We would like to thank the reviewer for the very useful comments, which help to improve the quality of the text. By green color we show edited or new text in the updated variant.

Anonymous Referee #1
Referee comment on "Columnar and surface urban aerosol in Moscow megacity according to measurements and simulations with COSMO-ART model" by Natalia Chubarova et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-83-RC1, 2022
The paper assesses the Moscow megacity's aerosol pollution, focusing on urban air pollution and its radiative effect. The authors employed both experimental and modeling approaches to give a comprehensive picture of the magnitude and temporal variability of urban pollution in Moscow. The paper is well written and logically organized. It has a comprehensive introduction and discusses new valuable observation results. It could be published after a minor revision according to the below comments.

General comments:

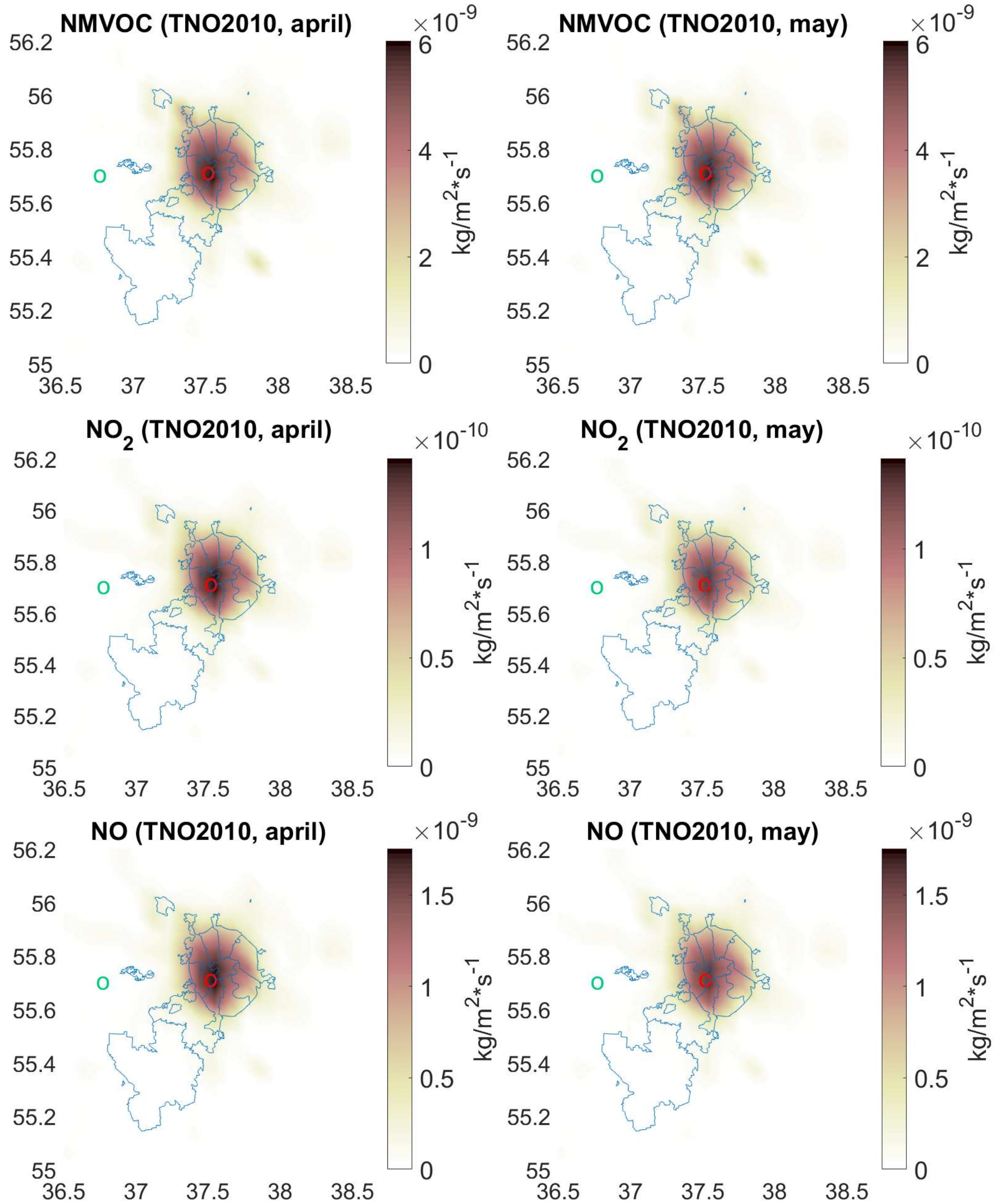
1/ The model itself and especially its aerosol component should be better explained. It would be helpful to discuss the spatial maps from the model to see how representative (spatially) the observations are.

Thank you. Yes, we added additional information about the details of aerosol evaluation in the model.

The new text is the following:

"The simulations of reactive gaseous and particulate matter are based on the enhanced KAMM/DRAIS/MADEsoot/dust model (Riemer et al., 2003a; Vogel et al., 2006, Vogel et al., 2010). In MADEsoot (Modal Aerosol Dynamics Model for Europe extended by soot) all aerosol modes are represented by lognormal distributions. Five modes for Aitken and sub-micron aerosol particles include one pure soot mode, secondary particles consisting of sulphate, ammonium, nitrate, organic compounds (SOA), as well as the modes representing aged soot particles, consisting of sulphate, ammonium, nitrate, organic compounds, water, and soot. It also includes coarse particle mode, which contains additional anthropogenic emitted particles. All aerosol fractions are subject to coagulation and condensation following Binkowski and Shankar (1995), Whitby et al. (1991), Kerminen and Wexler (1994). Schell et al. (2001). Odum et al. (1996). The soot particles are directly emitted into the atmosphere. Coagulation and condensation are accounted for in transfer of soot from external into internal mixture. The Aitken and sub-micron particles are formed due to the aging process. For each mode prognostics equations for the number density and the mass concentration are solved numerically. The standard deviations are kept constant. Since the number densities of the coarse mode are small the inter-modal coagulation between the coarse mode and the other modes and the intra-modal coagulation of the coarse mode particles are both neglected. Additionally, the aerosol distributions are modified by the sedimentation, advection and turbulent diffusion processes. More details can be found in (Vogel et al., 2010). The resuspension of urban dust with stronger winds currently is not taken into account. The chemical reactions of the gaseous species are calculated using the chemical mechanism RADMKA (Regional Acid Deposition Model Version Karlsruhe) based on RADM2 (Regional Acid Deposition Model; Stockwell et al., 1990) with the important updates described in (Vogel et al., 2010). The photolysis frequencies were simulated according to (Vogel et al., 2009). For the evaluation of the aerosol optical properties (the extinction coefficient, the single scattering albedo and the asymmetry factor) a special parameterization scheme is used based on the a priori calculations with the application of the approach described in (Bohren and Huffmann, 1983) and pre-calculated aerosol distributions. This procedure is based on typical size distributions and chemical compositions, which are simulated in the model domain."

Concerning the spatial maps: we added location of urban and pristine sites in Figure 2, where the spatial fields of emissions are shown. This clearly proves that the locations of both site are really typical for urban (MOscow MSU) and clear conditions (Zvenigorod). Please, find the updated Fig.2 in the supplemental file.



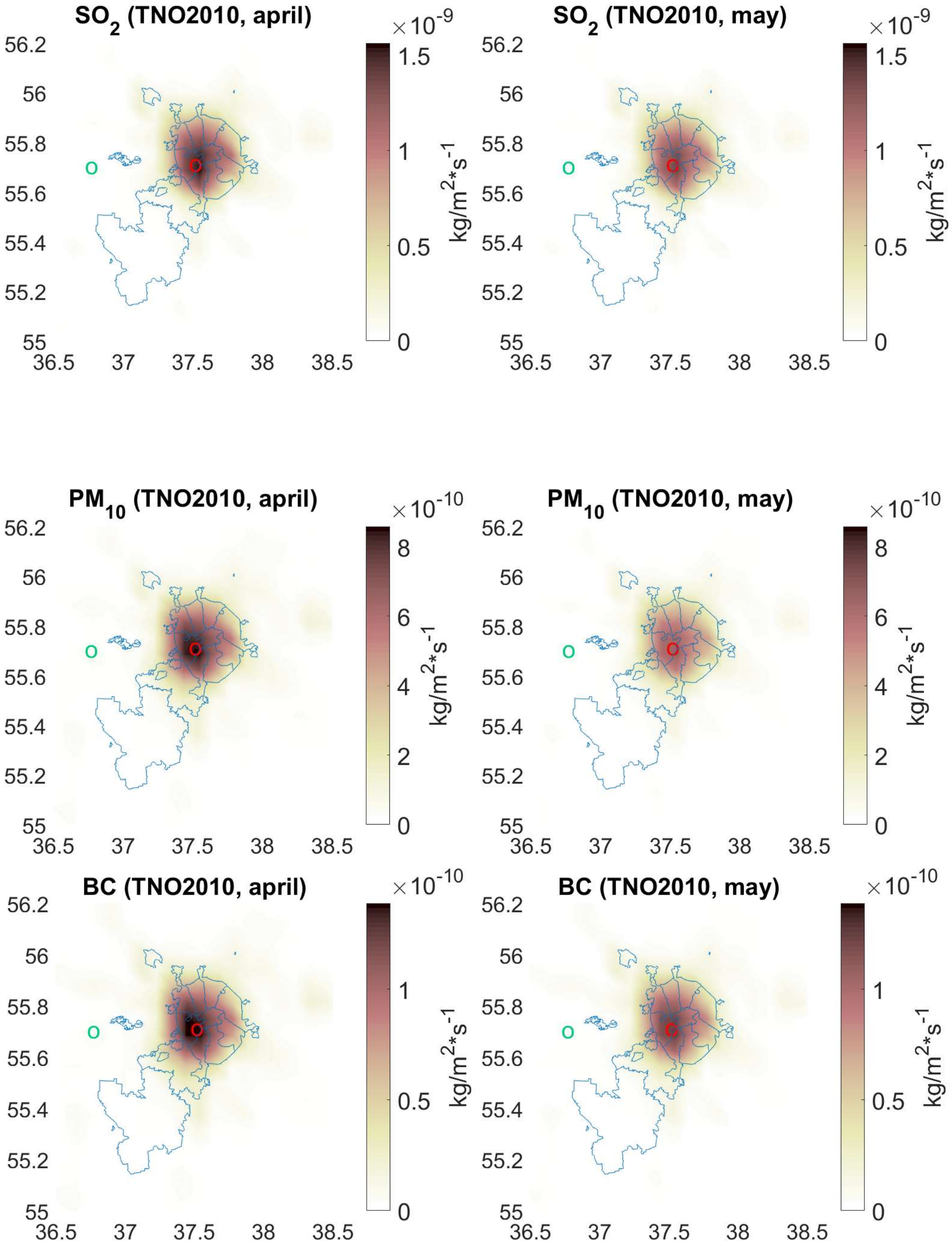


Figure 2: Monthly mean emissions of aerosol gas-precursors, PM10 and BC emissions according to the TNO2010 inventory in April and May in Moscow region. The location of the Moscow State University Meteorological Observatory is shown by red circle and Zvenigorod site – by green circle.

2/ I am not clear how does the model with 7-km grid spacing describe in-city aerosol concentration? Is any parameterization for the urban terrain used?

Yes, you are right. Local in-city variations are not represented by the model with this resolution, but the difference between the city and suburb stations provide reliable estimates of urban effects. The main aim of this study was to evaluate the features of air pollution using the difference between the data at Moscow State University Meteorological Observatory and at upwind background site at Zvenigorod (around 55 km to the west). Therefore, we do not analyze the spatial structure of pollution within the Moscow itself. In this situation it is possible to use 7 km grid. In this study no parameterization for the urban terrain is used but in our current studies with 2-km grid spacing it has been applied, along with TERRA_URB parameterization.

We added in the text the following clarification:

"Since the main objective of our study was to evaluate aerosol pollution in Moscow at the MSU MO and at upwind background conditions, we did not focus on detailed variations of aerosol inside the city and used 7 km grid step for model simulations."

3/ PM2.5 was not measured, and all information about a fine aerosol fraction came from AERONET retrieval?
It is unclear what are the natural aerosols that contribute to PM10?

Only the measurements of PM_{10} mass concentrations are currently available. However, according to Wu and Boor (2021, included in reference list) fine aerosol mode is the predominant fraction of PM10 in urban aerosol in Central and Northern Europe (see, for example, see Fig. 10 in this paper). So, we can consider that PM10 is mainly consisted of fine aerosol mode in Moscow as well, similar to columnar aerosol. Unfortunately, no information is available concerning the measurements of the properties of natural aerosol in Moscow background, except its total mass concentrations at surface. However, if take, in the first approximation, the data of CAMS reanalysis (Bozzo et al., 2020) for Moscow background conditions, we obtain that organic aerosol during warm period is mainly (more than 50%) contributed to the total amount of natural aerosol there. Sulphate aerosol is the second component in the range (20-40%), and the dust contribution lies within 0-14%.

4/ The model does not account for any natural emissions, including urban dust.
If observations are not available, it will help discuss aerosol vertical distributions, at least from the model perspective.
The model accounts for relatively small natural biogenic emissions of hydrocarbons in background region in spring (see Fig.2 for NMVOC emissions over the whole Moscow region), which are the gas precursors of organic aerosol. In addition, urban emissions include direct emission of particulate matter of undefined composition (as dust fraction) and soot, as well as the gas-aerosol precursors for sulphate, organic, and nitrate secondary aerosols. From the model perspective anthropogenic aerosol is emitted from the bottom levels of the model (up to 990 m) and decreases with height (see Figure below).

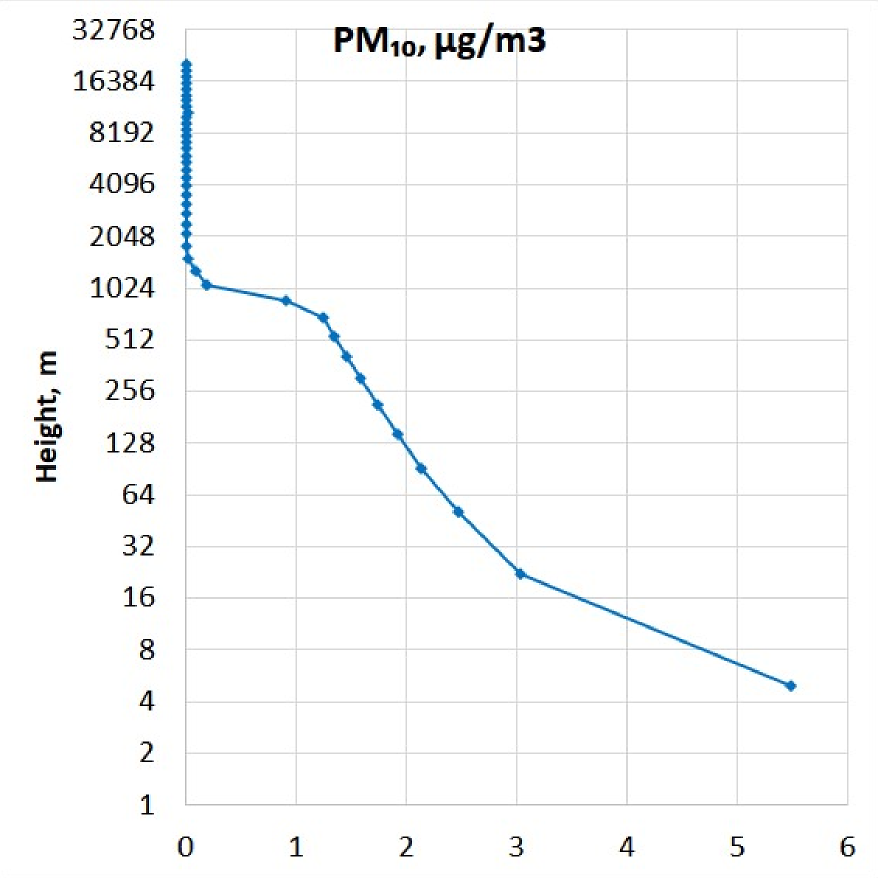


Figure 1C. This is an illustration of the PM10 vertical distribution (15/04/2018, 12UTC).

We thank you for the idea to study the vertical profiles of aerosol distribution, which we will certainly do in the separate paper. However, in this paper we focus mainly on the difference in aerosol characteristics between urban and background sites. That is why we do not account for the natural aerosol, which should be similar at these sites and will not affect the evaluation of urban aerosol component.

Some additional clarification has been added in the text:
"Urban aerosol sources according to TNO2010 include direct emission of particulate matter of undefined composition (as dust fraction) and soot, as well as the gas-aerosol precursors of sulphate, organic, and nitrate secondary aerosols. The model also accounts for relatively small natural biogenic emissions of non-methane volatile organic compounds from the Global Land Cover 2000 project, which are the gas precursors of organic aerosol."

Specific comments
L10: COSMO-ART is a regional meteorology and chemical transport model

Following (Vogel et al., 2009), where the first mentioning of the COSMO –ART was made, we specified the name of this model in the following way in this place and everywhere in the text:

"Urban aerosol pollution was analyzed over the Moscow megacity region using COSMO-ART (COSMO — COnsortium for Small-scale MOdelling, ART — Aerosols and Reactive Trace gases) online coupled mesoscale model system.."

L39: The aerosol forcing is negative
Yes, of course. We corrected the text.

"Radiative effects of the anthropogenic aerosol are negative and exceed 1 W m^{-2}…...."
L160-165: How is aerosol microphysics calculated?
We added the description of the aerosol block simulations in COSMO-ART:

"The simulations of reactive gaseous and particulate matter are based on the enhanced KAMM/DRAIS/MADEsoot/dust model (Riemer et al., 2003a; Vogel et al., 2006, Vogel et al., 2010). In MADEsoot (Modal Aerosol Dynamics Model for Europe extended by soot) all aerosol modes are represented by lognormal distributions. Five modes for Aitken and sub-micron aerosol particles include one pure soot mode, secondary particles consisting of sulphate, ammonium, nitrate, organic compounds (SOA), as well as the modes representing aged soot particles, consisting of sulphate, ammonium, nitrate, organic compounds, water, and soot. It also includes coarse particle mode, which contains additional anthropogenic emitted particles. All aerosol fractions are subject to coagulation and condensation following Binkowski and Shankar (1995), Whitby et al. (1991), Kerminen and Wexler (1994). Schell et al. (2001). Odum et al. (1996). The soot particles are directly emitted into the atmosphere. Coagulation and condensation are accounted for in transfer of soot from external into internal mixture. The Aitken and sub-micron particles are formed due to the aging process. For each mode prognostics equations for the number density and the mass concentration are solved numerically. The standard deviations are kept constant. Since the number densities of the coarse mode are small the inter-modal coagulation between the coarse mode and the other modes and the intra-modal coagulation of the coarse mode particles are both neglected. Additionally, the aerosol distributions are modified by the sedimentation, advection and turbulent diffusion processes. More details can be found in (Vogel et al., 2010). The resuspension of urban dust with stronger winds currently is not taken into account. "

L175-178: Are there any natural emissions in the model, e.g., biogenic? Do you account for the resuspension of urban dust? How significant is it?

There are biogenic emissions of hydrocarbons in the model. Figure 2 presents the distribution of all sources of aerosols gas precursors, including biogenic NMVOC, which shows the dominating urban source of these emissions.

In model simulations resuspension of urban dust with winds currently is not taken into account (urban dust is emitted as anthropogenic regardless of wind speed). It can be a source during strong winds, however, these situations are rare in spring in Moscow (mean wind velocity in spring is only V=2.6-2.8 ms-1 according to 60 years of measurements in Moscow (Chubarova et al., 2014, https://doi.org/10.3103/S106837391409005)).

We added in the description of the model the following sentence:

"The resuspension of urban dust with stronger winds currently is not taken into account."

L187: In observations, one site is urban, and the another is in the suburb. It is not much coverage. Could you tell from the model simulations that these two locations represent urban and pristine conditions?

Yes, these two locations represent urban and pristine conditions. The Figure 2 has been modified and the locations of both Moscow and Zvenigorod sites are shown now together with the fields of emissions. It is clearly seen that the maximum of anthropogenic emissions are over the MSU MO area and negligible emissions are seen over Zvenigorod site. Since we have prevailing westerlies, Zvenigorod station is not affected by the Moscow megacity anthropogenic aerosol emission, because pollutant plume from the city is transferred further to the east. MSU MO, on contrary, is affected by the anthropogenic emissions. In addition, we studied the specific conditions, when the effect of Moscow due to meteorological situations can affect Zvenigorod. The obtained differences in urban effects are not high (see the Discussion in 3.4).

In the text there is also the following information with reference to this Figure:

"Due to prevailing westerlies and location of the ZSS site far from local anthropogenic emissions (see Fig. 2) it can be characterized as a background pristine site."

The caption of Figure 2 has been modified accordingly:
"Figure 2: Monthly mean emissions of aerosol gas-precursors, PM10 and BC emissions according to the TNO2010 inventory in April and May in Moscow region. The location of the Moscow State University Meteorological Observatory is shown by red circle and Zvenigorod site – by green circle."

L190-192: It is irrelevant to have instant observations at the same time. The averaging over time would give more reliable results.

In the text it is written that "time difference between the two instant measurements in these sites is only 3 minutes." I guess this is the advantage to have the quasi simultaneous measurements of the initial data. Of course, then we averaged the data having the 1-hour resolution dataset, which has been further analyzed.

We added the following sentence in the text:

"Finally, all the data were combined in the 1-hour resolution dataset."

L225-230: The definitions of the fine and coarse aerosol fractions came from AERONET. What are coarse and fine aerosols in the model? Could you explain this in the text?
In the model there are different size ranges: Aitken mode, Accumulation mode and Coarse particle mode according to their standard definitions.
We added the information about the aerosol modes:
"Five modes for Aitken and sub-micron aerosol particles include one pure soot mode, secondary particles consisting of sulphate, ammonium, nitrate, organic compounds (SOA), as well as modes representing aged soot particles, consisting of sulphate, ammonium, nitrate, organic compounds, water, and soot. It also includes coarse particle mode, which contains additional anthropogenic emitted particles".
Please, find also the additional information about the model microphysics, which we have added above in the response.

L260-265: I suppose sulfate is low because SO2 is low. What are the secondary aerosols in your observations and simulations?
Yes, you are right, measured SO2 concentrations are much lower than those, calculated using the TNO anthropogenic emission dataset. We mentioned this in the text.
In simulations, along with other aerosol modes, we have different kinds of secondary aerosols including sulphate, nitrate and secondary organic aerosol. Their description has been added in the text. Unfortunately, currently there are no secondary aerosols measurements in Moscow. Concerning model simulations, secondary aerosols ratio in the total urban aerosol content is about 55% with maximum contribution for organic component (30%). This is mainly in accordance with the ranges provided in (Huang et al., 2014).

Huang, RJ., Zhang, Y., Bozzetti, C. *et al.* High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* **514,** 218–222 (2014). https://doi.org/10.1038/nature13774

We added this information in the text.
"This may indicate the importance of secondary aerosol generation in the urban atmosphere of Moscow. According to the model simulations, secondary aerosols are about 55% in the total urban aerosol content, which is mainly in accordance to (Huang et al., 2014), with the maximum contribution of the organic component (30%)."

L272-274: I suggest stronger winds more effectively generate coarse aerosols, like urban dust.

Yes, there is a coarse aerosol content dependence on wind velocity. However, in the model currently there is no parameterization of urban dust resuspension. We have added in the text the following remark:

"A decrease in AEE and, correspondingly, the decrease in the fine AOD fraction with the increase in wind speed may be also associated with less effective fine mode aerosol generation due to better ventilation conditions and with possible more effective urban coarse aerosol mode dust resuspension at stronger winds (Amato et al., 2009, Hosiokangas et al. 2004)."

L284-287: I would say that in unstable conditions, BC PM is more effectively dispersed vertically, the BC source is fixed, and the PM source gets stronger with stronger winds.

We have edited this sentence by adding the possible effects of resuspension. However, it should be studied further in details in special experiments. The dust fraction is not large over our region.

The changes are the following:

The smaller PM_{10} negative correlation with wind speed could be also explained by the effects of dust resuspension at stronger winds.

L308-309: Do all Moscow power stations work on gas? What about SO2 emissions from traffic?

Moscow power stations use gas as the main fuel, with other options for reserve fuels. Generally Moscow SO2 emissions are overestimated by TNO emission inventory as seen from our analysis. The Euro-5 motor fuel standard has been used in Moscow since 2016 that provides low SO2 emissions in the atmosphere.

We added this information in the updated text:

"In addition, Euro-5 motor fuel standard, which has been used in Moscow since 2016, provided low SO_2 emissions in the atmosphere."

L369: "results" > "result"

Done. Thank you.

L400: Do anthropogenic emissions have a diurnal cycle?

Yes, TNO inventory, which is used in the simulations, has a diurnal cycle with 1-hour resolution. We added this in the text.

"The one-hour resolution TNO2010 emission inventory has been developed using official reported emissions data by source category and combining them with other estimates, where needed (Kuenen et al., 2014)."