Interactive comment on “Atmospheric impacts of the 2010 Russian wildfires: integrating modelling and measurements of the extreme air pollution episode in the Moscow megacity region” by I. B. Konovalov et al.

Anonymous Referee #1

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General Comments:

This paper analyzes the contribution of wildfire emissions to surface concentrations of CO, PM10 and ozone over the Moscow metropolitan region during the summer of 2010. The pollutants are simulated from 28 May to 31 August 2010 using the regional chemistry transport model CHIMERE (forced by MM5 for meteorological fields) with the horizontal resolution of 0.2x0.1 degrees (dx∼20km). The emissions of wildfires are derived from the MODIS fire radiative power (FRP), and their intensity is adjusted to match the surface observations of CO and PM10. Authors estimate that 9.7 Tg of CO from fires are needed during this summer episode to reproduce the CO concentrations at the surface, which represents more than 85% of the total annual CO emissions from anthropogenic sources. Although this top-down estimate of the fire emissions is based on numerous assumptions and is therefore highly uncertain, it provides an interesting alternative to the more traditional direct estimates of fire emissions from satellite data (e.g. Wiedinmyer et al., 2006). The two approaches are complementary and the conclusions of the paper would be much stronger if authors could compare their results with other estimates/studies.

After adjusting the fire emissions, authors evaluate CO, PM10 and O3 concentrations against surface data. The sites used for this comparison were not used for the retrieval/optimization of the fire emissions but it is unclear if they were truly independent (located far enough from sites used for the optimization). The comparison at the surface shows that fires are the main contributors to elevated CO and PM10 levels during the 3-10 August episode and that by including their emissions the model provides a much better agreement with the daily mean observations of these species. However, the comparison with the truly independent MOPITT CO data shows large discrepancies in the boundary layer that need to be addressed. A very large gap (∼300 ug/m3) between observed and modeled ozone levels is also found during this intense fire episode (7 August). This indicates that the processes involved are not modeled accurately. For example, the emissions of ozone precursors might be inaccurate in terms of amount and vertical distribution, or the assumptions used to calculate the impact of aerosols on photolysis might be too crude for this case study. The reasons for these discrepancies need to be explained and justified in the paper.

Overall I find that the paper is suitable for publication in ACP after the concerns listed in this review have been fully addressed. I also find that Drs. Yurganov and Chubarova have raised important concerns in their comments, and it is very disappointing that they have not been addressed in the review process.

Major concerns:
1) Uncertainties in the fire emission estimates

- The approach applied in this study to determine the fire emissions is based on numerous assumptions. I would like to see all of the assumptions and the associated uncertainties summarized in a new Table. E.g. PM10 are assumed to be primary species during the fire event; associated uncertainty XX-XX% (reference if available).

- The fire emission flux is very sensitive to the choice of the correction factor C (p.12160, l.10-20). Authors use the value of \( \exp(1*\text{AOD550nm}) \), whereas the AOD at 4um should be used. According to the AERONET measurements in Moscow for the polluted day of 7 Aug. the \( \text{AOD}_{500\text{nm}} \) is \( \sim 2.5 \) times larger than the \( \text{AOD}_{1\text{um}} \), and likely even larger than the one at 4um. Therefore, for this particular day the correction factor and the emissions are overpredicted by at least a factor of 2.

In order to better constrain this parameter and reduce the uncertainties I suggest that authors scale the MODIS AOD550 as close as possible to the 4um wavelength using the ratios calculated daily from the AERONET dataset. AOD at 1um and 500nm could be used to determine this scaling ratio.

- The comparison with CO data from MOPITT shows some intriguing differences at 900hPa. Not only the background values seem low by 30ppb, but also the dCO due to fires is not well captured in the model during the first part of August. The increase due to fire emissions ranges from 5-10ppb in the model, whereas the observed values seem to be \( \sim 20\text{ppb} \) higher than the background (assuming that observations follow a similar temporal pattern). The comparison with this totally independent dataset suggests that there might be an underprediction of a factor of 2 of CO fire emissions in the model. Dr. Yurganov also mentioned this possible underprediction in his comment. This issue needs to be addressed in the revised paper. Showing that the surface CO data matches the observations after adjusting the emissions with surface stations (even if the two groups of stations are independent) is not a robust conclusion given the gap at 900hPa. Adding a plot of CO vertical profiles from CHIMERE might also help understand and explain this discrepancy.

- A map showing the location of surface stations used for the optimization and for the model evaluation needs to be added to the paper. It is unclear so far which sites have been selected (and how) for optimization vs. evaluation process.

- Finally, it would be very beneficial (especially given all these uncertainties/assumptions involved) to compare the Konovalov et al. fire emission estimates with other studies. The results of Yurganov et al., ACPD 2011 and the differences between the two approaches must be discussed in the paper. This might help quantify the uncertainties associated with the emission estimates of the present paper. As I already mentioned, it would of great interest (if possible) to compare the results of the present paper with the “traditional method” which uses the area burned to derive the fire emissions.

2) Impact on photochemistry and ozone levels As I already mentioned above, a very large gap (\( \sim 300 \text{ug/m3} \)) between observed and modeled ozone daily max levels is found during the fire episode (7 August) and the reasons for it need to be explained.

The increase in ozone precursors due to fire emissions seems to be responsible for a large increase in ozone (\( \sim 600 \text{ug/m3} \)) on 7 Aug. (compare TEST_3 and TEST_4 runs). These values seem very high. What is driving this increase? VOC emissions from fires could not be evaluated in the paper, but their ratios with CO could be compared to other studies from the literature.

Luckily this increase due to emissions of fire precursors is counterbalanced by the reduction in photolysis rates (\( \sim 40\% \)) that was assumed for absorbing (\( \text{ssa}=0.8 \)) and vertically uniformly distributed aerosols. However the model is still 300ug/m3 too high in comparison to the observations. This suggests that the assumptions on the aerosol feedbacks on photolysis might be too crude in the model and needs to be further examined.
Indeed, according to the short comments posted on the acpd website, the SSA measured in this region in presence of fires seems to be higher >0.9. This might be due to the presence of large fraction of secondary organic and inorganic material in this polluted region. I suggest that authors either calculate the SSA using the aerosol composition that was predicted by the model, or provide upper and lower estimates for ozone impact for SSA varying from 0.96 to 0.8. Authors should perform an offline calculation of photolysis rates and their attenuation using the TUV model which can be downloaded from: http://cprm.acd.ucar.edu/Models/TUV/ for this single day (7 Aug).

3) Introduction

I suggest that authors provide more details and literature references on the approach they have applied in the paper. The paragraph p.12144 l.27 – p.12145 l.7 could be expended and e.g. the results of Ichoku and Kaufman, 2005 further discussed. The results of previous modeling studies and data analysis of this 2010 episode or previous similar episodes in the Moscow region should be added too (e.g. work by Yurganov et al., and Chubarova et al.). The paragraph (l.7-19 p12145) that is listing the global model studies without however describing their major findings could be omitted and more focus could be put on regional CTMs studies.

Also, it is not clear (as presented here) why the approach using FRP is better for real time assimilation systems and forecasting purposes than the traditional approach using the area burned from that MODIS fire counts. To me it seems that the traditional approach is easier to use for this purpose as it only require MODIS data, whereas the approach presented here also requires data from the surface stations for the optimization process, and those data might be more difficult to get in real time.

I disagree with the statement p.12144 l.18-26. MODIS fire counts provide data at a much higher resolution i.e. 1x1km2 and are frequently used to determine daily mean fire emissions and to model their impacts on air quality using CTMs. This entire paragraph needs to be modified. The approach using FRP and the top-down approach should be presented as a complementary way of retrieving the fire emissions and the authors should express the need for comparing it with the traditional one.

p.12146 l.25: This sentence should be modified as we already know that this approach is feasible... as already done by others e.g Sofiev et al., 2009.

4) I think that the sensitivity study to determine the role of heterogeneous reactions should be removed, as it is highly uncertain due to uncertainties in the aerosols. Authors could just briefly mention that they did the sensitivity study, and that they found xx% difference with the reference simulation.

5) How is the model performing in terms of meteorological variables? Figure 9 shows the observed parameters. Please add on that same plot the model predictions. To my understanding, authors use MM5 at 1x1 degrees to drive the meteorology, however this resolution is too coarse to force their inner domain. Please clarify this in the paper.

Having the correct meteorological parameters during this episode is crucial in order to get the correct amount of dilution of the smoke plume. In particular, I would like authors to show that CO surface concentrations are not overpredicted during the nighttime in the model due to insufficient PBL mixing. Showing this is very important as authors are comparing daily mean values with the observations, and any overprediction during nighttime can greatly affect the average concentrations, and might hide the underprediction of the daytime fire emission fluxes. This needs to be addressed in the paper.

Minor modifications:

Authors should avoid using general terms such as e.g. “Atmospheric impacts” or “state-of-the-art” models as they do not reflect the work done here nor are appropriate for the tools used in this paper.

-Title: “Atmospheric impacts” Is not appropriate here as authors are only focusing at the impacts limited to the boundary layer and not treating the transport of the smoke into the upper troposphere or stratosphere. I suggest using “Tropospheric impacts” of
"Air quality impacts". I suggest that the term “megacity” be omitted from the title, as it is obvious that Moscow is a large metropolitan area, or written as “of an extreme air pollution episode in the megacity of Moscow” (in this case the regional aspect is lost).

Abstract p.12142: -l.4: “megacity” should be removed. -l.4-10: change to: “The paper analyses the evolution of the surface concentrations of CO, PM10 and ozone over the Moscow region. . . results of a mesoscale model. The CHIMERE chemistry transport model is used and modified to include the wildfire emissions of primary pollutants and the shielding effect of smoke aerosols on photolysis under the assumption of highly absorbing particles.” -l.15-16: change to “The model results show that wildfires are the principal. . . with the extremely high levels of daily mean CO and PM10 concentrations (up to 22 ppm and 700 ug/m3 . . . on 7 August).” -l.20-23: change the sentence starting by “In contrast to” “However, ozone concentrations were simulated to be very high (>1,000 ug/m3) even when the fire emissions were omitted in the model. It was found that the smoke has the tendency to increase ozone production by providing more precursors for ozone formation, but also to inhibit the photochemistry by absorbing the solar radiation.” Scattering cannot be used here as these results were obtained under the assumption of highly absorbing aerosols.

Introduction: -p.12143 l.6-7: Change to “Several severe air pollution episodes occurred during this period in number of Russian regions.”

-p.12143 l.11&15: The use of terms such as “extreme perturbations,” “extreme air pollution episodes” or “critical test” is not justified in this paragraph. It is unclear so far in the text to what these terms are referring to. Are we talking here about specific meteorological conditions that are unusual compared to the climatologically values, or very high levels of pollutants, or both? This needs to be explained, and quantified.

-The term “state-of-the-art models” is being used frequently in the paper and should be replaced by “current models”. I would argue that there is not such a thing as the state-of-the-art model especially when talking about CTMs or GCMs that rely on e.g. reduced chemical mechanisms, parameterized aerosol feedbacks, offline meteorology, tabulated photolysis rates, or missing fire emissions as discussed in the paper.

-p.12144 l.4: “wildfires” instead of “wild fires”

-p.12150 l.20-22: remove the following sentence “the network..” -p.12157 l.4: use “the model outputs were processed.”

-p.12160 l.24: l24-25: Replace the sentence starting by “therefore, setting..”, by “Therefore, k was set to 1 in this study, and the sensitivity to this parameter is examined in section 5.3.”

-Please justify this statement: “in the considered situation with.”.

-p.12168 l.4-7: this statement is inexact, I believe that organic aerosol formation is at least as complex as the ozone formation. And this erroneous statement is not an acceptable justification of the ozone low values found on 7 Aug, and the model gap found on this day. This needs to be revised.

-p.12168 l.8-13: this paragraph is more suitable for the introduction.

-p.12169 l.1: use “Small improvements in ozone simulations may.”

-p.12169 l.22-23: use “First, wildfire emissions favor.”

-p.12169 l.29: do not use base case for the FE run, as it gets confused with the reference case.. instead name this case the “fire case”.

-p.12172 l.21: MOPITT instead of MOPPIT. l.1: remove “megacity” from the title of this section.

-p.12173 l.20 modify to “satellite data and ground..” l.24: modify to “the model was modified to take into account”

-Conclusion: authors need to be more specific and quantify the differences between the model runs and with observations. P12173-l.25 add “due to the assumed shield-
ing effects of aerosols. P12174-l.5: add “at 550nm and assuming that aerosol single scattering albedo of 0.8.” P12174-l.10-11: this is not true for ozone on 7 Aug. P12174-l.14: add “performance at the surface”. P12174-l.24-29: this sentence is confusing and needs to be rephrased.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12141, 2011.