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 manuscript.

R1. It is appreciated that the authors have made great efforts to revise the previous manuscript. There is some more information added and the manuscript is improved. However, there are still large room for further improvement for its consideration in publishing in ACP. I only list a few points which may help, but the overall writing still needs considerable improvement.

A. Following your comments/ suggestions, the manuscript was further improved. Please, see detailed replies below.

R1. Why the unit of BC is ppb? Why not give mass concentration, are you modelling the number concentration of BC? I only see the unit of aerosol concentration is mass per unit volume (or mass) of air.

A. The Enviro-HIRLAM output of aerosol compounds is given in ppb. For clarification, as the obtained modelling results are the only available data of BC in Ukraine, we used the ppb units for BC, and to avoid confusion, we converted ground-level measurements of dust to ppb as well in order to unify our comparison.

R1. The map in Fig. 1 could be colored with terrain height for a better demonstration.

A. Fig. 1 was updated by including terrain relevant data to underline presence of the mountainous areas.

R1. The reason in selecting the three sites should be more explicitly given. The three targeting places should be also marked in the maps in Fig. 2-4. The reason in selecting these three sites needs to be explained by careful analysis of synoptic meteorology.

A. We clarified in Section 2.2 as (Lines 158-165):

"The air temperature vertical profiles were analyzed for 2–18 August 2010 at the following sounding stations: Kyiv (station code 33345), Kharkiv (34300), Odesa (33837), Rostov-na-Donu (34731), Kalac (34247) and Voronezh (34122). Unfortunately, there is a limited number of the sounding stations in the region of interest. Hence, Voronezh and Kalac were selected because these are situated within a radius of less than 400 km away from the forest fires area (being the nearest sounding stations to wildfires). Kharkiv and Rostov-na-Donu were selected for estimation of temperature inversion impact at some distance (within a radius of up to 600 km) away from the fires. Kyiv and Odesa were chosen as these are relatively distant (within a radius of up to 1000 km) to wildfires emission sources. The air temperature vertical profiles were considered for up to 3.5 km above the ground surface at 00 UTC."

Overall, we analyzed BC distribution in all grid cells over the region of interest (i.e. not only for these selected sites). The analysis and explanation in the section "Results" refer to all region of interest. In the same way, for Fig. 2 and Fig. 3 – analysis was made for the entire geographical territory, not for the specific locations. These sites are the sounding stations, and we used these locations in "Results" while describing vertical profiles in Fig. 5 considering in-situ aerological measurements. Air temperature profiles were analyzed for these 5 stations, but on plots we showed three of them as an example (one station depending on the distance).

The description of synoptic situation was amended as follows:

"…a blocking anticyclone caused severe hot weather, lack of precipitation, which lead to the occurrence of wildfires. The period lasted from the end of June to the second half of August 2010 over Eastern Europe and the south-western regions of Russia (see example Fig. A1). Hot air masses from Central Asia penetrated the territories on the north-west, and the anticyclone was detected throughout the whole troposphere before the highest pollution levels were transported and distributed out of burning areas. Continuous extreme weather and clear sky conditions together with the highest insolation in the middle latitudes caused the dominance of high temperature and low humidity regimes. These led to the most favourable conditions for drought formation that played a crucial role in initiating the wildfires and contributed to their rapid spread." (Lines 169-175)

"The observed anticyclonic conditions influenced the formation and development of spatio-temporal patterns for BC atmospheric transport and dispersion. The time-series for each grid point consisted of two maxima. These were related to the observed atmospheric circulation patterns. A typical clockwise air movement for anticyclones in the Northern Hemisphere caused an intensive atmospheric transport towards Ukraine during two periods: 7-8 and 13-16 August 2010." (Lines 183-186)

"The dominated hot and dry weather conditions interrupted in the second half of August 2010. It occurred when the blocking anticyclone weakened, and a cyclone arrived from the western sector." (Lines 204-205)

R1. The tile should be revised, what "BC episode" mean?

A. From definition, the using of word "episode" is attributed to an event or a group of events occurring as part of a sequence. The wording as "air pollution episode" is frequently used as well. BC is one of air pollutants, and we are using the wording "BC episode", when we discuss the episodic elevated BC concentration events in Ukraine.

To clarify, we suggest to edit the current title – "Estimation of elevated black carbon episode over Ukraine using Enviro-HIRLAM" to "Enviro-HIRLAM model estimates of elevated black carbon pollution over Ukraine resulted from forest fires".

R1. It is not clear throughout the text whether the presented BC concentration is columnar concentration (layer-integrated) or surface concentration. In Fig. 3, what is the integral near-surface?

A. BC concentration was modelled at 40 model levels. These details are given in Section 2.1 (Lines 116-117):

"The vertical structure included 40 model levels (up to 10 hPa) with a more detailed resolution in the boundary layer with 22 model levels from the surface up to 500 hPa. This provided a great opportunity to study the BC vertical atmospheric transport."

We analyzed BC concentration both for near-surface level and at the heights/ altitudes, but we did not analyze integrated columnar content.

For example, caption to Fig. 2 contains: "…with the highest BC content near the surface", to Fig. 3 "The integral value of the near-surface BC concentrations…". We identified pressure levels in the text when we analyzed BC content at the heights.

In the revised version, we have clarified throughout the text where it was not accurately mentioned.

The explanation of integral near-surface content is given in the Section 2.1 (Lines 139-143). It was clarified as the following:

"The spatial analysis of model output was carried out considering all grid cells without spatial averaging and interpolation. It enabled the detection of concentration changes within each grid caused by anticyclonic air movements. Evaluation of accumulated BC impact was performed by time integration of near-surface BC concentration at the lowest (near-surface) model level for the studied period. Therefore, the summed values represent the total amount (for the Aitken, accumulation, and coarse modes in ppb) transported by air movements through grid cells near the surface."

There are also more details in the Section 3.1 (Lines 207-215):
"In general, wildfire emissions had a large cumulative effect in the near-surface layer concentrations. The 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, respectively, 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 in Fig. 3). A large amount of combustion products was transported through the atmosphere to the south-west and deposited over territories of the Eastern Ukraine, the Azov and Black Seas. The integral values of BC for these territories exceeded values of 800 and 150 ppb for the accumulation and for the coarse modes, respectively. This is 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 a smoother distribution than the coarse mode."

R1. What is the difference between Fig. 2 and Fig. 3. What do you mean by "during the episode when highest surface concentration"?
A. We corrected caption to Fig. 2 as: "Figure 2: The spatial distribution of the BC for the Aitken (a,d,g), accumulation (b,e,h) and coarse (c,f,i) modes in the days of air movement towards Ukraine with the highest BC content near the surface."

Fig. 2 shows BC content at specific time and day with the highest BC content near the surface (when air masses moved towards Ukraine). Fig. 3 shows BC content near the surface integrated with time (it was done to estimate accumulative effect in the near-surface layer which has the largest impact for human health).

R1. It is unnecessary to show the sub-figures in Fig. 4, you may just use a log x-axis. The x-axis needs to be consistent among figures to be comparable.
A. The Fig. 4 was corrected.

R1. There are a lot of information which has not been conveyed clearly in Fig. 4. How the accumulation mode BC had changed so dramatically? This needs to be presented in a map along with synoptic analysis and fire points to show 1) how BC has been transported 2) how the particle size had evolved. The key question needs to be answered: how the size of BC and mass loadings has been changed. I still don't get the reason why BC could be so large for biomass burning, are those mixed with dust?
A. After we added new Fig. 4 and revised the text, the Sections 3.1 and 3.2 together with Figures 2-5 respond these questions:
"The main source of wildfire emissions was located outside of the Ukraine's territory and consisted of several burning areas (see Fig. 2b). The observed anticyclonic conditions influenced the formation and development of spatio-temporal patterns for BC atmospheric transport and dispersion. The time-series for each grid point consisted of two maxima. These were related to the observed atmospheric circulation patterns. A typical clockwise air movement for anticyclones in the Northern Hemisphere caused an intensive atmospheric transport towards Ukraine during two periods: 7-8 and 13-16 August 2010. During these periods, the elevated concentrations were also observed in the northern regions of Ukraine (as shown in Fig. 2)." (Lines 183-187)

"For these days, the highest BC values for the accumulation mode near the surface exceeded values of 400-600 ppb near the burning areas and values of 70-150 ppb over the selected territories of the eastern, central, and northern parts of Ukraine. For the coarse mode, the corresponding values were 300-450 ppb and 20-60 ppb for the same territories, respectively. The coarse mode BC mainly deposited close to the burning areas and only a small fraction was transported more than 500 km away from them (Fig. 2). In contrast, the accumulation mode BC was transported far from the burning areas by the prevailing wind, and it was the dominant BC mode at a distance of ca. 1000 km (Fig. 2). The highest values for Aitken mode BC did not exceed 6 ppb near the fires, and the simulated values were lower than 2 ppb at a larger distance from the burning areas. Moreover, the highest values of 10 ppb of the Aitken mode BC were intermittently observed over urban areas. Due to the geographical location of the wildfires, maxima in the BC concentration time-series shifted from the east and north-east to the west and south-west regions of Ukraine. Such a shifting depended on anticyclonic air mass movements. Nevertheless, during 7–8 and 13–16 August, the higher values in the western part of Ukraine were observed within 24 h after the maxima emerged in the east. During unfavourable circulation conditions, the pollution plumes were transported and dispersed at distances of more than 2000 km away from the original wildfire areas. The dominated hot and dry weather conditions interrupted in the second half of August 2010. It occurred when the blocking anticyclone weakened, and a cyclone arrived from the western sector." (Lines 192-205)

"During 3-18 August at nighttime, the whole European territory of Russia, and the central and eastern territories of Ukraine were characterized by a presence of surface air temperature inversions. This was indicated by diurnal variation of BC in the lowest 500 m layer. The deepest and strongest inversions, up to 655 m depth and up to 12.5°C, were observed during 4–7 August and on 16 August (see Fig. A2). At 500 m level, the air temperature was warmer by 10–12°C than at 2 m height. On the other nights, the inversions were weaker with an average difference of only 3–4°C in the 2-500 m layer.

In our study, the diurnal variations of the BC content in the boundary layer were well identified. Maxima was often observed at nighttime and during morning hours, whereas the daytime pollution levels were low in the lower troposphere (see Fig. 4). These diurnal variations occurred due to radiative surface cooling during summer nights, and especially, in the case of blocking anticyclones. Such anticyclones facilitated the formation of air temperature surface inversions. Together with the intensification of downward air movement at night, BC from the whole lower and middle tropospheric levels is accumulated within the boundary layer, reaching elevated values there. During the daytime, the processes of less intensive air descending and turbulence increasing resulted in a more homogeneous vertical distribution. These processes led to a decrease in near-surface BC content. As it happened throughout the Ukraine's territory with anticyclonic weather, spatial distribution of BC at the separate level in the lower troposphere was dominated by horizontal dispersion.

The vertical distribution of the coarse BC mode and accumulation modes was well detected in the lowest 3-km layer with the maximum observed in the boundary layer. Approximately at the 700 hPa level, BC concentration for both accumulation and coarse modes started to decrease very rapidly (Fig. 5). The levels of 660-630 hPa for the coarse mode and 590-450 hPa for the accumulation mode were identified as the highest altitudes where the influence of wildfire emissions was detected constantly during the daytime. The elevated BC concentrations were rarely detected at 590 hPa (for the coarse mode) and at 400 hPa (for the accumulation mode) levels, where the concentrations remained below 0.1 ppb. In other words, in the middle troposphere, diurnal variations of BC content were negligible because surface inversions did not reach these altitudes.

In contrast to the coarse and accumulation modes, the Aitken mode was observed throughout the entire troposphere (Fig. 5), however, the wildfire emissions of the Aitken mode did not dominate over the anthropogenic emissions. The Aitken mode rarely exceeded 0.2 ppb at the 950 hPa level. Therefore, concentrations of more than 1 ppb were observed only near the surface and had a clear maximum over urban areas (see Fig. 3). This is the reason why the Aitken mode near the surface was higher at more distant Odesa than Kalac." (Lines 237-270)

"As seen in Fig. 5, BC was rather equally distributed in the 1000–700 hPa layer during daytime hours. But at nighttime, the BC was observed mostly in the boundary layer, and especially, in the coarse mode. The air temperature inversions and descending air during the night hours resulted in more intense deposition of coarse mode, and hence, making it difficult to detect coarse particles at the distances of more than 1000 km away from the active fires (represented by Odesa in Fig. 5). However, the accumulation mode could be transported through the atmosphere over such long distances and such transport was observed within the lowest 3-km layer.
Overall, it was found that the accumulation mode BC was a predominant size fraction in the region of study. While concentrations of the BC Aitken mode were small and the coarse mode deposited mainly close to the wildfires, the accumulation mode was transported by prevailing winds far from the burning areas, and mainly within the lowest 3-km layer (up to 700 hPa). In particular, this size fraction caused elevated BC concentrations over the region of study, with the highest concentrations near the surface at nighttime and during morning hours." (Lines 278-288):

R1. Fig. 5 needs to be improved. It is not nicely shown, and I don't see the point to show this information.
A. The Fig. 5 was improved (Note, it is the Fig. 6 in the revised version).
The vertical distribution of BC and the ratio of other aerosol compounds at different distances from wildfires is important to analyze before the consideration of direct aerosol effects during the study period. It helps us to link observed direct aerosol effects in the atmosphere to aerosol content, and BC, in particular.

R1. There are still a lot of work to do for discussions, especially for the atmospheric processes. The current discussions are not sufficiently supported by the current analysis.
A. We improved the Discussion by adding the following more elaborated text (Lines 340-362):
"Wildfires typically produce large amount of BC emissions, which can be transported through the atmosphere far from the source (Eleftheriadis et al., 2009; Bond et al., 2013). During summer 2010, one of the most severe wildfires in Europe occurred, enhanced by an unusually strong heat wave (Konovalov et al., 2011; Witte et al., 2011; Galytska et al., 2018). During observed stationary anticyclonic conditions, atmospheric horizontal and vertical transport of BC illustrated specific features. The territory of Ukraine, being among the most affected geographical regions (Galytska et al., 2016; Galytska et al., 2018), has faced several elevated pollution episodes. The existing problems with national UA-AQMN made it impossible to perform an accurate analysis of different aerosol compounds, including BC distribution with Enviro-HIRLAM. Our results showed that the anticyclonic conditions caused the atmospheric transport of BC for more than 2000 km away from the burning areas. This elevated BC content was mainly formed by the accumulation mode, which is different to e.g., dust events (Kompalli et al., 2014), where coarse particles are the predominant size fraction. The Aitken mode contribution was much smaller in comparison to the accumulation and coarse modes, and urban emissions frequently defined the spatial distribution of the Aitken mode BC. At the same time, the coarse mode BC was deposited not far from the burning areas, and it was rarely observed at distances of ca. 500 km away from the fires. Overall, during these particular wildfires, the area affected by intense BC deposition was rather large in comparison to other wildfires (Hodzic et al., 2007).
Based on our results, BC was mostly transported within the lowest 3-km layer (up to 700 hPa). At these altitudes, the BC ratio was 8-20% among all aerosol compounds. Frequent and strong air temperature inversions determined deposition during August 2010. Its influence during the stationary anticyclone was compared to that which low-level temperature inversions impact in areas with complex terrain on the example of Slovenia (Glojek et al., 2022). These conditions defined diurnal variations of BC and its vertical profiles. Despite the fact that the wildfires had the dominant role in

the region, BC behaviour on a diurnal cycle did not differ much in comparison to urban emissions (Sahu et al., 2011; Chen et al., 2014). The BC maxima near the surface occurred at nighttime and during morning hours, and minima were dependent on the boundary layer height development during the afternoon hours."

R1. In conclusion, you have not really shown any diurnal variation.
A. In the revised version we added Fig. 4 with BC diurnal variability.

R1. The last two paragraphs of conclusion are loosely written.
A. Corrected as (Lines 416-426):
"The findings of this study are important and valuable for improving numerical weather prediction due to taking into account aerosol effects and for depicting the impacts of extreme events such as wildfires with atmospheric chemical transport modelling. The results are relevant for multi-scale assessment studies of atmospheric pollutants' impacts on population and ecosystem health, for climate adaptation and socio-economical related studies, for optimization and establishing of air quality monitoring stations and decision- and policy making processes.
Overall, the study needs to be further expanded in two directions. The first direction requires elaboration of ground-level BC measurements in Ukraine for modelling results validation and assessment of the negative BC impact on human health and local ecosystems in Ukraine with a future transition to mitigation measures and reduction strategy. The second direction should expand the analysis of direct and indirect aerosol effects by using seamless/ online-integrated modelling to distinguish the influences of different aerosol compounds."