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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluating Trends and Seasonality in Modeled $PM_{2.5}$ Concentrations Using Empirical Mode Decomposition" *by* Huiying Luo et al.

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Reply to interactive comments on "Evaluating Trends and Seasonality in Modeled PM2.5 Concentrations Using Empirical Mode Decomposition"

Anonymous Referee #4

General Comments: This paper introduces a new approach for process-based model evaluation of speciated PM2.5, which allows for the assessment of the performance of regional-scale air quality models like CMAQ on the intrinsic time-dependent longterm trend and cyclic variations in daily average PM2.5 and its species. The authors tested

the method with time series data at three sites. The data are generally sound, whereas some results and discussions of the study are still lack of persuasion.

One major concern is about how well the current approach's performance is compared with the previously published methods and some over-interpreted conclusions. The other is that it is not sure that the difference between the model and the new approach evaluation results can be simply explained by the inadequate description of nitrate or organics in the model. As the authors noted, they obtained abnormally low correlations of synoptic scale NO3 at ATL and calls for a better representation of nitrate partitioning and chemistry. What about the results for the other two sites? The authors need to provide more information on such issues to make the conclusion robust.

We appreciate the time and effort devoted by the reviewer to provide suggestions that helped improve the quality of our paper.

Our temporal decomposition approach applied to PM2.5 and its speciated components is not directly comparable with the other approaches reported in the literature. To avoid any over-interpretation of the analyses, we have refrained from exploiting model performance on the characteristic time scales and have carefully aligned our interpretation with IMFs that are statistically significant (almost all seasonal cycles are statistically significant from noise as shown in Fig. S5). Also, the differences between observed and simulated total and speciated PM2.5 are driven by several factors discussed in the paper. We cannot conclude exclusively that there is inadequate description of nitrate or organics in the model. Other potential issues such as the improper allocation of emissions also contributed to the difference between model simulations and observations. To be specific, description of secondary organic matter formation and magnitude and variation of primary sources are emerging areas of research; NO3 formation pathways are likely inadequately represented in the employed model version, and its predictions are also strongly influenced by the uncertainties in NH3 emissions.

CMAQ fails to simulate the magnitude of NO3 at all three sites with very abnormal

r_IMFn. Moreover, NO3 is the only component that has low correlation on the synoptic scale at ATL. The poor performance for NO3 mentioned above at all three sites calls for the modeler to look at the representation of nitrate partitioning and chemistry as summarized in the conclusions: "The consistent large under/over-prediction of NO3 variability at all temporal scales and magnitude in the trend component, as well as the abnormally low correlations on the synoptic scale NO3 at ATL, calls for better representation of nitrate partitioning and chemistry."

Specific comments:

1) Introduction: "Evaluation of ten-year averaged monthly mean of PM2.5 simulated with WRF/Chem . . ." how does the model performance of PM2.5 compositions simulation should also be summarized to provide an intact view on the previous results.

Unfortunately, Yahya et al. (2016) only compared the overall 10-year average of the PM compositions (sulfate, ammonium, nitrate, EC, and total carbon) from ground-based observations to that of the model simulations as the background map. Thus, we are not able to make any conclusions on the seasonality of PM2.5 components.

2) Line 36: "and other natural species..." what do natural species refer to?

Natural species refer to PM2.5 non-anthropogenic components such as crustal material. We have changed this to "crustal elements" in the revised manuscript to avoid confusion.

3) Line 47: "monthly or seasonal means" means of speciated PM2.5?

The sentence is rephrased as: "monthly or seasonal means of total and/or speciated PM2.5."

4) Line 48: what do you mean by "ten-year averaged monthly mean"?

It is the monthly mean averaged over a period of ten years: ten-year averaged mean for Jan., Feb., . . .

5) Line 51: "with a phase shift of few months" please explain phase shift.

The phase shift refers to that in Fig. 4c (copied below) in Yahya et al. (2016). The definition is similar to what we used in the evaluation of the cyclic signals: "the phase shift of an IMF n is defined as the days an IMF decomposed from modeled time series has to be shifted to maximize the correlation (R_max) with the corresponding IMF from observed PM2.5 time series."

6) Line 55-57: ". . .long-term trends or interannual variations driven by climate change, emission control policies or other slow varying processes..." what is the main reason? Are there any previous results?

Changes in air quality concentrations, such as PM2.5, are driven by changes in emissions and meteorological processes which highly impact the transport, chemical reactions and deposition of air pollutants. Thus, long-term trends reflect the impact of long-term changes in emissions (they might be governed by local control policies on anthropogenic emissions or climate-impacted natural emissions), long-term meteorological conditions (climate) and other slow varying processes (e.g. ENSO). There is no "main reason" among them. Here, we are simply stating that averaging over very long time periods can conceal signals driven by slow-changing processes: "In addition, averaging of those monthly or seasonal means across multiple years may conceal the long-term trends or interannual variations driven by climate change, emission control policies or other slow varying processes." We are not certain what the reviewer's query is directed at. Thus, we have left the sentence unaltered.

7) Line 68-74: I do not think this paragraph is necessary for the manuscript.

Following the reviewer's suggestion, the paragraph has been deleted in the revised manuscript.

8) Line 311: "RENO is located close to the border with California and is affected by large wildfire breakouts in the western U.S. . .." Is there any evidence for this demon-

stration?

The location of Reno, NV and the impact of California fire on July 10, 2008 is illustrated in Figure 1 (copied below) from Gyawali et al. (2009). We have also demonstrated the impact of 2008 fire season earlier in Section 4.1: "The small variation in the estimated characteristic period of IMF6 is because this monitoring site is located in a wildfire prone region on the border of Nevada and California. Clear evidence can be seen from Fig. 4a that an extra annual cycle in the IMF6 of observations in the summer of 2008 is depicted, which is very possibly driven by the 2008 California Wildfires spanning from May until November."

9) Line 327-: "To sum up, the long-term trend at QURE is well simulated by the model." This is unlikely consistent with the data presented in Table 1.

Our statement is based on the fact that the model has captured the decrease (i.e., rate of change), even though the absolute magnitude of the trend/long-term component is overestimated (which is what is shown in Table 1-now Table 2). We have re-phrased the sentence to: "To sum up, the decreasing long-term trend at QURE is well simulated by the model."

10) Lines 333-335: "Species other than those in the available dataset may also play a considerable role in driving the agreements or disagreements between model simulations and observations of total PM2.5" What are the contribution of these species to PM2.5 at the studied sites?

We have decomposed the remaining components (Rem) and added an 8th line of figures for the trend component in Rem in Fig. 6. The overall concentration share (%) of the remaining components can be found in the newly added Table 1. We have also added Figure S6 in the supplement that shows time series of the concentration share of each component (e.g. OC/Total PM2.5 %).

11) Lines 367-368: "Both observed and simulated annual cycles at the RENO site are

largely contaminated by the extreme events lasting for several months that are not properly simulated" is it possible to remove the data of extreme events before simulation, in order to eliminate the contamination?

These extreme events are very likely caused by large wildfires. We can eliminate emissions from wildfires in model simulations, but there is no straightforward way to eliminate contributions of wildfires in the observations. Thus, we kept the original observations and CMAQ model simulations, which included wildfire emissions.

12) Lines 384-387: "Specifically, the anti-correlation likely points to an inaccurate representation of the seasonal variation of the non-carbonaceous portion of organic matter due to an improper representation of organic aerosols in the model version analyzed here; this problem has since been corrected in more recent releases of the CMAQ model." This sentence needs to be rewritten for clearance. And what does the noncarbonaceous portion of organic matter refer to?

The long sentence has been revised for clarity: "Specifically, the anti-correlation likely points to an inaccurate representation of the seasonal variation of the non-carbonaceous portion of organic matter due to an incomplete representation of organic aerosols in the model version analyzed here; newer versions of the CMAQ model include updated treatment of organic aerosols (e.g., additional SOA formation pathways, improvements in representation of primary OM emissions) which is likely to correct the mentioned features (Appel et al., 2017; Murphy et al., 2017; Xu et al., 2018)."

The non-carbonaceous portion of organic matter refers to the portion of organic matter consisting of oxygen, hydrogen, and nitrogen.

Minor:

13) Line 17: "chloride (Cl) organic"

Corrected.

14) Line 311: "U.S. as can been seen"

Corrected.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-1079/acp-2019-1079-AC2-supplement.pdf

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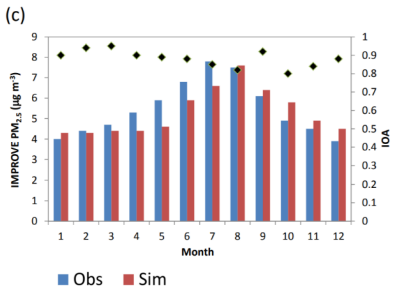

Fig. 4c in Yahya et al. (2016).

Fig. 1.

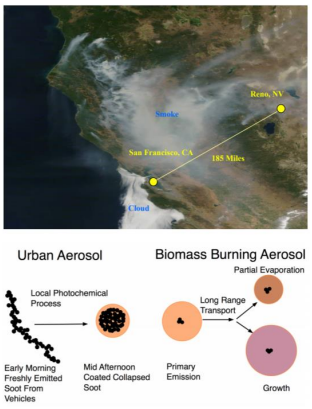

Fig. 1. Upper panel: Satellite image of smoke extending from northern California to Reno, Nevada on 10 July 2008. The smoke sources and wind trajectory were similar for much of July. Beneath panel: Conceptual model of emission and aging of urban and biomass burning aerosol.

Fig. 2.

Table 1. Concentration share (%) of different components in total $PM_{2.5}$. It is estimated by dividing the mean trend components of each species by that of total $PM_{2.5}$ for both OBS and CMAQ, multiplied by 100. The concentration share of the remainder species (*Rem*) is estimated by subtracting all the available species share from 100 to compensate for the small discrepancies caused by the rounding up process and uncertainty in the mode decomposition. "-" indicates the data is not available (same applies for all other tables).

		SO_x	NO_y	NH_x	OC	EC	Cl	Rem
QURE	OBS	38	7	-	19	5	1	30
	CMAQ	19	15	-	14	5	1	47
RENO	OBS	7	13	5	46	11	-	20
	CMAQ	11	4	2	30	7	-	45
ATL	OBS	28	6	10	24	8	-	24
	CMAQ	22	10	8	17	9	-	33

Fig. 3.