#### I. Editor comments related to the response to the previous referee comments

Referee #3:

1) Q. Not entirely clear how statistical measures are averaged (lines 336-338). Are measures calculated for each site and then those values for each domain are averaged (e.g., the 5 Relative Humidity NRMSE values for the 5 KEN2K weather stations are averaged to produce the KEN2K NRMSE value?) Or are the observed and modelled data for all the sites within a domain used together to calculate the average measure?

A > The statistical analysis both for WRF and for CHIMERE has been done calculating the statistics for each station individually and the averaging all station together so that e.g., the 5 values of the individual relative humidity NRMSE are averaged to produce the final NRMSE value for the domain. The calculation has been done on the original hourly values from observations and model outputs and consider hourly values from the model only if the corresponding hourly observation is present. According to comments made by reviewer 4 and 5, MNB and RMSE have been substituted by MFB and MFE in the validation of WRF and CHIMERE.

# Editor: Is this information included in the text? If not, please do so.

A >> A new subsection of Methodology has been introduced following the editor's comments: 2.5 Statistical parameters. Inside it this information has been clearly stated: "The statistical analysis both for WRF and for CHIMERE has been done calculating the statistics for each station individually and the averaging all station together. The calculation has been done on the original hourly values from observations and model outputs and consider hourly values from the model only if the corresponding hourly observation is present." (Line 321 – 324)

2) Q. It would be helpful to specify how wind direction statistics were calculated. Since wind direction is a circular variable, calculating means, RMSE, etc. is different than for linear variables. Also, I'm not sure that normalized measures, MNB, NRMSE make sense for wind direction.

A > The statistics presented originally in the manuscript has been calculated as follows:

$$MNB = \sum (Mi - Oi) n i=1 \sum (Oi) n i=1$$
  
 $RMSE = \sqrt{\sum (Mi - Oi) n 2 i=1 n}$ 

As the review suggests these operators can be used for linear variables such as temperature and relative humidity but they haven't the same meaning for what concern circular variables like in the case of the wind direction. Moreover, they rely also on the number of observations point included in the denominator and the final value can be misleading. For this reason, the statistical analysis in the new manuscript has been changed and the MNB and RMSE values substituted with mean fractional bias and error (MFB and MFE) originally used only for the validation of CHIMERE. Moreover, for WRF we also use the Index of Agreement calculated as follows:

IOA = 1 - 
$$[\sum (O-M) n \ 2 \ i=1 \sum n \ (|M-O|+|O-O|) \ 2 \ i=1]$$

# Editor: Please add these equations to the manuscript, equivalently to Eqs. 2 and 3

A >> The equations relative to Pearson Coefficient R and Index of Agreement IOA have been added to the pre-existing equations of Mean Fractional Bias (MFB) and Error (MFE) in the new subsection 2.5 Statistical Parameters. The equations of MNB and RMSE haven't been added because these operations haven't been used in the final statistical analysis

3) Q. In the discussion of statistical evaluation of meteorological parameters, it would be helpful to include criteria for what constitutes "good agreement" (line 361), "acceptable agreement" (line 443), etc.

A > These qualitative terms have been deleted and the paragraphs modified to include quantitative statements.

Editor: In lines 522 and 525, you still use 'acceptable', in I. 410 'reasonable' without quantifying it. Please either avoid such statements or define them properly. In particular, you may want to consider whether the paragraph at the end of section 3.1.2 is completely needed. – What do you want to say here – specify 'acceptable' for what.

A >> The sentence in I.410 has been modified as follows: "The observed and modelled wind speeds in UGA2K, KEN2K and ETH2K <u>suggest an overestimation by the model</u> of 0.9, 0.8 and 0.2 m s<sup>-1</sup>, respectively (Table 3)."

The last paragraph at the end of section 3.1.2 has been modified as follows: "Nevertheless, the more detailed analysis of the urban weather stations revealed discrepancies in the reproduction of relative humidity and wind direction for the station of Kampala (UGA2K) that could affect the deposition, removal and transport processes simulated by CHIMERE and will be object of future investigation to further improve the meteorological performance of WRF. Even if the bias found for some variable in the calculation of the averaged statistics over all stations was high, the individual weather stations close to the urban areas of interest showed smaller bias and levels of MFB and MFE inside the goal or criteria range of performance and therefore considered acceptable for simulations." (Line 526 – 532)

4) Q. In Figure 8 the data for Nanyuki show what appears to be a nearly constant baseline PM2.5 concentration of around 2 to  $2.5 \mu g$  m-3 . Why would this be occurring?

A > The observations used to validate CHIMERE performance for Kenya comes from previous work by Pope et al., 2018 [1]. In that work the site of Nanyuki was chosen as rural spot in a location of minimum local air pollution influence. The data from Nanyuki has been used for the calculation of the net urban increment subtracting the rural background concentrations of Nanyuki from the urban concentrations in Nairobi. The average concentrations around 2  $\mu g$  m-3 in the period between the 4th and the 11th are the levels of the rural background in absence of any external influence from meteorological parameters and in absence of local sources. The peak of concentrations visible is the other days are between 4 and 15  $\mu g/m3$  that is in any case a low value in comparison with the concentrations from the urban area. The difference in the baseline concentrations is given by the big difference between the days with possible transport of pollutants from days where this phenomenon is not visible, but it is exaggerated by the low scale of the concentrations (0-16  $\mu g$  m-3 )

# Editor: Please add the relevant information on baseline PM to the manuscript.

A >> A new paragraph has been added at **line 709 – 714**: "The site of Nanyuki was chosen by Pope et al. (2018) as rural spot in a location of minimum local air pollution influence. Data from Nanyuki was used for the calculation of the net urban increment subtracting the rural background concentrations of Nanyuki from the urban concentrations in Nairobi. On the one hand, the average concentrations around 3-4  $\mu$ g m-3 in the period between the 4th and the 11th are, therefore, levels of the rural background in absence of any external influence from meteorological parameters and in absence of local sources."

5) Q. In presenting data table results, the text is often mainly just stating the values that are already shown in the tables. (e.g., sections 3.1.2, 3.2.1, 3.2.2) These sections could be condensed and/or modified to include additional description and discussion of what the data values mean.

Editor: In the revised manuscript, there are still instances of such descriptive text only listing values that are reported in the table without any discussion. Condense such texts (e.g. l. 418 -443) and add more interpretation as it has been done around l. 400.

A >> The discussion relative to the statistical parameters has been modified and the made more descriptive of the numbers reported in the text. The paragraphs are at **line 406 – 451** have been modified accordingly.

6) Technical corrections: Throughout the manuscript the authors mention "low air quality index". This could be interpreted as a low numerical value of the air quality index, indicating good air quality, but from the context it seems the authors are instead describing poor, or low, air quality. It would be better to use a different word than "low".

Editor: This comment was not addressed in the previous response. It is a fair concern as 'low air quality index' may be interpreted either as 'low index for air quality' or 'index for low air quality'. Please address it and replace 'low air quality (index)' by a less ambiguous expression.

A >> the expression low air quality index has been modified with poor air quality index throughout all the manuscript.

#### Referee #4:

### 1) Introductory comment:

.... In its current shape, this article sometimes looks like a technical report on the feasibility of a particular forecast system for specific regions, which is not really what is expected from a research article. I think that with the additions above, this article could give many more indications on the specificities on Particulate matter composition in this region, and yield more interesting questions for future research. I feel this article will deserve publication because they obtain a great performance in reproducing pollution in areas where this has rarely been attempted; Once major changes are brought (making the statistical discussion more straightforward and give more scientific material from the model outputs), I feel that this may become a breakthrough article for air quality modelling in Africa.

Editor: I agree with the referee that your article raises many important questions regarding air quality in Africa. I respect your response that your study is focused on presenting the model performance for a few locations but may be a considered a starting point for future analyses and additional aspects. Your paper would indeed benefit if you added a few sentences along those lines towards the end of the paper as an outlook on further research question that should be explored in forthcoming studies. This could be part of the conclusion section.

A >> A new paragraph has been added in the conclusion: "The present work represents a first step in the use of numerical models for atmospheric chemistry simulations in East Africa with particular focus on urban conurbation. The aim of the present work was to assess the possibility to perform simulations with results close to observations in order to open the road for more detailed works. The natural next step of the present research aims to refine the quantity and quality of the input data used for the validation of both modelling system in order to improve the reliability of the predictions. Moreover, a more detailed analysis of the secondary inorganic and organic components of PM2.5 will be conducted for the three domains. Finally, the performance of CHIMERE will be tested in the reproduction of

gaseous species too in order to give a wider vision of the capabilities and opportunities of numerical modelling in this area of the world with present data." (Line 901 – 909).

Fig. 9: It is not useful to compare modelled values in Nanyuki to observed values in Nyeri, 60km away in a mountain / plateau environment. No statistical link between the two timeseries can be expected a priori. I do not understand the point of the authors here, this should maybe be explained more.

A > The analysis of concentrations observed in Nanyuki takes in account that the location chosen by Pope et al. (2018) for the sampling of PM was a rural spot in a location of minimum local air pollution chosen to calculate the net urban increment subtracting the rural background concentrations of Nanyuki from the urban concentrations in Nairobi. The comparison that is proposed by Figure 8 it is only one of the options that can be taken in account considering the combined effect of meteorological parameters and location with higher contamination levels near Nanyuki that could influence the local level of PM. A first element to take in account to explain the peaks of contamination in Nanyuki could be the presence of local sources not accounted in the emission inventory used in CHIMERE. Despite this there is a clear change of trend in the concentration levels between February and March, in presence of local sources misrepresented we should see peaks at high concentration also in March but instead they are absent. A second element to take in account is the possible presence of precipitation during the period of March were the average concentrations of PM2.5 doesn't exceed the 2 µg/m3 but (Pope et al., 2018) affirm In their work that no rain was observed in that period and WRF model also doesn't model any in that particular period. We are aware that to support the thesis of transport phenomena additional further analysis (e.g., trajectory analysis) are required as well as more observational point along the way between Nyeri and Nanyuki. Further analyses are planned to go in that direction, what we argue in this paper is to give a possible explanation with the extent of the data available at the moment.

Editor: I am not convinced that your response fully addresses the referee's concern. Please explain in the manuscript why the comparison as performed in Figure 8 is justified.

A >> A new paragraph has been added to make the motivations for the analysis of Figure 8 more robust and justifiable: "The site of Nanyuki was chosen by Pope et al. (2018) as rural spot in a location of minimum local air pollution influence. Data from Nanyuki was used for the calculation of the net urban increment subtracting the rural background concentrations of Nanyuki from the urban concentrations in Nairobi. The average concentrations around 3-4  $\mu$ g m-3 in the period between the 4th and the 11th are, on one hand, levels of the rural background in absence of any external influence from meteorological parameters and in absence of local sources.

On the other hand, the presence of higher hourly peaks in before and after the 4th to 11th can be linked to different reasons: the presence of local emission sources contributing to the peaks or the dispersion of polluted air masses from elsewhere towards the site of Nanyuki. It is important to observe that model and observations seems to agree particularly well in the description of the difference in magnitude between the different time periods excluding the possibility that the observed values can be influenced by local emission sources not accounted in the emission inventory. It seems more likely that those concentration levels are transported to Nanyuki from neighbouring areas with higher levels of PM2.5 contamination. To investigate this possible role of PM2.5 dispersion towards Nanyuki, we consider the closest MIDAS weather station to the sampling area of Nanyuki, in the town of Nyeri (0.43°S, 36.95°E altitude 1916 m a.g.l.) (n10 in Figure 2). Nyeri is only 60 km from the Nanyuki site and is situated between Mount Kenya (0.10°S, 37.30°E, altitude 4341 m a.g.l.) to the west and the Aberdare Range (0.46°S, 36.69°E, altitude 3441 m a.g.l.)." (Line 709 – 725)

#### **II. Additional Editor comments:**

A. Comments regarding content and structure

I. 15: Add the model resolution here.

A >> Model resolution has been added.

I. 219: (1) which conversion factor from organic carbon to aerosol mass was applied? (2) replace 'for' by 'with' (multiplied with...)

A >> The conversion factor's value has been added and the text modified according to the suggestion.

I. 220: Why is it assumed that PM2.5 is only composed of carbon-containing components? How about other compounds, such as sulphate etc?

A >> The reason for the creation of PM2.5 using the carbon-containing component is motivated by the nature of the DICE emission Inventory that focuses on emissions particular diffuse and inefficient combustion emission sources (e.g., road transport, residential biofuel use, energy production and charcoal production and use). The variable PM2.5 created from DICE in this way has been then merged with the variable PM2.5 from EDGAR global emission inventory that contain information about inorganic fractions such as sulphate.

I. 320 – 325: This paragraph is neither a result nor a discussion of your results. Therefore, either connect it better to the results or remove it, as it seems out of place and redundant here.

A >> The paragraph has been deleted.

I. 327 - 375: This text is still a description of the methodology and therefore should be a subsection of Section 2. Lines 327 - 349 could be included in a subsection 'Statistical parameters'; I. 350 - 375 describes 'Model resolution and simulations.

A >> The manuscript has been modified accordingly and the information in line 327-375 moved in the new sections 2.5 Statistical Parameters and 2.6 Model resolution and simulations design

#### I. 585: 'CHIMERE 'better reproduces' than what?

A >> The text has been modified: "Despite this and considering the daily average concentrations in the urban sites, the R coefficients were found to be between 30 and 42 % suggesting that CHIMERE better reproduces the concentrations of PM2.5 <u>using daily than hourly values</u>." (Line 590-592)

I. 682 - 693: The model-observation comparison in Fig 7a shows clearly that the model tends to overestimate the PM2.5 concentration. If the emissions in the model were correct, one would expect the opposite trend – as you correctly describe, i.e., lower predicted values as compared to observations since the latter represent point measurements whereas the former are gird-averaged values. However, in Figure 7a, there seems to be a period where model/observation agreement is particularly poor (~ 28/02 – 05/03) that shows a very distinct trend, opposite to the expected one. What was different during this period? If indeed this discrepancy is due to an

# incomplete/inappropriate emission inventory in the model, can the characteristics of the air mass give a hint on the missing/wrong emissions as a function of air mass type/history?

A >> Thanks for highlight this aspect in the simulation. The period between the 28<sup>th</sup> of February and the 5<sup>th</sup> of March do actually show hourly peaks of PM2.5 at high magnitude not found in the observation. A Similar event is present also in the period 10<sup>th</sup> 11<sup>th</sup> of March. The reason for this particular behaviour is not straightforward. Assuming that the observations from the field sampling are correct, one reason can be connected to the values of wind directions modelled by WRF towards that particular grid cell corresponding to Tom Mboya Street where in particular hours of the day additional amount of PM2.5 is moved from the from neighbouring cells. An additional explanation can be connected to the inorganic and organic secondary formation of PM2.5 that can contribute to the high hourly peaks modelled but not observed.

Nevertheless, the magnitude of the emissions in itself should not represent a problem because the average description of the minimum and maximum levels of PM2.5 modelled by CHIMERE show reasonable agreement with the observations both in urban area (where the emissions are supposed to be high) and in rural area (where the emissions are supposed to be low).

Despite this, as the reviewer suggests the magnitude of the emissions can be generally appropriate to the average levels of PM2.5 but can still be misrepresented at hourly level in urban area. It will be absolutely important to continue the work on refining the anthropogenic emissions used for this type of simulations and a possible step ahead is the creation of local emission inventory at high resolution able to increase the level of detail of the real levels of PM2.5 in urban areas.

Table 7: Is this table necessary? It provides the same information as in I. 794 – 798 and in some of the following lines. I suggest removing it as it is neither a result nor part of their discussion.

A >> Table 7 has been deleted.

I. 847: The simulation of 'weather patterns' were not the main goal of this study but simulation of trends of air pollution.

A >> The sentence has been modified substituting weather patterns with variables that have actually analysed for the validation of the model.

- B. Technical comments (language, journal standards etc)
- I. 18: replace 'tool' by 'model'

A >> The text has been modified accordingly.

I. 37: define 'WWP' and add it as database to reference list as detailed on the journal website

https://www.atmospheric-chemistry-and-physics.net/submission.html#manuscriptcomposition

A >> The reference has been added.

I. 43: Can you give a reference to the data base? – Add to reference list.

A >> The reference has been added.

I. 54: add 'UN Habitat, 2017' to reference list

A >> The reference has been added.

I. 98/99: A verb seems to be missing in this sentence.

A >> The text has been modified accordingly.

I. 123: (1) Figures should be numbered according to their reference in the text. Since here Fig. 3 is cited before Figure 2, please change them accordingly. (2) remove 'a, b, c' here and in the remainder if the manuscript – see my comment below regarding 'panel 3d'.

A >> The figures have been moved in the manuscript and the legend modified.

I. 247 – 249: This sentence is quite convoluted given its rather simple message. How about "The emissions used in this work might not reflect the true values due to missing emission sources and the mismatch of the simulated time period and the date of the emission inventories."

A >> The text has been modified accordingly.

I. 276 – 278: I do not understand this sentence.

The sentence has been modified. The new sentence is: "The period chosen for the simulations of meteorology has to be representative of the average weather conditions of the analysed area and avoid unusual weather conditions (e.g., extreme events) that could impact the physical and chemical processes described in the CTM and affect the final concentrations of secondary pollutants simulated" (Line 283-286).

Table 2: (1) Spell out Latitude, Longitude, Elevation. (2) Use consistent terminology for latitude. Here you use – whereas later in the text, you specify S, N.

A >> The table has been modified accordingly.

Figure 3: Remove 'd)' from the last panel. It is a legend and therefore does not need a label. In the caption, replace 'in table d' by 'in the legend'.

A >> The image has been modified accordingly.

Table 3: Do not use random abbreviations in the table and caption. Spell out all words (obs., rel., ...) or define them in the caption (e.g., relative humidity (RH) which then can be used as RH in the table).

A >> The text has been modified accordingly.

I. 321: (1) remove 'from the real world'. (2) replace 'systems' by 'simulations'

A >> The text has been modified accordingly.

I. 345 & 347: See my comment above regarding referring to Figures in the correct order. For simplicity, I suggest removing the text in the parentheses here. You can refer to it later.

A >> The text has been modified accordingly.

I. 603: A subject is missing in this sentence (that starts with 'Is therefore...')

A >> The text has been modified accordingly.

I. 721 and 729: Correct the units (m s-1)

A >> The units have been modified accordingly.

I. 756: 'and' seems wrong here ('and large hotspots...') - should it read 'a'?

A >> The text has been modified accordingly.

I. 786: why 'e.g.,'? Is 25  $\mu g$  m-3 a limit set by the WHO for comparable areas?

A >> The text has been modified accordingly.

Table 6: (1) Replace 'WHO exceeding limit' by 'Exceedances of WHO limit'; (2) The last two columns do not include essential information: 'Ratio' is unclear and not very meaningful; 'model false positive' is described in the text and therefore does not need to be repeated here.

A >> the text has been modified and the last two column of Table 6 deleted.

Data availability: Please provide at a minimum the model input and output data in a public repository <a href="https://www.atmospheric-chemistry-and-physics.net/policies/data">https://www.atmospheric-chemistry-and-physics.net/policies/data</a> policy.html

A >> The emissions used for the simulations made by CHIMERE are available already in a public repository: https://doi.org/10.25500/edata.bham.00000695

The information relative to the availability of the other input data/models have been added in the section "Data Availability". This includes links for the download of both models and the observations of Meteorology. Observations of PM<sub>2.5</sub> for Nairobi are available upon request to Pope Francis and his team, while for Addis Ababa and Kampala are available upon request to the respective U.S. Embassies.

1127 – 1129: Provide complete information for these references.

A >> The references have been modified.