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Evaluating trends and seasonality in modeled $PM_{2.5}$ concentrations using empirical mode decomposition

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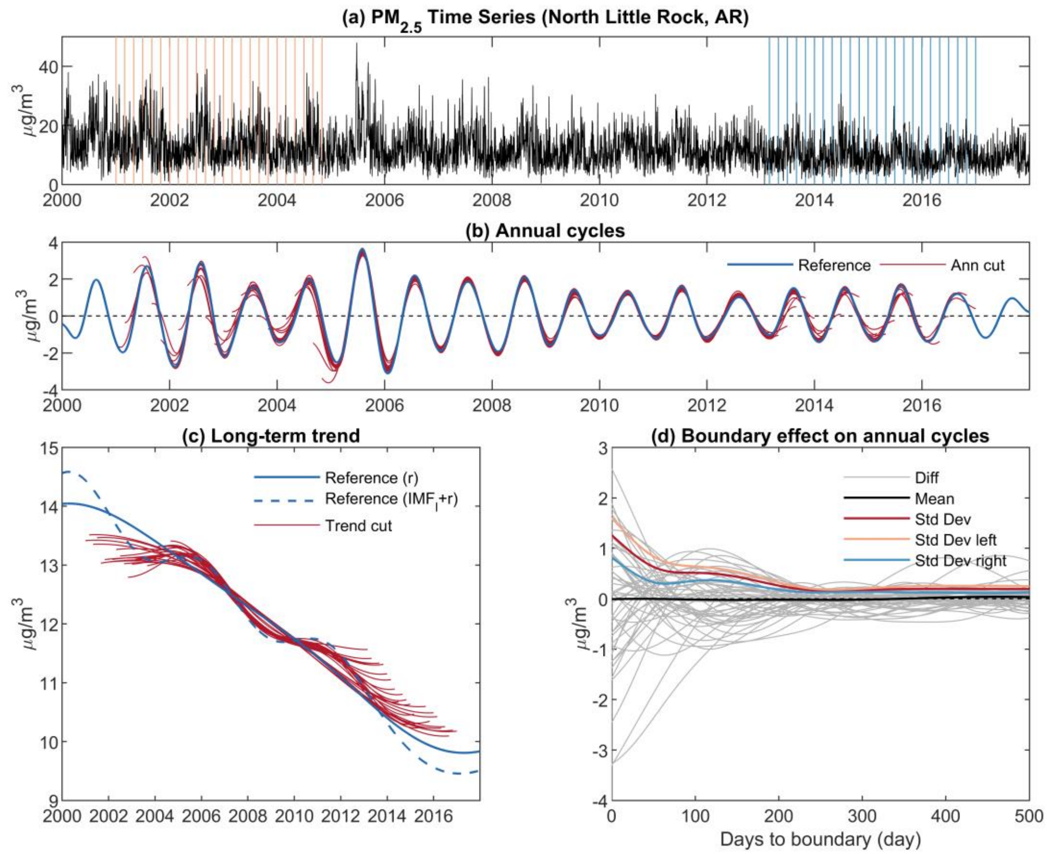

Fig. S1. Evaluation of the boundary effect on annual cycles and long-term trend with eighteen years of total $PM_{2.5}$ observation at North Little Rock, AR. (a) time series of total $PM_{2.5}$ (average data coverage: 97%; gaps are filled with most recent available observation); (b) annual cycles; (c) long-term trend; (d) boundary effect on annual cycles. Vertical colored lines in (a) indicate the 48 hypothetical boundaries by cutting the data before the left boundaries (light red) and after the right boundaries (light blue). In (b) and (c), annual cycles and long-term trend decomposed from the entire studied period are used as reference (blue) and those from the cropped time series are plotted in red. Specifically for the long-term trend, the summation of the residual and the last IMF (IMF_l) with longest temporal scale are shown in dashed blue for better comparison with the trend of the shorter cropped time series. The boundary effect in (d) is assessed by the statistics of the difference (*Diff = Ann cut - Reference*) in annual cycles as a function of number of days to the hypothetical boundaries: mean of all difference in black, standard deviation of all difference in red, standard deviation of those from the left/right boundaries in light red/blue as in (a). Difference in the level of standard deviation of left/right boundaries is very likely driven by the amplitude of the local annual cycles.

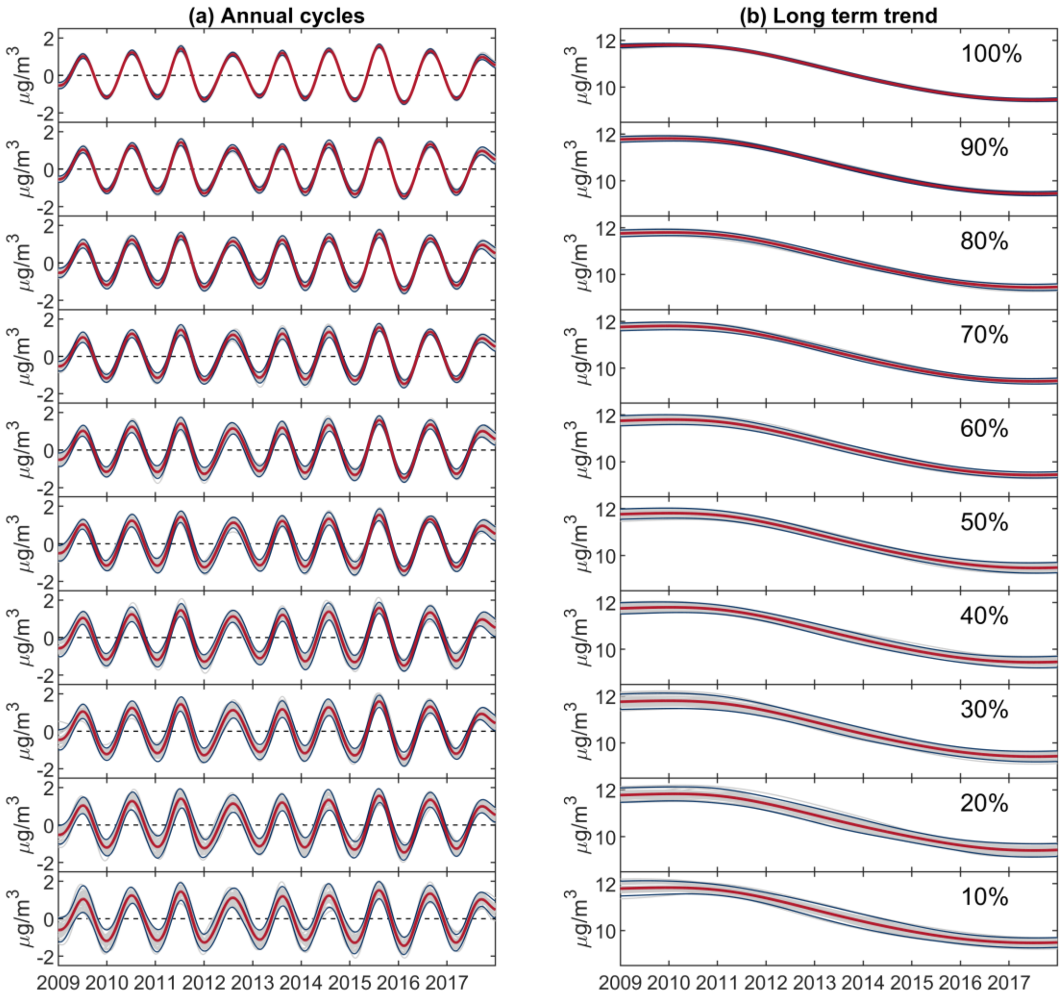

Fig. S2. Time series of (a) annual cycles and (b) long-term trend decomposed from the same total $PM_{2.5}$ observation as in Fig. S1, but for the period of 2009-2017 when average data coverage is almost 99%. Light gray lines indicate the components decomposed from $PM_{2.5}$ time series with data coverage of 100%, 90%…,10% from top to bottom (40 realizations in each data completeness scenario; in each realization, certain percentages of observations are randomly drawn from the original time series and filled with linear interpretation before the decomposition). The red thick line represents the average of those 40 realizations in each scenario and the thin dark blue lines indicate the average $\pm$ two times the standard deviation.

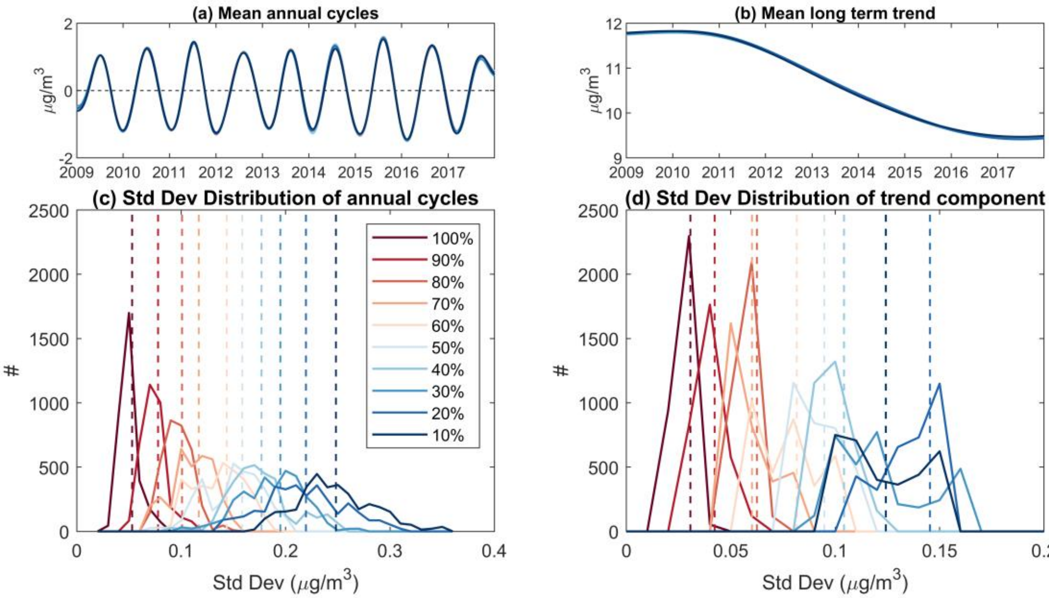

Fig. S3. The impact of data completeness on decomposed annual cycles and long-term trend. Time series of (a) averaged annual cycles and (b) averaged long-term trend of 40 realizations in each data completeness scenario as shown with red thick lines in Fig. S2; standard deviation distribution (9*365 data points in each distribution) of (c) annual cycles and (d) long-term trend for different data completeness scenarios. Same color scheme applies to all subplots as shown in (c). Vertical dashed lines in (c) and (d) are their corresponding median value of the distribution. Note the perfect alignment of the components shown in (a) and (b).

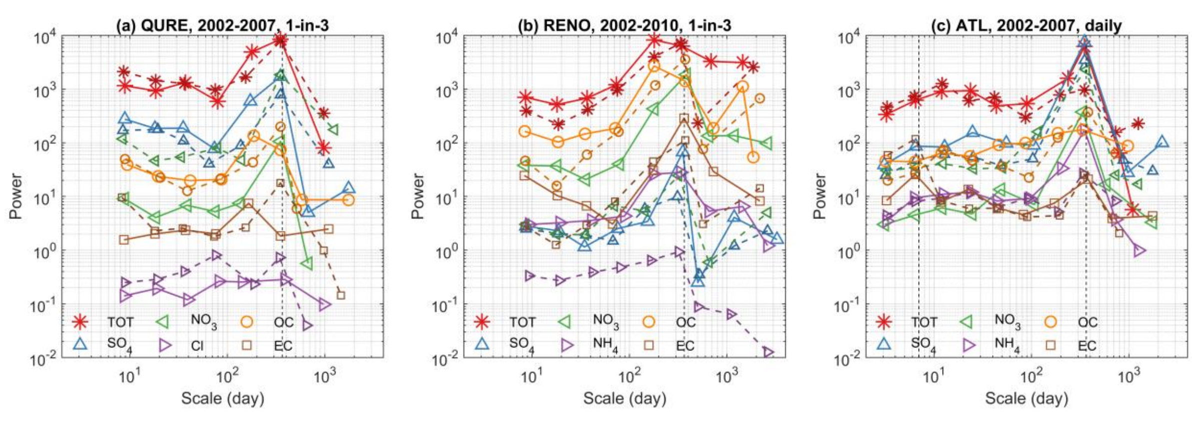

Fig. S4. The peak of power spectrum of each IMF of observed and simulated total and speciated $PM_{2.5}$ for (a) QURE, (b) RENO and (c) ATL. IMF1 to the last pair of IMFs with increasing characteristic periods are shown from left to right. Species decomposed from observations are connected by solid lines, while species decomposed from simulations are represented by smaller markers in darker shades connected by dashed lines.

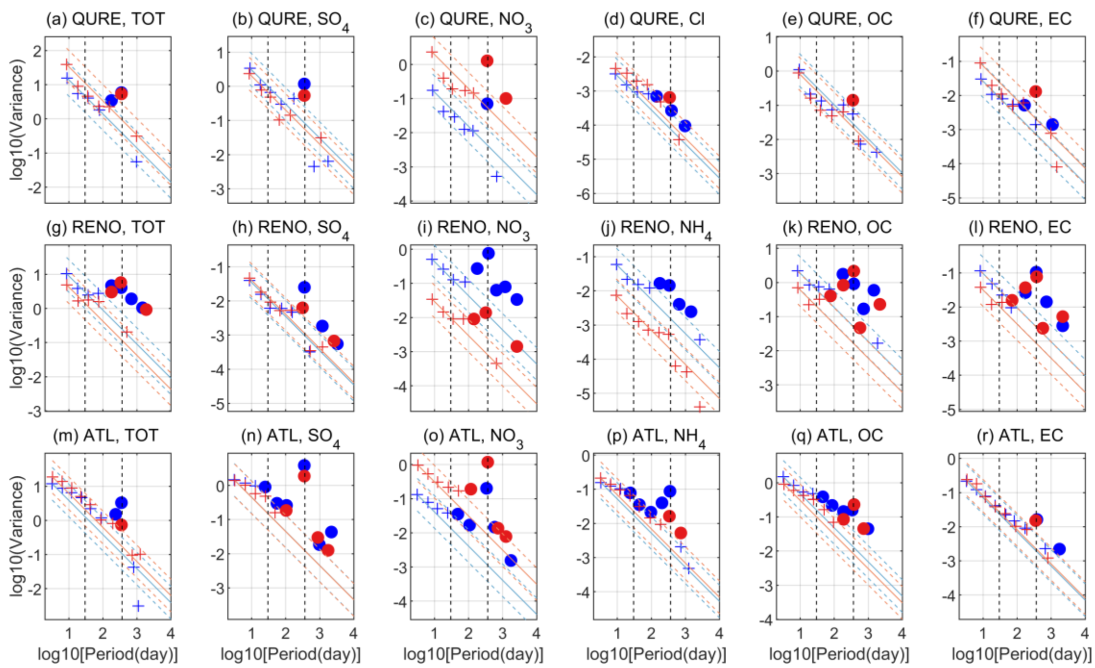

Fig. S5. Statistical significance test results of IMFs decomposed from observed (blue) and simulated (red) total and speciated $PM_{2.5}$ for (a-f) QURE, (g-l) RENO and (m-r) ATL following the method used in Wu et al. (2007). IMF1 to the last IMF with increasing characteristic periods are shown from left to right with filled circles representing statistically significant IMFs and plus signs indicating those not distinguishable from the corresponding IMFs of a pure white noise series. Vertical dashed black lines indicate the period of a month (left) or year (right). The colored solid lines indicate the variance of IMFs of white noise with its first IMF containing the same variance as that decomposed from observation (light blue) or model simulation (light red). The dashed lines above/below the solid line in the corresponding color are the upper/lower bound of a 95% confidence interval estimated by taking three times/one third of the variance of the white noise decomposed IMFs. Any IMFs above the upper bound is deemed statistically significant from those decomposed from white noise.

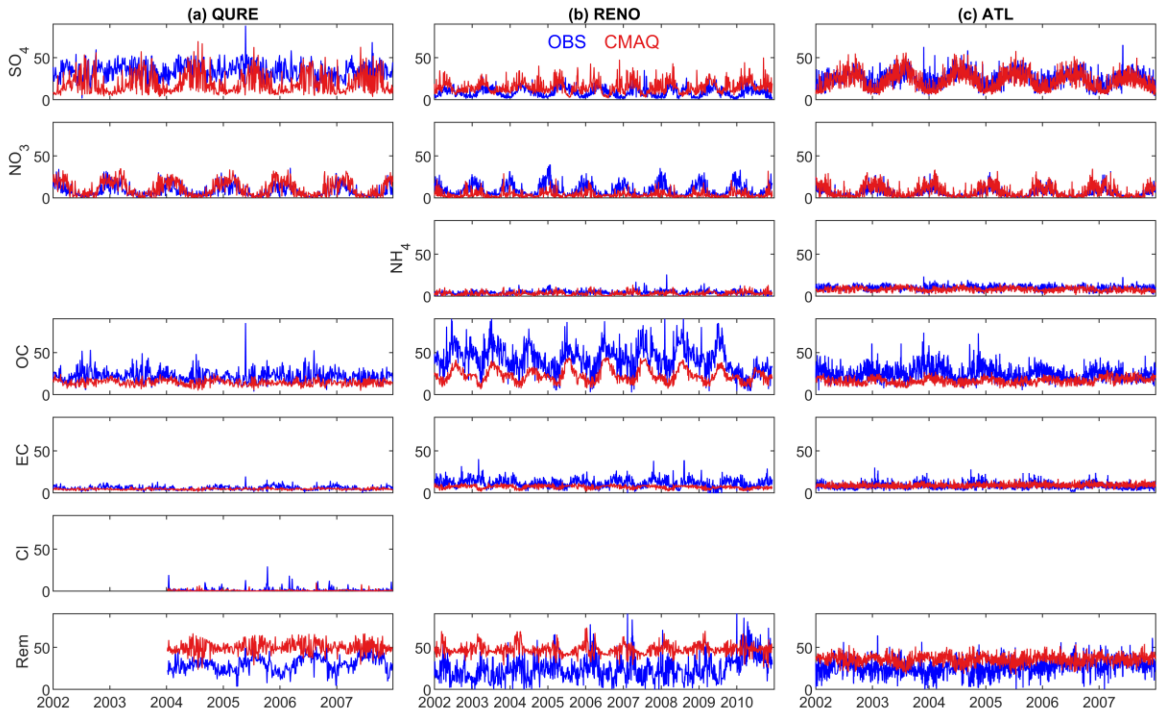

Fig. S6. Variations of the concentration share (%) of different components in total $PM_{2.5}$. It is estimated by dividing daily concentrations of each species by that of total $PM_{2.5}$ for both OBS and CMAQ, multiplied by 100. The concentration share of *Rem is* estimated by subtracting all the available species share from 100.