**Atmospheric
Chemistry
and Physics
Discussions**

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.

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Response to the comments of the anonymous referee # 1

We thank the reviewer for the thorough critical evaluation of our paper. All of the reviewer's comments are very carefully addressed in the revised manuscript. Below we describe our point-to-point responses. In earlier interactive comments (Konovalov, 2011a,b; Beekmann, 2011), we already had responded to some of the critics, and we had regretted that the review does not take into account the paper's positive results.

Comment: 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).

In a general case, magnitudes of uncertainties in the estimated fire emissions depend on the temporal and spatial scales considered (for example, they are expected to be larger for individual grid cells but smaller for big regions). Uncertainties in many factors (of quite different nature) contributing to the overall uncertainty of our calculations are unknown, and it is also unknown how these uncertainties (e.g., uncertainties in FRP data) combine and propagate in the preliminary data processing and the consecutive model simulations. We can assess the overall accuracy of our emission estimates only in an indirect way by comparing model results with measurements (similar to what was done in many other modeling studies). Taking these facts into account, we do not see a way to summarize our whole procedure and uncertainties associated with individual steps in one table, without oversimplifying the situation and without presenting unjustified numbers. However, to address this reviewer's comment we have introduced an additional section (Section 4.3) summarizing major assumptions and potential uncertainties in our results. In particular, we discuss potential biases in our emission estimates which should be considered together with random uncertainties evaluated formally by means of the Monte-Carlo method.

Comment: 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*AOD550nm), whereas the AOD at 4um should be used. According to the AERONET measurements in Moscow for the polluted day of 7 Aug. the AOD_500nm is 2.5 times larger than the AOD_1um, 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 500 nm could be used to determine this scaling ratio.

As it was already noted in the initial paper (Section 4.1), "relating AODmodis and tau_4 is not easy, because not only optical properties of aerosols but also the spatial structure of tau_4 on fine scales which are not captured by AODmodis data should be taken into account. For example, heavy smoke over a single fire could completely obscure it from a satellite sensor, but, at the same time, corresponding AODmodis value (representing a much larger territory) would not be significantly different from a background value." Accordingly, we believe that the required AOD at the 4um wavelength cannot be obtained in the considered situation by scaling the MODIS AOD550 using the ratios calculated from the AERONET dataset. We tried to further clarify this point in the revised version of the paper.

Comment: 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 20ppb 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.

This reviewer's comment was already addressed in the interactive comment by M. Beekmann (2011). In particular, he argued that "we used the comparison with MO-PITT measurements only as a way to demonstrate the spatial extent of air pollution

C8802

caused by wildfires" and that "this comparison does not permit deriving any quantitative conclusions about CO emissions".

To put less emphasis on the attempted comparison of model results with MOPITT measurements in the paper, the figure showing time series of CO mixing ratios at 900 hPa (Fig.16) is replaced by the plots presenting spatial distributions of maximum perturbations of surface CO and ozone concentrations during the considered period. Additionally, to strengthen our arguments about an important role of boundary conditions in our simulations, we have performed a model run using zero CO mixing ratios as both the top and lateral boundary conditions. It was found that in unperturbed atmosphere (without fire emissions) the average CO mixing ratios (processed with the MOPITT averaging kernels) at 900 hPa over the CER region during July and August 2010 were more than three time less than the corresponding value calculated with the boundary conditions from MOZART. This test confirmed a strong impact of the boundary conditions on the model results compared with the MOPITT measurements. A corresponding remark is added to the revised manuscript.

At the same time, we recognize (please see Section 5.1) "that we cannot claim that our estimates concerning the European part of Russia or the whole Europe are sufficiently constrained by measurements, because measurements in Moscow are mainly sensitive to emissions in the CER region." That is, uncertainties in the fire emissions and their vertical distribution in the troposphere may also contribute to the differences between our simulations and the MOPITT data, as noted in Section 5.4 of the revised manuscript. To illustrate the range of possible uncertainties in available emission estimates, a comparison of our estimates with GFED3 data and results by Yurganov et al. (2011) is added to the revised paper (see Section 5.1). In particular, we found that our estimates are, on the average, about a factor of three larger than the corresponding values obtained from GFED3. This means, in particular, that using GFED3 data in our simulations could not bring our results closer to the MOPITT measurements. On the other hand, the estimate obtained by Yurganov et al. for a much larger region than

C8803

that considered in our study is only about a factor of two larger than a corresponding GFED3 estimate. That is, if we assumed that both the spatial distribution of fire emissions in the GFED3 inventory and the estimate by Yurganov et al. are correct, we would get an even larger discrepancy between our simulations and the MOPITT data.

Comment: 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.

Maps showing the location of air pollution monitors in Moscow are added to the revised manuscript (Fig. 2). It was already mentioned in the initial paper (p.12151, l.9) that the sites were distributed into two groups randomly (please see also the comment by M. Beekmann, 2011 on this issue).

Comment: 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.

For comparison with our estimates, we calculated CO emissions for the selected regions using the gridded (with the 0.5 by 0.5 degree resolution) monthly data of the Global Fire Emissions Database version 3 (GFED3) (URL: http://www.globalfiredata.org). These data (for the year 2010) were not publicly available when the discussion paper was in preparation. The corresponding values are added to Table 4. The results of Yurganov et al. (2011) are also discussed in the revised manuscript. In particular, we argue that even disregarding potential uncertainties in the estimates obtained by Yurganov et al., the direct comparison of their estimates

with our results is not possible, because of large differences in the considered regions (Yurganov et al.: all of Russia plus parts of Kazakhstan and Mongolia; our study: a European part of Russia).

Comment: Impact on photochemistry and ozone levels. As I already mentioned above, a very large gap ($\sim$300 ug/m3) between observed and modeled ozone daily max levels is found during the fire episode (7 August) and the reasons for it need to be explained. The increase in ozone precursors due to fire emissions seems to be responsible for a large increase in ozone ($\sim$600 ug/m3) on 7 Aug. (compare TEST_3 and TEST_4 runs). These values seem very high. What is driving this increase? VOC emissions from fires could not be evaluated in the paper, but their ratios with CO could be compared to other studies from the literature. Luckily this increase due to emissions of fire precursors is counterbalanced by the reduction in photolysis rates ($\sim$40%) that was assumed for absorbing (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).

To address this comment, we have introduced the TUV (v.5.0) model into the meteorological interface of CHIMERE, and this important modification allowed us to perform off-line calculation of photolysis rates of each model species for each grid cell of the three-dimensional domains of the model as a function of the zenith angle and aerosol

optical depth measured by MODIS at 550 nm. All our simulations were then repeated with the improved photolysis scheme and SSA=0.95. The results presented in the paper are accordingly updated. As it was expected (Konovalov, 2011a), this change had a very small effect on CO and PM10 concentrations and fire emission estimates. The changes in daily variations of ozone were more significant; however, the performance statistics changed quite insignificantly, and the major conclusions of the study remained the same. The comparison of the updated results with our earlier simulations indicated that the aerosol effect on photolysis rates was strongly underestimated by the employed rough parameterization (see Eq. 4 of the discussion paper) in episodes with very high AOD values observed over the Moscow. The agreement of simulated concentrations with measurements on August 7 is improved, although it is yet not perfect. Additionally, we present a simulation obtained with SSA=0.8 (see Fig. 15, TEST_5 in the revised manuscript). The results of this test show that ozone concentrations at surface during the considered episode are rather insensitive to changes of SSA (the differences between the two case do not exceed 7 percent, but mostly much smaller). We think that this test provides an exhaustive answer to the concerns expressed by the anonymous referee and by N. Chubarova (2011).

In the updated simulations, the ratio of VOC to CO emissions from forest fires was exactly the same (see Sections 4.1 and 4.2) as that recommended by Wiedinmyer et al. (2006). Note that this ratio was increased by about 50 percent in comparison to a value specified in the earlier simulations (please compare the last sentence in Section 4.2 of the both versions of the paper); the ozone response to the corresponding increase of ozone precursor emissions in the strongly polluted atmosphere was found to be relatively small (less than 7 percent). The ratio of VOC to CO emissions from peat fires was also based on values provided in literature discussed in Section 4.1.

Comment: 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.

The discussion of the approach applied in the paper is extended in the revised manuscript. The references to publications by Yurganov et al. and Chubarova et al. are added. The paragraph listing the global model studies is shortened and merged with the next paragraph.

Comment: 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.

This question was already addressed in the interactive comment by M. Beekmann (2011). The discussion of this point in the revised manuscript is accordingly modified. In particular, we note that this approach seems to be very suitable for near real time data assimilation systems, particularly because it allows (at least, in principle) estimating emissions from currently active fires and taking into account differences in smoke emission rates from different active fire pixel. We do not claim, however, that this approach is better than the traditional approach, because special studies are needed to justify this.

Comment: 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.

We meant that available data of global fire emission inventories (like GFED) cannot be used in regional models without additional processing aimed at increasing their spatial and temporal resolution. This paragraph is modified following the reviewer's recommendations.

Comment. 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.

This comment was also addressed in the interactive comment by M. Beekmann (2011). The mentioned sentence is modified, such that our goal is formulated more accurately.

Comment: 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.

This sensitivity test is removed. Instead, a corresponding remark is added to Section 3.1.

Comment: 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.

Evaluation of the meteorological data is far beyond of the scope of this paper. In particular, we believe that a meteorological model cannot be properly evaluated by comparing simulated data with measurements made in just one point (especially in the case of precipitation and instantaneous wind speed). Nonetheless, in order to satisfy the reviewer's request, we added model predictions for temperature and precipitation to

Fig. 9. The data for the wind speed (at 00 UT) are removed entirely to insure readability of the figure and also taking into account low spatial representativity of daily variations of the available observations of this characteristics. Our choice of the resolution of the meteorological model is explained in Section 3.1 of the revised manuscript.

Comment: 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.

We considered the daytime and nighttime CO and PM10 concentrations separately and concluded that the model reproduced mixing processes during night-time rather adequately, although not perfectly. The average values of the daytime and nighttime concentrations are reported in a special paragraph added to Section 5.2.

Comment: 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).

We believe that the current title is not misleading. Indeed, it consists of two parts: the first part indicates the broad topic of interdisciplinary research (and appeals to a larger audience), while the second part specifies the concrete subject of this study. By saying: "an extreme air pollution episode in the Moscow region" we imply that the

paper focuses on the impact of the fires on the boundary layer in a specific region. The term "megacity" is removed from the title of the revised manuscript.

Comment: 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 widlfires are THE principal.. with the extremely high levels of daily mean CO and PM10 concentrations (up to XX ppm and 700 ug/m3 : : : on 7 August)."

The requested changes are made.

Comment: l-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.

The changes are made.

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

The requested change is made.

Comment: 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 meterological conditions that are unusual compared to the climatologi-

cally values, or very high levels of pollutants, or both? This needs to be explained, and quantified.

The sentence is revised. We tried to make it clear that we are talking about "the extreme perturbation of atmospheric composition" which provided a critical test for the current understanding of atmospheric "chemical and meteorological" processes. The extent of perturbations is qualitatively outlined in the previous paragraph complemented (in the revised version) by two additional references where a reader can find some quantitative details. The considered statement could not be quantified in the given context because, on the one hand, too many atmospheric parameters were strongly perturbed, but, on the other hand, there are so far very few reliable publications on this subject which could be cited in a scientific paper.

Comment: 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.

The term "state-of-the-art models" is not used in the revised manuscript. However, we would like to note that, this term is quite frequently used in scientific publications (Google returned 1,770,000 references), and that acceptable simplifications which make CTM's computationally affordable should be considered as "state-of –the art" within CTM's.

Comment: -p.12144 l.4: "wildfires" instead of "wild fires"

The correction is made.

Comment: p.12150 l20-22: remove the following sentence "the network.." -p.12157 l.4: use "the model outputs were processed.."

The corrections are made.

Comment: 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.

The corrections are made.

Comment: -p.12164 l.9-10: Please justify this statement:" in the considered situation with..".

The statement is revised.

Comment: -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.

The words "reflecting a more complex nature of ozone evolution" are removed.

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

In this paragraph, we discuss the results of our analysis of monitoring data; these results could not be presented in Introduction before Section 2.2 where the source and processing of monitoring data are described.

Comment: -p.12169 l.1: use "Small improvements in ozone simulations may.."

The sentence is revised.

Comment: p.12169 l.22-23: use "First, wildfire emissions favor.."

The change is made.

Comment: -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".

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The change is made.

Comment: p.12172 l.21: MOPITT instead of MOPPIT. l.1: remove "megacity" from the title of this section.

The change is made.

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

The change is made.

Comment: Conclusion: authors need to be more specific and quantity the differences between the model runs and with observations. P12173-l.25 add "due to the assumed shielding 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.

The conclusions are revised following the reviewer's recommendations. Specifically, we added more quantitative details concerning consistency of simulated ozone concentrations with measurements.

References:

Beekmann, M., Interactive comment on "Atmospheric impacts of the 2010 Russian wildfires: integrating modeling and measurements of the extreme air pollution episode in the Moscow megacity region" by I.B. Konovalov et al., Atmos. Chem. Phys. Discuss., 11, C5821–C5825, 2011.

Chubarova, N., Interactive comment on "Atmospheric impacts of the 2010 Russian wildfires: integrating modeling and measurements of the extreme air pollution episode in the Moscow megacity region" by I.B. Konovalov et al., Atmos. Chem. Phys. Discuss., 11, C2519–C2521, 2011.

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Wiedinmyer, C., Quayle, B., Geron, C., Belote, A., McKenzie, D., Zhang, X., O'Neill, S., and Wynne, K. K.: Estimating emissions from fires in North America for air quality modeling, Atmos. Environ., 40, 3419–3432, doi:10.1016/j.atmosenv.2006.02.010, 2006.

Yurganov, L. N., Rakitin, V., Dzhola, A., August, T., Fokeeva, E., George, M., Gorchakov, G., Grechko, E., Hannon, S., Karpov, A., Ott, L., Semutnikova, E., Shumsky, R., and Strow, L.: Satellite- and ground-based CO total column observations over 2010 Russian fires: accuracy of top-down estimates based on thermal IR satellite data, Atmos. Chem. Phys., 11, 7925-7942, doi:10.5194/acp-11-7925-2011, 2011.

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