. and Dudhia, J.: Coupling an advanced land-surface / hydrology model with the Penn State / NCAR MM5 modeling system. Part 1: Model description and implementation, *Mon. Wea. Rev.*, 129, 569–585, 2001.
- Hong, S. Y., Noh, Y., and Dudhia, J.: A new vertical diffusion package with an explicit treatment of entrainment processes, *Monthly Weather Review*, 134, 2318–2341, 2006.
- Hong, S. Y.: Stable boundary layer mixing in a vertical diffusion scheme, *The Korea Meteor. Soc.*, fall conference, Seoul, Korea, Oct. 25-26, 2007.
- Kain, J. S.: The Kain-Fritsch convective parameterization: An update, *J. Appl. Meteor.*, 43, 170–181, 2004.
- Péré, J. C., Mallet, M., Pont, V., and Bessagnet, B.: Impact of the aerosol direct radiative forcing (ADRF) on the radiative budget, surface heat fluxes and atmospheric dynamics during the heatwave of summer 2003 over Western Europe. A modelling study., *Journal of Geophysical Research*, 116, d23119, doi:10.1029/2011JD016240, 2011.

**This is quite relevant for figure 6, which might suggest that emissions from the fire are being injected too high.**

*As previously shown in details in the work of Péré et al. 2014, except for a lack of CALIOP measurements below 0.6 km, the model is within or close to the uncertainty range of observations except at 2 to 4 km where the model tends to largely overestimate CALIOP values (model biases from 30 to 350 % compared to observed median values). However, this part of the aerosol profile represents less than 30 % of the total aerosol extinction, suggesting that model uncertainties on the extinction vertical distribution remain limited.*

**3) What is TUV doing with sub-grid clouds? The grid cells (30 km) are somewhat coarse even for a regional simulation when the study is focusing on a plume**

*In TUV, clouds are assumed to be horizontally homogeneous layers. Hence, sub-grid cloud processes are not taken into account.*

*The following sentence has been added in the section 2.2 of the revised manuscript :*

*« In TUV, clouds are assumed to be horizontally homogeneous layers and are considered to be of three types : low, middle and high altitudes clouds. »*

**4) Page 7066, lines 10-23: While there is no denying that photolysis is critical for ozone formation in the atmosphere, the authors should also mention briefly the importance of VOCs in catalyzing ozone formation. The explanation given seems incomplete. This issue further relates back to the importance of emission inventory uncertainties compared to photolysis rate estimation uncertainties.**

*The reviewer is right. To clarify this point, the following sentence has been added in the Section 2.3 of the revised manuscript :*

*« The sequence of reactions (R1 to R4) is generally initiated by the reaction of various VOC with the OH radical. »*

**5) Figures 2 and 3: Please consider putting percent signs near the color axes for easy reference.**

*OK, This is now added.*

**Also, are these data for the lowest layer of the model or some column average?**

*Yes, the simulated values are for the lowest model layer. The term « near-surface » has been added in the captions.*

**May I suggest the authors replace “diurnal-average” with “daytime average” here and throughout the text?**

*Ok, this is now done.*

**6) Page 7069, lines 6-9: This statement seems true at first read, but certainly depends on the aerosol concentration, which will vary day to day and isn't necessarily a minimum at midday.**

*We agree with this remark. This statement has been removed and replaced by discussions on the daytime average values (please see response above about the major issue 2).*

**7) Figure 8: Are the SO<sub>4</sub> and SOA species for PM<sub>2.5</sub> aerosols ?**

*Yes, they are present in the fine aerosol mode.*

**Presentation Issues and Typos:**

**1) Page 7065, line 28: Please consider putting the description of the simulations in their own section so that it is clearer to the reader that the computational experiment is being described. As it is now, the information is somewhat buried.**

*The subsection « On-line coupling between CHIMERE and TUV » has been renamed « Simulation set-up » in the revised manuscript.*

**2) Page 7058, line 15: In “terms” of air quality ...**

**3) Page 7058, line 26: “the exceedance of ...” and please change exceed to exceedance as appropriate throughout the text.**

**4) Page 7060, line 6: Please replace ‘emphasize’ with ‘focus’. There are many other English language typos throughout the paper. These don’t detract from the scientific quality of the paper, but they do distract. Please have someone look over the manuscript with the intention of correcting these mistakes**

*Thank you, the corresponding sentences have been now corrected in the revised manuscript. Also, we have now put a significant effort in improving its grammar quality.*