Answer to Reviewer 1

Dear Anonymous Reviewer,

We are thankful for your constructive criticism and comments to the manuscript "Estimation of elevated black carbon episode over Ukraine using Enviro-HIRLAM" submitted to the ACP journal. Please, see below our replies and changes/ modifications to the original manuscript.

R1. This is a modelling study about black carbon particles but without any validation from measurements, at least some ground measurements of BC are required.

A: We totally agree with the comments on validation and included and clarified more details in the text concerning that aspect. Unfortunately, validation data is a big and serious problem for Ukraine. There are no monitoring sites that provide black carbon (BC) measurements and this is valid for a wide variety of chemical species. Hence, the modelling remains the only available tool for the Ukrainian territory, which allowed to estimate the spatio-temporal distribution of species like BC, organic carbon, etc. As a result, there is a gap in scientific knowledge about BC in Ukraine, and there are no assessments how BC is distributed; how it influences local ecosystems, human health; and, consequently, no relevant mitigation measures were taken.

Moreover, the existing official Ukrainian air quality monitoring network (UA-AQMN) was developed and established in the former USSR, and it was not significantly improved or upgraded during the last decades. The monitoring sites (more than 120 in total) of the network do not measure PM; and dust – is the only pollutant (measured by official air quality stations) which could describe aerosol content and concentration. To note the reader on this, we added details on it in the "Methods"-section, including the definition of "dust" according to the existing local regulations in Ukraine. Of course, the process for EU Directives implementation has been already started in Ukraine, but it is still in slow progress and not finished yet.

As soon as BC measurements will be launched as part of UA-AQMN, it would be possible to study the spatio-temporal variability of BC more accurately and precisely. But nowadays only the modelling approach can be used. Nevertheless, the first assessment such as the current work, are necessary to raise awareness for interests and needs of Ukrainian national economy following UN towards Sustainable Development Goals, and in particular, SGDs 3, 9, 13.

In practice, we described and explained this in the revised version of the manuscript.

Accordingly, the following changes/additions were incorporated in the revised manuscript:
in Section "Introduction":

"Described wildfire events frequently occur in Ukraine or in territories of neighboring countries that develop into elevated pollution events. Unfortunately, the existing official Ukrainian air quality monitoring network (UA-AQMN) does not provide any measurements of BC. Hence, the modelling remains the only possible way to estimate the spatio-temporal variability of BC in Ukraine, and to explore the consequences of elevated pollution episodes and assess the short-term impacts in the region. The lack of observation capacity on BC on a national level caused a gap in knowledge on how BC is distributed; what are the impacts on local ecosystems; and which mitigation measures are needed to improve the situation. This study is the first in Ukraine which describes the elevated spatio-temporal BC content on the three-dimensional scale, explores the distribution of BC in particle sizes and compares the BC ratio between other aerosol components in the atmosphere with the emphasize on extremely hot weather episodes. Furthermore, we aimed to estimate the impact of wildfire

emissions on the surface by considering direct aerosol effects, as the response of radiative and temperature regimes varying in different regions and depending on the ratio of aerosol components."

& in sub-section 2.2 "Additional data for analysis"

"Unfortunately, UA-AQMN does not provide measurements of BC, moreover PM also is not measured. Dust – is the only available pollutant which could be compared to the modeled aerosol species. The dust is measured at more than120 monitoring sites, and measurements contain all coarse aerosol particles regardless of their origin (Nadtochii et al., 2019). Dust concentration on these sites is measured using the method of weighing the total suspended particles (RD, 1991).

R1. The modelling setup is too rough.

A: The section 2.1 "Enviro-HIRLAM setup" was updated with including more details. The following text was added for meteorological and atmospheric composition input for the model runs:

"The initial and boundary conditions (ICs/BCs) for meteorology include components of winds speed, air temperature and specific humidity extracted at all model levels and at every 3-hour interval from ERA5 model archives at the European Centre for Medium-Range Weather Forecasts (ECMWF). The sea surface temperature and the conventional BUFR (The Binary Universal Form for the Representation of meteorological data) observations (for data assimilation) were extracted at every 12-hour and 3-hour interval, respectively. The ICs/BCs for gas and aerosol concentrations included 3D fields of mixing ratio of aerosol components (dust, hydrophilic and hydrophobic organic matter and black carbon, sulphates) as well as gases (O_3, SO_2, NO_2, NO, hydrogen peroxide (H_2O_2), hydroxyl radical (OH), nitrate radical (NO_3), hydroperoxyl radical (HO_2), dimethyl sulfide (DMS)) were extracted at all model levels and at every 3-hour interval from Copernicus Atmosphere Monitoring Service (CAMS) of ECWMF."

The following text was added for emission inventories used for the model runs:

"A suite of emission inventories (EIs) was utilized in the model runs, including anthropogenic and biomass burning (wildfires). In more detail, the EIs (given in geographical latitude/longitude domain) – for the global biomass burning (wildfires) emissions (IS4FIRES; Sofiev et al., 2012), Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants (IIASA's ECLIPSE, 5th version) (gridded emissions of gases and aerosols of SO_2, NO_x, NH_3, nmVOC, BC, OC, $PM_{2.5}$, PM_{10}, CO, CH_4 for SNAP codes (Selected Nomenclature for Air Pollution) such as industry, transportation, agriculture, etc.), shipping emissions (include SO_2, BC and OC), emissions of DMS (Nightingale et al., 2000). The mission preprocessor includes both vertical and temporal profiles for the model setup."

R1. Not just that, but all the presentation throughout the texts shows the authors have not really performed any meaningful data analysis.

A: We expand our analyses with results by adding the analyses of aerosol component ratio, direct aerosol effects on downwelling surface radiation fluxes and air temperature. There was added Aitken mode for previously presented results. Based on this work, the section "Results and Discussions" was revised.

The section 3 "Results and Discussion" was updated with including more details.

In particular, new sub-section "3.3 Ratio of aerosol compounds and observed direct aerosol effects during the wildfire episode" was included with Figures 5-6-7.

"Aerosol impacts on the atmospheric composition during wildfire events depend on the intensity of the fire (Vadrevu et al., 2015), on the fuel (Christian et al., 2003; Lee et al., 2018) and the stage of the fire (e.g., open fire, smoldering fire (Lee et al., 2010; Popovicheva et al., 2019)). These features will influence for example the ratio of CO/CO_2, amount of black carbon emissions as well as the vertical distribution of BC. Enviro-HIRLAM simulations were conducted considering five main aerosol compounds: BC, organic carbon, sulfate, sea salt and mineral dust. The highest values of BC-to-total aerosol content ratio observed in the lower 200-m layer (up to 990 hPa) over fire areas reaching 20% (see Fig.5a). BC normally accounts for 10% at the distance from wildfires by the prevailing wind (Fig. 5d-5i). On the example of Odesa (the most distant city among presented), it is well seen the influence of local urban BC emissions, where BC ratio increased to 15-24% near the surface (see Fig. 5j-5l). Vertical distribution of BC ratio was rather uniform from the surface to the altitude of 2 km (≈800 hPa).

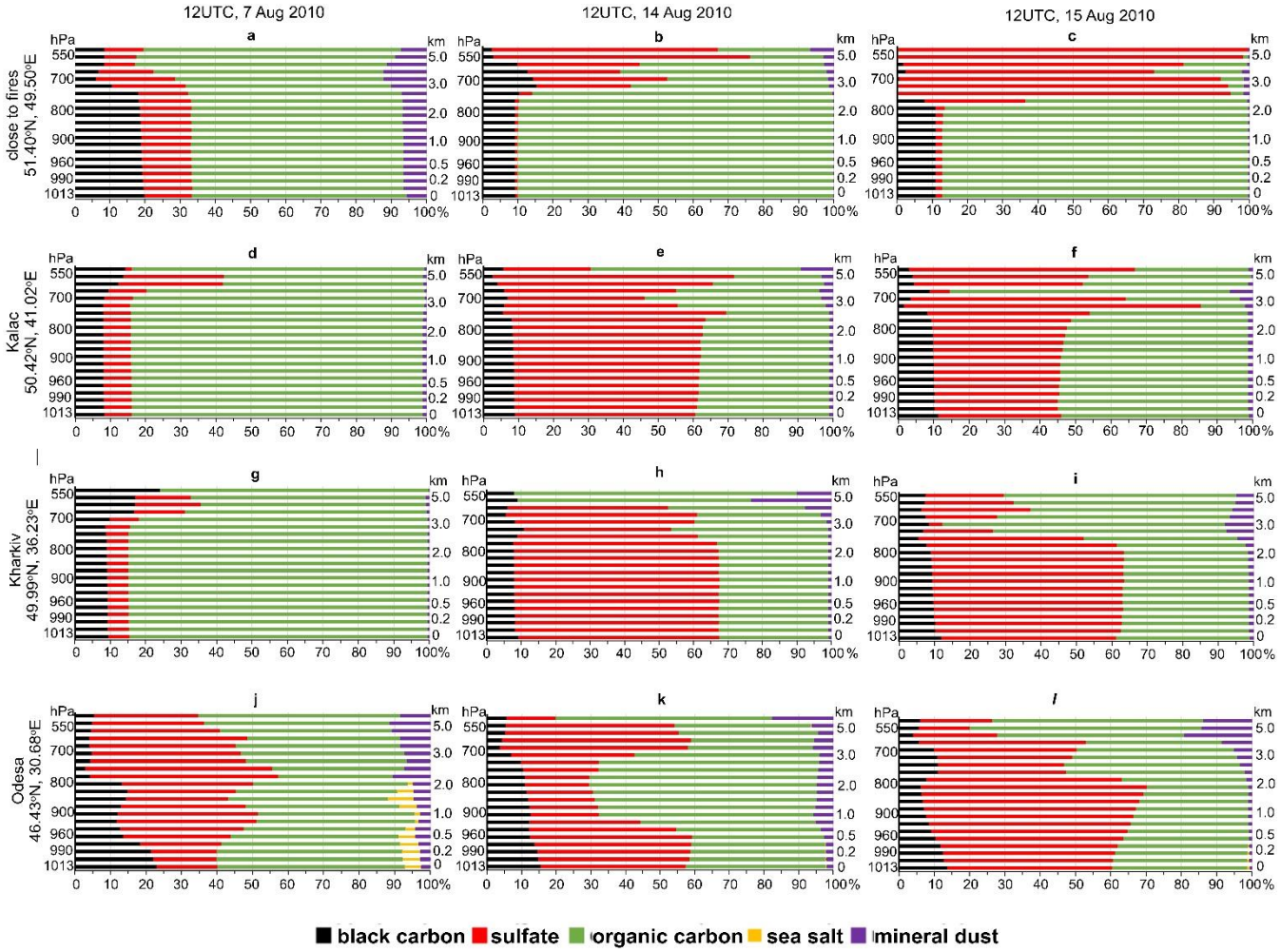

Figure 5: Vertical distribution of ratio of aerosol compounds in the days of air movement towards Ukraine at 12UTC on 7th August (a,d,g,j), 14th August (b,e,h,k) and 15th August (c,f,I,l).

Organic carbon very often is the main aerosol component in the lower 2-5-km layer (≈800-550 hPa), especially near the fires, but also at the surface in general (e.g., Jimenez et al. 2009). Based on our results, sulfates prevail throughout the lower and middle troposphere at the distances, and over 3 km (≈700 hPa) level near the wildfires. Hence, aerosol effects over the region were mainly driven by organic carbon and sulfates aerosols. The largest BC contribution during anticyclonic weather conditions seem to be in the boundary layer. In our analysis, mineral dust ratio rarely exceeded 10%, whereas sea salt was detected in the atmosphere only near the sea (see Fig. 5j, 5l).

Based our modeling results, continuous wildfires and aerosol emissions over the region as a consequence influenced the radiative transfer via direct aerosol effects. Enviro-HIRLAM DAE run showed both increase and decrease of downwelling surface long-wave and short-wave radiation. The most intense changes caused by direct aerosol effects were observed during the midday reaching over ±20 W/m^2 for long-wave and over ±100 W/m^2 for short-wave radiation (see example on Fig. 6). The observed direct effects on the downwelling surface radiation co-aligned with the total aerosol impact. Our results indicate that when the BC concentration and the coarse mode exceeded 30 ppb and BC-to-total aerosol mass ratio was over 10% in the lower troposphere, the downwelling surface long-wave radiation tended to increase while the changes of short-wave radiation varied within -200…25 W/m^2.

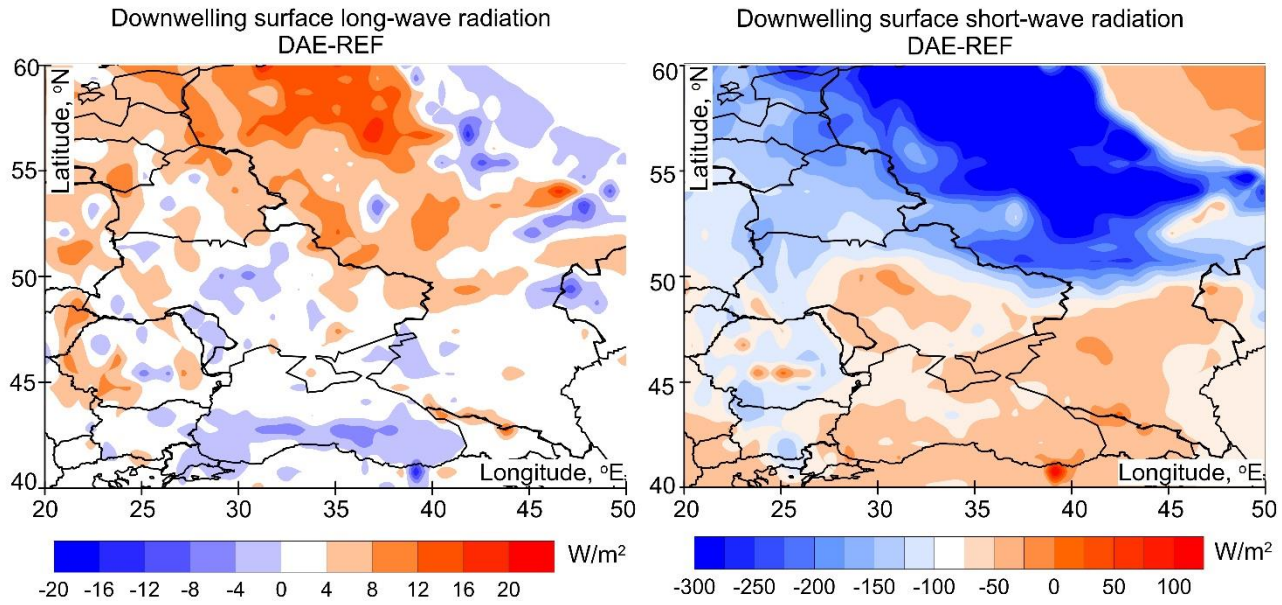

Figure 6: Differences between DAE and REF for downwelling surface long-wave and short-wave radiation at 12UTC on 14th August 2010.

In our simulations, direct aerosol effects had an impact on temperature as well. The difference between DAE and REF runs showed that 2-m air temperature during the midday varied from -3°C to 3°C (see Fig. 7). The 2-m air temperature decreased due to the direct aerosol effects prevailed during the wildfire episode. However, the 2-m air temperature was higher with direct aerosol effects in areas where DAE downwelling surface long-wave radiation was higher 12 W/m^2 and DAE-REF difference for surface short-wave radiation was not lover than -150 W/m^2. Moreover, the 2-m air temperature was 1-4°C higher at the distance from the wildfires over the territories where the BC coarse mode exceeded mixing ratios of 20-30 ppb.

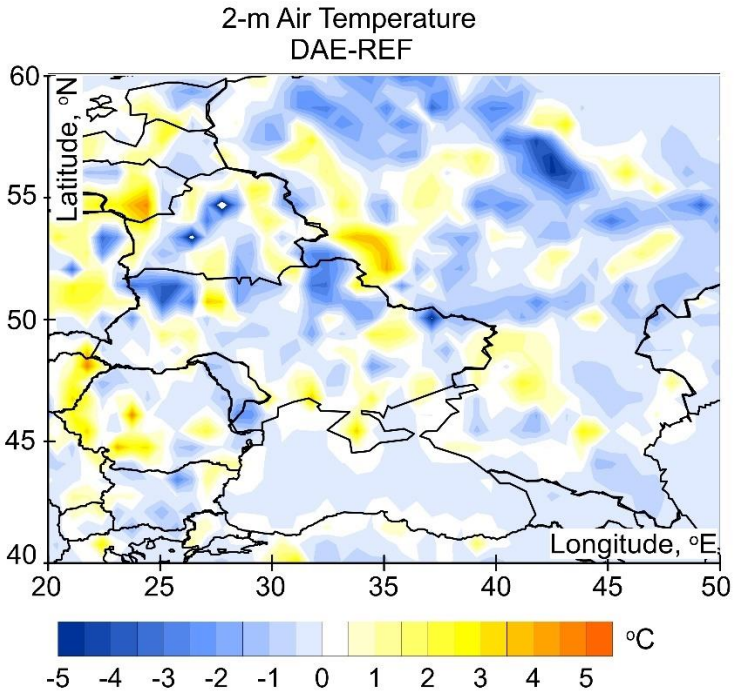

Figure 7: Differences between DAE and REF for 2-m Air Temperature at 12UTC on 14th August 2010.

Moreover, the sections 3.1 and 3.2 were also improved by addition more detailed analysis for the Aitken mode (including updated Figures 2-3-4), and, in particular, by adding the following text:

for sub-section 3.1:

"At the beginning of August 2010 before the wildfire episode, the BC content over Ukraine was low and mostly it was represented by the accumulation mode with the values of 1-8 ppb. The highest BC concentrations reached 16 ppb (accumulation mode) and was observed at the near-surface level near small local wildfires in the central part of Ukraine. The BC concentrations in the Aitken and coarse modes did not exceed 0.5 ppb."

&

"In general, the wildfire emissions have a large accumulative effect in the near-surface layer. Total accumulated amount of BC for the period 3–18 August 2010 reached 13500 ppb (for the accumulation mode) and 2200 ppb (for the coarse mode) in the lower tropospheric layer near the burning areas (Fig. 3). The total accumulated amount of BC Aitken mode caused by wildfires was about 15-30 ppb, whereas the maximum accumulated impacts were observed in the cities (see e.g., Aitken mode on Fig. 3). A large amount of combustion products was transported through the atmosphere to the south-west and deposited over the territories of the Eastern Ukraine, the Azov and Black Seas. The integral values of BC on these territories exceeded values of 800 and 150 ppb for the accumulation and for the coarse modes, respectively. This is well seen in Fig. 3 where the regions were affected by intensive deposition processes. Due to the smaller sizes of the particles, the accumulation mode had a larger spatial extent and more smooth distribution than the coarse mode."

&

"For the studied period, in general, the ground-based dust measurements also showed elevated levels connected with the forest fires. Almost all the geographical regions except northern part of Ukraine experienced a clear maxima of ground-based dust concentrations during August 2010. The highest exceeding over the urban background values in August were observed on the eastern territories. In this case, the integral value of the near-surface BC concentrations for the coarse mode was higher than 250 ppb. For the cities on the east, the dust concentrations were 27–47% higher than the average

dust content in 2010. Moreover, these values were also 23–72% higher than multi-year average concentrations for the month of August. Overall, the dust content on the east Ukraine was 0.02–0.07 $\mu g/m^3$, which was higher than usually occurred in August.

Large difference in the dust content between August and other months was observed on the seashore of the Azov Sea and in the central part of Ukraine. The concentrations were higher by 0.05–0.25 $\mu g/m^3$ than usually in the same month. A majority of cities in the central part of Ukraine showed 17–73% higher dust concentrations than average in 2010 and concurrently 8–45% higher than usually in August. In the western parts of Ukraine, the integral values were lower than 100 and 500 ppb for coarse and accumulation modes, respectively (Fig. 3)."

for sub-section 3.2:

"In contrast to the BC coarse and accumulation modes, Aitken mode was observed throughout the entire troposphere up to 200 hPa level (Fig. 4) height, however the wildfire emissions did not prevail over the anthropogenic emissions. The BC Aitken mode rarely exceeded 0.2 ppb at 950 hPa level. Therefore, concentrations more than 1 ppb were observed only near the surface and had clear maximum over urban areas (see Fig. 3). This is the reason why the BC Aitken mode near the surface was higher in more distant Odesa than in Kalac region."

& sub-section 3.4 "Discussion" was added:

"The BC dispersion and vertical distribution during stationary anticyclonic conditions had their own specific features. The most typical cases were a uniform vertical profile up to 700 hPa (≈3 km) with a BC ratio of 8-20% among all the aerosol compounds. Very often there was no clear maximum within the lowest 3-km layer especially for the coarse and accumulation modes. The urban areas with additional anthropogenic emissions were an exception where a maximum formed near the surface. This distribution differed from the other synoptic conditions, which resulted in a maximum BC concentration outside the emission areas at an altitude of 1.5 km (Kostrykin et al., 2021).

The decrease of radiation forcing (Chubarova et al., 2012) and a reduction of shortwave radiation (Pere et al, 2014) were observed after wildfire aerosol emissions during summer 2010. Previously it was shown that the shortwave radiation is smaller on 70-84 W/m^2 in the diurnal averages (Pere et al., 2014). Our results confirmed the decrease in the downwelling surface short-wave radiation at the background as the result of the direct aerosol effects were up to -250 W/m^2 during the midday hours. However, numerous spots were observed, where the downwelling surface short-wave and long-wave radiation increased. The 2-m air temperature was higher in spite of considering the direct aerosol effects of the areas where the long-wave radiation increased up to 20 W/m^2 and both BC coarse and accumulation mode exceeded values of 30 ppb. These hot-spots might coaligned with the BC impacts at certain locations, as the BC influence is known by its localizing effects with no clear directs link between the pattern of forcing and pattern of temperature change (Modak and Bala, 2019). Overall, the direct radiative forcing and the consequent air temperature increase was estimated for BC regardless of the emission source (Ma et al., 2018; Zhuang et al., 2019; Kostrykin et al., 2021); however cooling effects also was detected (Ma et al., 2018). In combination with other aerosol types the effects on the radiative forcing also is variable (Kirkevag et. al, 1999). Hence, not all areas with air temperature increasing or decreasing are not directly connected with the prevailing influence of certain aerosol compound but a net effect including physical, chemical and meteorological effects are being experienced.

In the absence of BC measurements, ground-based monitoring network in Ukraine represents a challenge for the results validation. Nevertheless, it gives an overall picture of the BC distribution during unfavorable weather conditions and makes it possible for a provision of assessments in Ukraine on the quantify of BC influenced human health and local ecosystems in Ukraine as a whole.

Moreover, the fact that the wildfire emissions can cause local the air temperature to increase amid elevated pollution levels is crucial for the understanding the consequences for vulnerable people. This is especially relevant during the heat wave events such as in August 2010, which is discussed in this study.
“

R1. Is the unit of BC concentration in ppbm? What is ppbm by the way.

A: The units are in ppb, corrected throughout the text of the manuscript and Figures.

R1. There are a number of places using quotation marks when referencing. You need to extract the key relevant information and write down in your own way rather than using the original words and quotation marks.

A: We put quotations just for indication the places in the text with references. We do not use the original words or expressions. It is our own analysis of literature/ references/ sources. We excluded all unnecessary quotations. Moreover, the literature analysis was also improved.

In particular, in section “Introduction” the following was added:

“In contrast to other the aerosol compounds, BC typically cause radiative a positive forcing (Bond et al., 2013; Stjern et al., 2017) which intensity depends on the particle size (Matsui et al., 2018). Consequently, the heating effect is generally observed from wildfire emissions (Kostrykin et al., 2021), and also from anthropogenic sources (Zhuang et al., 2019). Sometimes a cooling effect was also detected (Ma et al., 2018). All these effects, however, are localized (Modak and Bala, 2019).”
&

To support this improvement, additional references were included in sub-section 3.4 “Discussion”.

R1. The quality of figures needs large improvement.

A: All figures in the manuscript were improved by re-drawing with a higher resolution and clarity included a re-written caption-text to the figures in a more self-explanatory way. Some of the figures (in particular, synoptic map from www.wetterzentrale.de & figure on air temperature vertical profiles from the vertical sounding stations Kalac and Kharkiv) were moved to Appendix A. Also, we added new material in sections in the “Results and Discussion” which was supported by new illustrations with corresponding analysis.

R1. What is the size fraction of PM10, did you use the mass concentration of PM10 or PM10 minus PM2.5. The measurement needs to be explicitly given. Also how you have defined the PM10 is necessarily dust.

A: We have corrected and clarified text in the “Methods”-section. The material and explanation about the dust measurements on UA-AQMN stations was also added. We clarified that dust includes all the aerosol particles regardless their origin with the size not less than PM_{10} fraction.

R1. For modelling, a description of anthropogenic emission inventory you used is needed.

A: Sub-section 2.1 for the Enviro-HIRLAM model setup includes now also a description of all emission inventories utilized in this study for the model runs.

R1. Fig. 7 is simply part of the open biomass burning emission inventory used (the burn area fraction), not footprint.

A: After a re-evaluation of the "Result and Discussion"-section, we excluded/ omitted the Fig. 7 (in old numeration) and provided an explanation about the possible influence of local fires.

R1. Overall, this manuscript is not qualified for commenting in details.

A: Following your constructive remarks and comments above, the original manuscript was revised, with new sections (3.3. and 3.4) that were added, illustrations/figure that were re-drawn with high resolution and for clarity. In general, we added more relevant material and improved on the detailed analysis, as requested by the reviewer.