B. Rooney et al., "Impacts of Household Sources on Air Pollution at Village and Regional Scales in India"

Responses to Referee Comments

We sincerely appreciate the care with which the Reviewers evaluated this manuscript. This was a particularly challenging project, combining gaseous and particulate emissions inventories over India, WRF meteorological modeling, gas- and aerosol-phase chemical transport modeling by GEOS-Chem and CMAQ, and analysis of in situ measurements in north-central India. (The paper includes 21 authors from 7 institutions in the United States and India.) In the following responses, we address each of the comments of the Reviewers. All the simulations in the work have been redone. This includes comprehensive meteorology with WRF, domain boundary conditions with GEOS-Chem, and gas-phase and particulate matter concentrations with CMAQ. GEOS-Chem was run for each simulation time period (September 2015, December 2015, and September 2016) to supply the boundary conditions to CMAQ. The meteorology input was re-generated by WRF based on more accurate parameterizations and the most up-to-date initialization data. We now include more detailed statistical analyses of predictions and observations than in the original manuscript. Use of updated meteorology resulted in considerably improved agreement between predictions and observations for all PM components. As a result, a major fraction of the paper has been rewritten in accord with the revised computational simulations. In the accompanying revised manuscript, all new material is presented in red typeface.

Referee 1

General comments:

The residential source is a major source sector for air pollution in India. Studying the impact of household source to air pollution is important for policymaking. The topic of this study is suitable for the ACP. However, major revisions are needed to further improve the study. The contents need to be re-organized, and the model performance should be evaluated cautiously before analyzing the results and drawing the conclusions. A discussion on the uncertainty and limitation of this study is also needed.

Specific comments:

1. Line 10-15 on page 6: "Thus, daily emission rates are generated for all species and sectors, except for the residential sector. All emissions are assumed to occur at Earth's surface." How do the authors derive the daily and hourly emission rates of different emission sectors for the CMAQ model? It seems limited information is available for the temporal profiles of most of the sectors other than residential. And assuming all the emissions occur at Earth surface will overestimate the impact of high-stack emissions sources to the ground-level concentrations, such as power and some industrial sectors.

We have replaced the previous material with a new description, see page 5, lines 5 through line 31.

2. Line 25-30 on page 10: Why not directly run the GEOS-Chem model in 2015 and 2016 for the exact corresponding dates of CMAQ simulation?

We have now run GEOS-Chem directly for September 2015, December 2015, and September 2016. All CMAQ simulations have been re-run with the appropriate GEOS-Chem boundary conditions.

3. Page 14-15: One and a half pages are spent to review the previous studies on ozone simulation in India. The review is too long, which distracted the reader's attention. It's better to first present the author's results, and then have proper discussion with information from previous studies. Please re-organize the contents.

We have shortened the review of previous studies on O_3 in India. Lines 7-22 on p. 14 of the original manuscript have been removed.

4. Line 20-25 on page 9: Why do the authors choose Sep. 2015, Dec. 2015 and Sep. 2016? Do the three periods have some relevance? What's the intention to choose three discrete periods?

The periods September 2015, December 2015, and September 2016 were chosen because actual measurements ($PM_{2.5}$, O_3, temperature, and wind direction) were carried out in the region of interest during these months.

5. Page 41: The comparison between simulation and observation shows that the model always underestimates the ozone concentration during night, close to zero. What is the reason? Is this reasonable?

At night, when radiation is minimal, O_3 is consumed by NO titration reactions. In the present study, CMAQ simulations overpredicted NO, particularly at night when NO peaks, leading to the underestimation of ozone at night. This effect could be exacerbated by an underestimated planetary boundary layer height or weak vertical mixing.

6. Line 25-28 on page 15: "This overall comparison of predictions and observations would appear to be driven by the accuracy of the meteorological fields generated by the model." Indeed, the performance of air pollutant predictions can be largely affected by the meteorological conditions simulated by WRF. It's necessary to evaluate the model performance of WRF prior to CMAQ evaluation, which may provide hints for the inconsistency between the observation and simulation.

We now have added a new figure and a new table in the text to show the evaluation of WRF simulated meteorology against the available surface observations at different sites during the same periods. The new Figure 5 shows that there is generally good agreement of surface temperature between WRF and observations for all three months. The surface wind direction is also found consistent between model and observations for each site and each month (Table 9). The simulated near-surface wind speeds are overestimated by WRF, with an averaged mean-bias (MB) of about +1.5 m/s. Such a bias is partly a result of the difference in the definition of “near surface” between the model and observations. The new Figure 5 and new Table 9 follow.

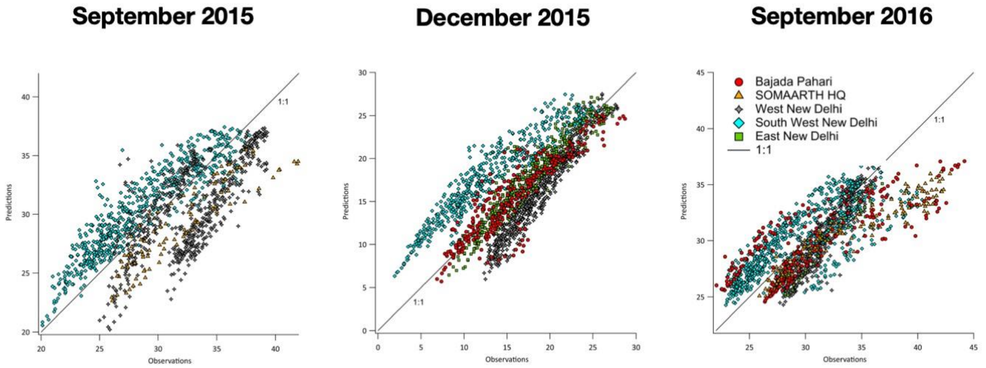

Figure 5. Evaluation of WRF simulated surface temperature versus ground observations.

		Bajada Pahari			**SOMAARTH HQ**			**West New Delhi**			**South New Delhi**		
		Sep '15	**Dec '15**	**Sep '16**	**Sep '15**	**Dec '15**	**Sep '16**	**Sep '15**	**Dec '15**	**Sep '16**	**Sep '15**	**Dec '15**	**Sep '16**
Temperature (°C)	**PRE**	-	15.28 *(4.59)*	30.10 *(3.19)*	29.27 *(3.48)*	-	30.22 *(3.06)*	30.45 *(3.79)*	16.59 *(4.91)*	30.07 *(3.05)*	30.32 *(3.74)*	17.59 *(4.82)*	29.96 *(30.3)*
	OBS	-	15.62 *(4.91)*	30.86 *(5.67)*	32.15 *(4.12)*	-	33.26 *(5.31)*	32.80 *(3.60)*	19.04 *(3.66)*	31.46 *(2.33)*	28.48 *(4.30)*	12.58 *(5.52)*	29.22 *(4.22)*
	MB	-	-0.34	-0.76	-2.89	-	-3.04	-2.35	-2.45	-1.38	1.84	5.02	0.74
	ME	-	1.60	3.08	2.92	-	3.07	3.03	2.58	1.54	2.11	5.02	2.37
	RMSE	-	2.20	3.71	3.39	-	3.99	3.58	2.99	1.88	2.50	5.33	2.75
Wind Speed ($m \cdot s^{-1}$)	**PRE**	-	2.91 *(1.17)*	2.31 *(1.07)*	-	-	2.01 *(0.66)*	-	-	2.57 *(1.28)*	2.80 *(1.27)*	2.72 *(1.08)*	2.74 *(1.39)*
	OBS	-	1.18 *(0.75)*	0.73 *(0.40)*	-	-	0.55 *(0.30)*	-	-	1.03 *(0.51)*	1.26 *(0.83)*	0.94 *(0.71)*	1.18 *(0.79)*
	MB	-	1.72	1.58	-	-	1.46	-	-	1.54	1.54	1.77	1.56
	ME	-	1.75	1.62	-	-	1.50	-	-	1.58	1.61	1.82	1.62
	RMSE	-	1.96	1.85	-	-	1.66	-	-	1.88	1.85	2.01	1.84
Wind Direction (°)	**PRE**	-	247 *(111)*	116 *(45)*	272 *(70)*	-	111 *(51)*	-	-	179 *(98)*	206 *(118)*	254 *(97)*	191 *(96)*
	OBS	-	259 *(57)*	102 *(41)*	255 *(58)*	-	110 *(48)*	-	-	181 *(97)*	198 *(45)*	224 *(44)*	228 *(50)*
	MB	-	0.14	14	16	-	-0.14	-	-	-6	9	35	-34
	ME	-	51	38	44	-	32.71	-	-	49	94	74	75
	RMSE	-	66	51	64	-	47.50	-	-	64	106	87	90

Table 9. Quantification of WRF model biases in meteorological fields. PRE is mean predictions; OBS is mean observations; MB is mean bias; ME is mean error; and RMSE is root mean square error. Standard deviation of predictions and observations are noted in parentheses.

7. Line 27-28 [on page 15]: "In general, the degree of agreement between predicted and observed O_3 levels in New Delhi over these periods should be considered as reasonable." The conclusion here is not convincing. Please first do the statistics for the model evaluation, and then compare with previous studies to see whether it's reasonable or not.

We have now redone all CMAQ simulations. We address the agreement between the new O_3 predictions and observations in New Delhi on page 12. Statistics are included in Table 10.

8. Line 15-28 on page 15: Please explain the reasons of underestimation and over estimation of ozone for different monitoring sites.

The results of ozone simulations in the present study are generally consistent with those of previous simulations over India. Virtually all studies of ozone modeling in urban areas in the literature show sites where concentrations are overestimated and sites where concentrations are underestimated. For example, also using WRF-CMAQ, Kota et al. (2018) showed that the relative bias in ozone simulation ranges from −30% to +50% in major cities of India. In South New Delhi, for example, the bias in O_3 predictions in the present study lies between -2.67 and +7.01 μg m^{-3}, as compared to the observations of 29.28 to 62.76 μg m^{-3} (see Table 10).

Kota, S.H., Guo, H., Myllyvirta, L., Hu, J., Sahu, S., Garaga, R., Ying, Q., Gao, A., Dahiya, S., Wang, Y., and Zhang, H., 2018. Year-long simulation of gaseous and particulate air pollutants in India, Atmospheric Environment, 180, 244-255.

9. Page 16-17: Somehow, the model performance for $PM_{2.5}$ is also not satisfactory: large underestimation occurred in SOMAARTH HQ in Sep. 2015, Bajada Pahari in Dec. 2015, whereas over estimation occurred in Bajada Pahari in Sep. 2016, and New Delhi in Sep. 2015 and Sep. 2016. Before moving further, the authors should evaluate the model performance in details, analyze the reasons for the underestimation and overestimation, and compare the model performance with other studies.

All CMAQ simulations have been redone with updated meteorology from WRF (see response to Referee 1, Comment 6), updated GEOS-Chem boundary conditions (see response to Referee 1, Comment 2), and updated emissions inventory to include biogenic VOC emissions (see response to Referee 1, Comment 1). The comparisons of predictions and observations are now entirely revised and with new Figures 6 through 9. A discussion of model performance is provided on pages 15-16 and statistics are detailed in Table 10.

10. Line 2-4 on page 18: "September 2015 and 2016, household energy-use activities account for up to 33% of ambient $PM_{2.5}$ at SOMAARTH HQ and up to 28% at Bajada Pahari in September 2016." With current model performance, it's hard to trust the reliability of the results. Since a large proportion of $PM_{2.5}$ mass concentration is missing compared with the observation.

All CMAQ simulations have been redone (see response to Referee 1, Comment 9). Statistical comparisons of predictions and observations are now included, see Table 10. New discussion of the contribution from household energy-use activities is provided on page 11, line 22 through page 12, line 2.

11. Now that the study also analyzed the household contribution to SOA, a model evaluation for SOA is also needed.

Direct measurements of the SOA fraction of PM are, unfortunately, not available in this region. Nonetheless, information is given throughout the manuscript on the levels of SOA predicted.

12. A discussion on the uncertainty and limitation of this study is missing.

A discussion of uncertainty is now provided, see page 13, lines 31-36.

Referee 2

General comments:

Rooney and coauthors have undertaken a worthwhile modeling study on the impact of residential combustion sources on air quality in rural and urban India. The goal, scope, and methods of the paper are well-suited to ACP. They should also be commended for their efforts processing emissions, meteorology and land use data for this simulation – all challenging tasks. The conclusions drawn are a good start to a conversation about contributions from residential burning to $PM_{2.5}$ and ozone. However, I find significant areas of improvement possible in the explanation of methods, consideration of underlying assumptions, and discussion of results. Please address the concerns below.

1. Quantitative and statistical analysis. The manuscript includes many statements like "whereas for the end of the month, predicted levels match closely those observed" (page 15, line 23), "In general, the degree of agreement between predicted and observed O_3 levels in New Delhi over these periods should be considered as reasonable" (page 15, line 27-28) and "the finer resolution computations generally predict somewhat higher $PM_{2.5}$ concentrations than the coarser computations" (page 16, lines 9-11). These kinds of statements need to be supported with rigorous statistical analysis. I would expect some subset of bias, error, RMSE, fractional bias, fractional error, correlation coefficient, index of agreement, etc. to be provided in any contemporary air modeling study with access to observation data. Aggregate metrics can often fail us if we rely too much on them, but they do help summarize the overall performance of the model against other studies.

The authors also rely primarily on timeseries plots comparing model predictions to observations for raw concentrations. It is usually useful to look at these comparisons with diurnal plots as well as raw timeseries to see if there are persistent issues at certain times of day; I recommend adding these for the data presented in figures 6-11. It's also important to add error bars to illustrate variability on all diurnal timeseries plots, including figures 12 and 16.

I also would have expected to see maps of output data for metrics like fractional SOA contribution or fractional residential combustion contribution. Of course, it is just a model result, but I would think experts in air quality issues in India would find such maps interesting to ponder since this data cannot be obtained with existing measurements alone.

Statistical analyses of predictions and observations are now described in detail, see Table 10. Timeseries plots comparing model predictions to observations have been completely revised to include updated model predictions and diurnal plots.

2. Species-level evaluation. Are there any observed speciation data that can help evaluate aspects of the model like the POA/SOA split? For example, OC/EC ratios might be instructive. It would also be useful to know if the model is predicting individual inorganic ion components well; do these data exist?

Unfortunately, there are no species-level data in this region for this period. We have now included discussion of model predictions of $PM_{2.5}$ speciation, see pages 10-11 and Table 10.

3. Meteorology evaluation. An evaluation of the meteorology fields has been completely omitted. This must be provided (e.g. supplemental information) or referenced if published somewhere else. The met evaluation would also enhance the value of the timeseries plots of raw concentration data. If performance trends are seen across the month, it is important to find out if they are correlated with biases in any meteorological parameters.

See response to Referee 1, Comment 6.

4. Model spinup. This lack of model spinup for the individual scenarios is potentially troubling, considering that the authors are focusing on periods with meteorological conditions that favor accumulation (i.e. low wind speeds, low boundary layers, cold temperatures, etc.). Spin up is happening at the GEOS-Chem domain, so it is not as bad as starting from clean conditions, but there is a resolution change in going from the GEOS-Chem field to the CMAQ parent field and again for the nested field. It would be safer to at least consider omitting some of the timeseries for model spin-up. I recommend one of the following: a) rerun WRF and CMAQ simulations beginning about 10 days before each time period (i.e. August 20 for September simulations). For emissions, if you don't have specific emissions for the spinup days, it should be safe to reuse appropriate days from the main simulation. For example, if August 20 is a weekday, make sure you pick a weekday from the main simulation. b) alternatively, cut off the first 7-10 days of each month from the existing results. Based on the timeseries data, it looks like making this revision will sacrifice minimal model-obs pairs for the PM dataset anyway.

All CMAQ simulations have been redone (see response to Referee 1, Comment 9), with slightly longer time periods. The September 2015 and September 2016 simulations now begin five days earlier. Additionally, the first five days of each simulation were excluded as spin-up from analysis and plots.

5. Impact of Conclusions. Finally, the authors have summarized in the abstract and conclusions sections some important results of their work: a) residential combustion makes a significant contribution to air quality impacts in India, b) the model sometimes performs well, but sometimes does not, and this is a result of meteorological and emissions errors, and c) SOA makes up a significant but minor fraction of the total OA, with varying temporal profile between urban and rural areas. The authors have assembled a wealth of data and numerical predictive techniques to create this dataset; are there any more conclusions that can be drawn from this work if analyzed deeper?

For example, if one population-weights the emission rates of total PM or SOA precursors across the parent domain, what sources stand out as the most impactful? The authors have shown that residential emissions do not matter as much in New Delhi as they do at the rural sites, but how does this translate to people impacted? And what does the emissions dataset say are the most important sources of primary $PM_{2.5}$, secondary $PM_{2.5}$ precursors, and Ozone precursors?

Another question to consider (aspects of this are repeated in some specific issues below) is how confident are the authors in their estimate of SOA contributions given the uncertainty in emissions speciation of SVOCs and IVOCs, SOA yields of aromatics, and potential degradation pathways like homogeneous fragmentation or particle-phase reaction/photolysis, among other uncertainties? I'm not calling for a suite of sensitivity studies here, but considering the author list contains unique experts in emissions and chemistry, it would be great to understand and document where this team thinks most of the uncertainty lies at this point for SOA predictions in India. Or, alternatively, what parameters need more investigation before such conclusions can be drawn confidently?

Specific Issues and Typos:

1. Page 1: Consider mentioning residential heating as well as cooking since it does become a significant part of the discussion in the results section.

Residential heating is not in the inventory, see response to comment 5 below.

2. Page 2, line 4: Consider replacing "pollution species" with "pollutants" since I wouldn't consider $PM_{2.5}$ to be a species, technically.

Correction made.

3. Page 3, Introduction: There's little to no mention of ozone here, which seems strange given its importance in the discussion later. Consider highlighting it here in parallel to PM.

Now included.

4. Page 4, line 16: What does this sentence mean? There is no "prediction" for SOA in India? Do you mean there is no inferred measurement of it? Or that there is a dearth of reported model predictions to compare to? Please be more precise.

We have clarified this statement. Whereas the CMAQ model predicts the fraction of $PM_{2.5}$ that is SOA, ambient measurements do not delineate the fraction of SOA.

5. Page 7, line 14: This statement asserts that there are no emissions from heating in the study, but Figure 2 clearly shows parameters used to inform the heating emissions. Please explain the heating emissions more clearly.

Residential emission rates for $PM_{2.5}$, black carbon (BC), organic carbon (OC), CO, NO_x, CH_4, CO_2, and total non-methane hydrocarbons (NMHC) were generated from SPEW. While SPEW incorporates temperature-dependent heating combustion activity, the inventory assumes temperatures too high for this activity to take effect. Thus, our inventory has no emissions from heating. Nonetheless, Figure 2 shows the fraction of daily household emissions from all quantifiable energy-use activities.

6. Page 7, line 15: This sentence makes it sound like there are no solvent emissions because the simulation is in August. I suggest rewriting and stating why there are not any solvent emissions. I assume it's from a lack of input data?

This statement has now been clarified. Solvent emissions are not included in the simulations for lack of specific input data.

7. Page 8, line 5: Burning of biomass fuel can give compounds with quite high OM:OC. How robust are your results to this simplifying assumption?

The issue is the accuracy with which the mass of biomass fuel emissions is simulated by CMAQ. Specific data on the OM:OC ratio of biomass burning emissions in this region are not available. CMAQ has been used in other studies to predict the mass of combustion emissions based on the correlations in CMAQ. While no specific data are available to evaluate the extent to which the CMAQ algorithm accurately represents the concentration of biomass burning emissions, $PM_{2.5}$ was speciated using Jayarathne et al. (2018), who studied emissions from burning wood and from burning a mixture of wood and dung, but not purely dung. The PM emission profiles for these fuels are now given in Table 3.

8. Page 8, line 6: PNCOM, PMC and PH_2O are ignored in the emissions. Do you have references for the validity of assuming they are negligible? I understand omitting coarse PM since you are studying $PM_{2.5}$. However, studies from airsheds like LA have shown that how a model treats inorganic aerosol in the coarse mode can be important for fine mode predictions. What is your basis for assuming negligible effect?

Coarse PM is included in the emissions (see Tables 1-3), and this line has been revised accordingly. PNCOM and PH_2O are omitted for lack of information.

9. Page 8, line 9-10: How much of the $PM_{2.5}$ emissions is carbon? Shouldn't it be included in this equation?

The equation and lines have been corrected, see page 6, lines 28-30.

10. CMAQv5.2 assumes one volatility distribution for all POA, while this study considers POA from a variety of fuels and burning methods. What uncertainties are introduced here and what impact do they have on conclusions?

No explicit information is available on the volatility of POA. However, it is likely that POA has a sufficiently low volatility, as assumed in CMAQ v5.2. It is not expected that this assumption introduces any significant uncertainty.

11. The resolutions of the coarse and fine grids should be mentioned in the emissions methods discussion as it would help the reader understand right away the challenge involved in specifying emissions for this simulation.

The resolution is now given.

12. Page 11, line 9: Did you use 4-D data assimilation to 'nudge' the met model to observations within the domain? If not, how often did you reinitialize the met model with realistic fields? One of these approaches should probably have been used to keep the met model realistic over a month-long simulation. If neither were used, it makes a met evaluation all the more important to demonstrate or reference to better understand any growing biases.

We do not use any nudging or data simulation approach in our WRF simulations. Three one-month simulations were performed without any re-initialization, which is a typical way to run the WRF model for regional weather/climate simulations [*Wu et al.*, 2017; *Kota et al.*, 2018]. Our simulated meteorological fields also show no systematic drift-away within a month, and they are quite consistent with ground-based observations, as discussed in our response to the major comment 3 above.

Kota *et al.*, Year-long simulation of gaseous and particulate air pollutants in India, Atmospheric Environment, 180, 244-255 (2018).
Wu *et al.*, WRF-Chem simulation of aerosol seasonal variability in the San Joaquin Valley Atmos. Chem. Phys. 17, 7291-7309 (2017).

13. Page 13: Could the authors please discuss why they feel it is appropriate to omit the pcSOA species, which is based on the SIMPLE method of Hayes et al. (2015)? Is this because the speciation studies used for these residential emissions are believed to account for all of the particle and vapor phase mass that may be missed in inventories of the United States NEI? Page 13, line 11 says SVOCs could not be quantified. This mass is potentially important and represents part of what the pcSOA is designed to address. Can the authors be more explicit about what classes of organic compounds are accounted for by their emissions speciation and which might be underrepresented? Discussing this in the context of volatility would suffice. For example, are IVOC precursors for SOA missing as well?

We have now included pcSOA in our revised CMAQ simulations.

14. Page 14: Much of the content in the first four paragraphs of section "ozone" is better suited for the introduction section than the results.

As noted above, the discussion of ozone simulations has been significantly shortened.

15. Page 16, line 12: Recommend replacing "closeness" with "similarity". How did you verify this statement? Perhaps make a timeseries of the emission rate of residential primary PM (median and error bars) across the child domain for the 4 km and 1 km cases?

The 1 km simulations have been removed from the revised manuscript as the initial results showed little difference between the two resolutions. Additionally, non-residential emissions were adapted from an Indian inventory with a native resolution of 36 km and boundary conditions have a resolution of 2° × 2.5°. Because of the coarseness of these inputs, it is unlikely that the difference between 1 km and 4 km chemical transport simulations is significant.

16. Page 16, lines 17-20: Is this statement true at all sites? Or is there variation in the importance of daily-varying emissions like agricultural burning?

All CMAQ simulations have been updated with new inputs and the comparisons of predictions and observations are now entirely revised (see response to Referee 1, Comment 9).

17. Page 18, lines 19-21: This sentence confused me. It says the emission inventories are the same, but that the main differences between scenarios is explained by differences in POA? I'm probably not understanding what scenarios the authors are comparing (e.g. seasons or resolution or sensitivities). It would be helpful to be more precise here.

With the new simulations, this comment is no longer relevant.

18. Page 19, lines 11-18: I'm confused again. Which conclusions are from Fleming et al., and which are new to this study? If they're all from Fleming et al., how do they compare with the contribution CMAQ is predicting for this work? Again, if aromatics are currently the dominant source of SOA, how much could be coming from IVOCs and potentially being missed?

The summary of results from Fleming et al. has been removed.

19. Figure 1: 4 km grid cell should read 4 km grid domain.

Figure 1 has been revised.

20. Figure 4: Recommend adding some visual cues for the flow of information from the parent domain all the way through to the nested domain. For example, add "4 km resolution" to the "Gridded Airborne Concentrations" box. Then duplicate that box, but with "1 km resolution" instead. You could then color the arrows based on whether they are relevant for the 1 km or 4 km simulation. For example, purple for 4 km and orange for 1 km. So the arrow from GEOS-Chem to IC-BCs would be purple. And the arrow labeled "Nested Domain" would be orange. The arrows connecting IC-BCs, Emissions and Met to CMAQ would be both orange and purple. Also, shouldn't there be an arrow from GEOS-Chem to the Met box? And from the Met box to Emissions? Maybe this is more detail than you want to include? You probably don't want to use the phrase "Parent Domain" in the GEOS-Chem box since I assume this was a global simulation? CMAQv5.2 Chemical Transport "Module" should read "Model".

With the omission of the nested 1 km simulations, we have removed Figure 4.

Referee 3

This manuscript presents a multi-resolution modeling study of ozone and particulate matter in India, with a focus on the impact of residential combustion on ozone and particulate matter concentrations. The authors apply the CMAQ model at several different horizontal grid resolutions for several different time periods in 2015/2016. They then analyze the total concentrations of ozone/$PM_{2.5}$ as compared to observations and examine the contribution of residential combustion of sources for cooking to the total $PM_{2.5}$ and total SOA. Overall, the manuscript is generally well written. However, I agree with the points made by the other anonymous referees, and concur that the manuscript would benefit greatly from some major revisions. I will not repeat the general suggestions made by the other referees here, only to say that I agree with their major points. I will however provide some specific comments in addition to those made by the other reviewers.

General Comments:
Very briefly, I agree that the authors spend a lot of time discussion previous work upfront. It would be nice more details of their own analysis could be provided and then compared to the previous studies as appropriate. More details and analysis of both the WRF and CMAQ simulations is required, particularly the WRF simulations. The point was made by the other referees, but I think it is necessary to reiterate here the importance of providing an analysis of the performance of the WRF simulations. Hopefully there are observations available to provide such an analysis. Giving the reader a clearer picture of the strengths and weaknesses in the WRF simulations would be very beneficial to supporting the analysis of the CMAQ simulations and the conclusions the authors are attempting to make. Related, there are large biases in the CMAQ simulations that at bring into question some of the conclusions made by the authors, particularly those attributing percent contribution of residential cooking to total $PM_{2.5}$ and total SOA. Providing a clearly picture of the CMAQ model performance would help support these conclusions (I refer the authors to the comments provided by the other referees). The authors should provide summary statistics (e.g. RMSE, bias, correlation) of the various simulations.

See responses above to comments of Referees 1 and 2.

Specific comments:
Page 7, line 30: How different are the profiles for wood and dung? Why not use a combination of the two if the percent contribution in a grid cell is known?

$PM_{2.5}$ was speciated using Jayarathne et al. (2018), who studied emissions from burning wood and from burning a mixture of wood and dung, but not purely dung. The PM emission profiles for these fuels are now given in Table 3.

Page 8, line 21: This appears to be sentence fragment to me. Please fix.

The sentence in question is no longer in the paper.

Page 10, line 26: As mentioned the other referee, it's a strange setup to be using the same month/day of the GEOS-Chem simulation to support the various CMAQ simulations. Why was this done?

This has been addressed. See response to Referee 1.

Page 11: What input data did the authors use to drive the WRF simulations? Reanalysis data (if so, what was the horizontal resolution)? Were the same data used for all three horizontal domains? How many vertical layers were used in the WRF simulations, and what was the height of the lowest layer? What Cumulus Parameterization (CP) scheme was used? Was a CP scheme applied for all the simulations?

Now we update our meteorological forcing by switching to the latest version of ECMWF-ERA5 which was just released in January 2019. These reanalysis data are on a 30 km grid and resolve the atmosphere using 137 levels from the surface up to a height of 80 km. In the WRF model, we use 24 levels in vertical, consistent with our setup in the CMAQ model. The lowest layer is about 50 m. No cumulus parameterization is turned on in our simulation, as we only have one domain with 4 km resolution.

Page 11, line 18: What version of MCIP was used? Also, please provide a relevant reference for MCIP.

MCIPv4.4 (Otte et al., 2010) was used to process WRF for CMAQ.

Otte, T. L. and Pleim, J. E.: The Meteorology-Chemistry Interface Processor (MCIP) for the CMAQ modeling system: updates through MCIPv3.4.1, Geosci. Model Dev., 3, 243-256, https://doi.org/10.5194/gmd-3-243-2010, 2010.

Page 11: What was the CMAQ configuration (e.g. vertical layers, mixing scheme, etc.)? As mentioned by the other referee, it does not appear that a spin-up was applied. I agree with the referee that it would be beneficial to provide a spin-up period, even if not starting from profile ICs.

Analysis of the updated CMAQ simulations excludes five days for spin-up (see response to Referee 2, Comment 4).

Page 15, line 25: As mentioned by the other referees, the author's statement that the "overall comparison of predictions and observations would appear to be driven by the accuracy of the meteorological fields" is on point, however no analysis of the accuracy of those fields is provided, so the reader has no idea what role the meteorological fields are actually playing in the analysis. So, these comparisons need to be provided since they are no doubt very important to the analysis and conclusions the authors are making.

See response to Referee 1, Comment 6.

Page 16, line 13: The statement that "The closeness of the 4 km and the1 km simulations reflects the closeness of the respective inventories" seems to only capture part of the story. What about differences in the meteorological performance between the two simulations? It would seem that the reader is intended to assume that the meteorological fields are also very similar between the 4 km and 1 km simulations. Is that really the case?

The 1 km simulations have been omitted from the revised manuscript (see Referee 2, Comment 15).

Page 19, lines 5-20: I'm confused whether the authors are describing new results from their own analysis or simply summarizing the results of Fleming et al.? Please clarify whether the statements are new results or simply summary of results from Fleming.

Discussion of results from Fleming et al. has been removed as not essential to the present work.

Page 20, line 10: The statement that "overall good agreement between observed and predicted levels O_3 levels" is a bit generous. Based solely on the time-series plots and without any summary statistics, I think the authors at best could call the results reasonable. There are certainly frequently very large biases that make calling the results "good" difficult to support. However, I believe that even reasonable results are sufficient to support the conclusions the authors are making regarding ozone performance.

Summary statistics are now provided in Table 10. See response to Referee 1, Comment 7.

Page 21, line 5: The statement regarding the importance of replacing household combustion devices with modern technology is not strongly supported by the analysis presented. If anything, it would seem that reducing the impacts from agriculture burning would go much further in improving people's exposure to $PM_{2.5}$ in this region. That is not to say that improving technology is not beneficial, it's just not the message I think that is delivered by the analysis as it is currently presented.

As noted above, all CMAQ simulations have been redone corresponding to more accurate meteorological fields. As a consequence, predictions of the average fractions of total anthropogenic $PM_{2.5}$ and secondary organic $PM_{2.5}$ due to household emissions increased (see Figure 12). In addition, the minimal impact of the residential sector on $PM_{2.5}$ remains consistently higher than the previous simulations. We feel it is appropriate to state that more modern cooking technologies would be beneficial. Therefore, we have left the sentence in question in the paper.

New text as the result of revising the manuscript in response to the reviews appears in red.

Impacts of Household Sources on Air Pollution at Village and Regional Scales in India

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Abstract. Approximately 3 billion people worldwide cook with solid fuels, such as wood, charcoal, and agricultural residues. These fuels, also used for residential heating, are often combusted in inefficient devices, producing carbonaceous emissions. Between 2.6 and 3.8 million premature deaths occur as a result of exposure to fine particulate matter from the resulting household air pollution (Health Effects Institute, 2018a; World Health Organization, 2018). Household air pollution also contributes to ambient air pollution; the magnitude of this contribution is uncertain. Here, we simulate the distribution of the two major health-damaging outdoor air pollutants (PM$_{2.5}$ and O$_3$) using state-of-the-science emissions databases and atmospheric chemical transport models to estimate the impact of household combustion on ambient air quality in India. The present study focuses on New Delhi and the SOMAARTH Demographic, Development, and Environmental Surveillance Site (DDESS) in the Palwal District of Haryana, located about 80 km south of New Delhi. The DDESS covers an approximate population of 200,000 within 52 villages. The emissions inventory used in the present study was prepared based on a national inventory in India (Sharma et al., 2015, 2016), an updated residential sector inventory prepared at the University of Illinois, updated cookstove emissions factors from Fleming et al. (2018b), and PM$_{2.5}$ speciation from cooking fires from Jayarathne et al. (2018). Simulation of regional air quality was carried out using the U.S. Environmental Protection Agency Community Multiscale Air Quality modeling system (CMAQ), in conjunction with the Weather Research and Forecasting modeling system (WRF) to simulate the meteorological inputs for CMAQ, and the global chemical transport model GEOS-Chem to generate concentrations on the boundary of the computational domain. Comparisons between observed and simulated O$_3$ and PM$_{2.5}$ levels are carried out to assess overall airborne levels and to estimate the contribution of household cooking emissions. Observed and predicted ozone levels over New Delhi during September 2015, December 2015, and September 2016 routinely exceeded the 8-hour Indian standard of 100 µg m^{-3}, and, on occasion, exceeded 180 µg m^{-3}. PM$_{2.5}$ levels are predicted over the SOMAARTH headquarters (September 2015 and September 2016), Bajada Pahari (a village in the surveillance site, September 2015, December 2015, and September 2016), and New Delhi (September 2015, December 2015, and September 2016). The predicted fractional impact of residential emissions on anthropogenic PM$_{2.5}$ levels varies from about 0.27 in SOMAARTH HQ and Bajada Pahari to about 0.10 in New Delhi. The predicted secondary organic portion of PM$_{2.5}$ produced by household emissions ranges from 16% to 80%. Predicted levels of secondary organic PM$_{2.5}$ during the periods studied at the four locations averaged about 30 µg m^{-3}, representing approximately 30% and 20% of total PM$_{2.5}$ levels in the rural and urban stations, respectively.

1 Introduction

Although outdoor air pollution is widely recognized as a health risk, quantitative understanding remains uncertain on the degree to which household combustion contributes to unhealthy air. Recent studies in China, for example, show that 50-70% of black carbon emissions and 60-90% of organic carbon (OC) emissions can be attributed to residential coal and biomass burning (Cao et al., 2006; Klimont et al., 2009; Lai et al., 2011). Moreover, existing global emissions inventories show a significant contribution of household sources to primary PM$_{2.5}$ (particulate matter of diameter less than or equal to 2.5 micrometers) emissions. The Indo-Gangetic Plain of Northern India (23-31° N, 68-90° E) has among the world's highest values of PM$_{2.5}$. In this region, the major sources of emissions of primary PM$_{2.5}$ and of precursors to secondary PM$_{2.5}$ are coal-fired power plants, industries, agricultural biomass burning, transportation, and combustion of biomass fuels for heating and cooking (Reddy and Venkataraman, 2002; Rehman et al., 2011). The southwest monsoon in summer months in India leads to lower pollution levels than in winter months, which are characterized by low wind speeds, shallow boundary layer depths, and high relative humidity (Sen et al., 2017). With the difficulty in determining representative emissions estimates (Jena et al., 2015; Zhong et al., 2016), simulating the extremely high PM$_{2.5}$ observations in the Indo-Gangetic Plain has remained a challenge (Schnell et al., 2018).

Approximately 3 billion people worldwide cook with solid fuels, such as wood, charcoal, and agricultural residues (Bonjour et al., 2013; Chafe et al., 2014; Smith et al., 2014; Edwards et al., 2017). Used also for residential heating, such solid fuels are often combusted in inefficient devices, producing black carbon (BC) and organic carbon emissions. Between 2.6 and 3.8 million premature deaths occur as a result to exposure to fine particulate matter from household air pollution (Health Effects Institute, 2018a; World Health Organization, 2018). In India, more than 50% of households report use of wood or crop residues, and 8% report use of dung as cooking fuel (Klimont et al., 2009; Census of India, 2011; Pant and Harrison, 2012). Residential biomass burning is one of the largest individual contributors to the burden of disease in India, estimated to be responsible for 780,000 premature deaths in 2016 (Indian Council of Medical Research et al., 2017). The recent GBD MAPS Working Group (Health Effects Institute, 2018b) estimated that household emissions in India produce about 24% of ambient air pollution exposure. Coal combustion, roughly evenly divided between industrial sources and thermal power plants, was estimated by this study to be responsible for 15.3% of exposure in 2015. Open burning of agricultural crop stubble was estimated annually to be responsible for 6.1% nationally, although more important in some areas.

Traditional biomass cookstoves, with characteristic low combustion efficiencies, produce significant gas- and particle-phase emissions. An early study of household air pollution in India found outdoor total suspended particulate matter (TSP) levels in four Gujarati villages well over 2 mg m^{-3} during cooking periods (Smith et al., 1983). Secondary organic aerosol (SOA), produced by gas-phase conversion of volatile organic compounds to the particulate phase, is also important in ambient PM levels, yet there is a dearth of model predictions to which data can be compared. Overall, household cooking in India has been estimated by various groups to produce 22-50% of ambient $PM_{2.5}$ exposure (Butt et al., 2016; Chafe et al., 2014; Conibear et al., 2018; Health Effects Institute, 2018b; Lelieveld et al., 2015; Silva et al., 2016), and Fleming et al. (2018a,b) report characterization of a wide range of particle-phase compounds emitted by cookstoves. In a multi-model evaluation, Pan et al. (2015) concluded that an underestimation of biomass combustion emissions, especially in winter, was the dominant source of model underestimation. Here, we address both primary and secondary organic particulate matter from household burning of biomass for cooking.

Air quality in urban areas in India is determined largely, but not entirely, by anthropogenic fuel combustion. In rural areas, residential combustion of biomass for household uses, such as cooking, also contributes to non-methane volatile organic carbon (NMVOC) and particulate emissions (Sharma et al., 2015, 2018). Average daily $PM_{2.5}$ levels frequently exceed the 24-hour Indian standard of 60 µg m^{-3} and can exceed 150 µg m^{-3}, even in rural areas. The local region on which the present study focuses is the SOMAARTH Demographic, Development, and Environmental Surveillance Site (DDESS) run by the International Clinical Epidemiological Network (INCLEN) in the Palwal District of Haryana (Figure 1). Located about 80 km south of New Delhi, SOMAARTH covers an approximate population of 200,000 in 52 villages. Particular focus in the present study is given to the SOMAARTH Headquarters (HQ) and the village of Bajada Pahari within DDESS, coinciding with the work of Fleming et al. (2018b), who studied cookstove non-methane hydrocarbon (NMHC) emissions and ambient air quality. Demographically, with a coverage of almost 308 sq km, the DDESS has a mix of populations from different religions and socioeconomic and development statuses.

The climate of the region of interest in the present study is primarily influenced by monsoons, with a dry winter and very wet summer. The rainy season, July through September, is characterized by average temperatures around 30 °C and primarily easterly and southeasterly winds. In a study related to the present one, Schnell et al. (2018) used emission datasets developed for the Coupled Model Intercomparison Project Phases 5 (CMIP5) and 6 (CMIP6) to evaluate the impact on predicted $PM_{2.5}$ over Northern India, October-March 2015-2016, with special attention to the effect of meteorology of the region, including relative humidity, boundary layer depth, strength of the temperature inversion, and low level wind speed. In that work, nitrate and organic matter (OM) were predicted to be the dominant components of total $PM_{2.5}$ over most of Northern India.

The goal of the present work is to simulate the distribution of primary and secondary $PM_{2.5}$ and O_3 using recently updated emissions databases and atmospheric chemical transport models to obtain estimates of the total impact on ambient air quality attributable to household combustion. With respect to ozone, the present work follows that of Sharma et al. (2016) who simulated regional and urban ozone concentrations in India using a chemical transport model and included a sensitivity analysis to highlight the effect of changing precursor species on O_3 levels. The present work is based on simulating the levels of both O_3 and $PM_{2.5}$ at the regional level based on recent emissions inventories using state-of-the-science atmospheric chemical transport models.

2 Emissions Inventory

2.1 Non-Residential Sectors Emissions

The present study uses an emissions inventory conglomerated from two primary sources: (1) an India-scale inventory for all non-residential sectors prepared by TERI (Sharma et al., 2015, 2016) and (2) a high-resolution residential sector inventory detailed here. Emissions data from each source were distributed to a 4 km grid for the present study. The TERI national inventory was prepared at a resolution of 36 × 36 km^2 using the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS ASIA) emission model (Amann et al., 2011). GAINS ASIA estimated emissions based on energy and non-energy sources using an emission factor approach after taking into account various fuel-sector combinations. Following the approach of Kilmont et al. (2002), the emissions were estimated using the basic equation:

$$E_k = \sum_l \sum_m \sum_n A_{k,l,m} ef_{k,l,m} \left(1 - \eta_{l,m,n}\right) \cdot X_{k,l,m,n} \tag{1}$$

where E denotes the pollutant emissions (in kt); k, l, m, and n are region, sector, fuel or activity type, and control technology, respectively; A the activity rate; ef the unabated emission factor (kt per unit of activity); η the removal efficiency (%/100); and X the application rate of control technology n (%/100) where $\sum X = 1.$ Energy sources considered include coal, natural gas, petroleum products, biomass fuels, and others and categorized into five sectors – transport, industries, residential, power, and others. The model uses the state-wise energy data and generates emissions of species such as PM, NO_x, SO_2, NMVOCs, NH_3, and CO.

For activity data of source-sectors, TERI employed published statistics (mainly population, vehicle registration, energy use, and industrial production) where possible. Energy use data for industry and power sectors were compiled based on a bottom-up approach, collected from the Ministry of Petroleum and Natural Gas (MoPNG, 2010), the Central Statistics Office (CSO, 2011), and the Central Electricity Authority (CEA, 2011). Transportation activity data were compiled from information on vehicle registrations (Ministry of Road Transport and Highways, 2011), emission standards (MoPNG, 2001), travel demand (CPCB, 2000), and mileage (TERI, 2002). Emission factors for energy-based sources from the GAINS ASIA database were used. Speciation factors are adopted from sector-specific profiles from Wei et al. (2014), primarily developed for China as there is a lack of information for India. In the transportation sector, the Chinese species profiles are dependent on fuel type but not technology.

The TERI inventory was compiled on a yearly basis, with monthly variations for brick kilns and agricultural burning, at a native resolution of 36 × 36 km^2 then equally distributed to grid resolution of 4 × 4 km^2 for this study. Emissions for non-residential sectors have no specified diurnal or daily variations; thus, the inventory for non-residential sectors is the same for each simulated day. Transportation sector emissions were estimated using population and vehicle fleet data at the district level and distributed to the grid using the administrative boundaries. Industry, power, and oil and gas sector emissions were assigned to the grid by their respective locations. Emissions from agriculture were allocated by crop-types produced by state in India. The inventory was vertically distributed to three layers with the lowest layer extending to 30 – 43 m, the middle layer to 75 – 100 m and the top layer to 170 – 225 m layers. VOC emissions were assumed to occur only in the bottom layer. Industry and power emissions were distributed based on stack heights and allocated to the second and third layers.

We incorporated biogenic emissions by using daily-averaged emission rates of isoprene (0.8121 moles s^{-1}) and terpenes (0.8067 moles s^{-1}) per 4 km grid cell, predicted by GEOS-Chem for the region of study. The TERI inventory additionally includes isoprene emissions from the residential sector, so isoprene from natural sources was calculated as the difference of the total rate predicted by GEOS-Chem and the rate of emissions solely from the residential sector. Terpene emissions are assumed to occur only in non-residential source-sectors. Isoprene and terpene emission rates were applied to all computational cells as an hourly average (with no diurnal profile) in the non-residential inventory.

2.2 Residential Sector Emissions

To examine local and regional impacts of residential sector emissions in greater detail, an update to the TERI inventory was performed using various sources to consider more granular input data specific to the residential sector (Table 1). Bottom-up estimates of delivered energy for cooking, space heating, water heating, and lighting were informed by those used in Pandey et al. (2014) and converted to fuel consumption at the village level using population size and percentage of reported primary cooking and lighting fuels from the 2011 Census of India (Census of India, 2011). Urban areas of the domain were assumed to have the average cooking and lighting fuel use profiles of the average urban areas of their district. Fuel consumption was converted to emission rates using fuel-specific emission factors informed by a review of field and laboratory studies, which was used to update the Speciated Pollutant Emissions Wizard (SPEW) inventory (Bond et al., 2004) and to generate summary estimates by fuel type. Hourly emissions were generated using source-specific diurnal emissions profiles (Figure 2). The same diurnal emissions profile is applied to all species from a source category and were informed by real-time emissions measurements taken in homes during cooking reported by Fleming et al. (2018a,b). Profiles for fuel-based lighting were informed by real-time measurements of kerosene lamp usage data reported in Lam et al. (2018). The residential sector inventory represents surface emissions with a native spatial resolution of 30-arc seconds (~1 km).

In deriving summary estimates of emission factors, priority was given to emission factor measurements from field-based studies. Several studies have shown that laboratory-based measurements of stove and lighting emissions tend to be lower than those of devices measured in actual homes (Roden et al., 2009), perhaps due to higher variation in fuel quality and operator behavior. Field-based emission factors utilized in this study include those for non-methane hydrocarbons, measured from fuels and stoves within the study domain (Fleming et al. 2018a,b). $PM_{2.5}$ speciation from cooking fires was informed by Jayarathne et al. (2018) (Tables 2 and 3). Residential emission rates for $PM_{2.5}$, black carbon (BC), organic carbon (OC), CO, NO_x, CH_4, CO_2, and total non-methane hydrocarbons (NMHC) were generated from SPEW, which estimates emissions from combustion by fuel type. As such, solvent emissions are not included for lack of specific input data. Additionally, while SPEW incorporates temperature-dependent heating combustion activity, the inventory assumes temperatures too high for this activity to take effect. Thus, our inventory has no emissions from heating.

We employed various methods to account for pollutant species not explicitly reported by SPEW (Tables 1 and 2). Gas-phase SO_2 and NH_3 emissions were informed by existing residential emissions in the TERI inventory (Sharma et al. 2015); NO and NO_2 were estimated from NO_x emissions assuming a NO:NO_2 emission ratio of 10:1. Total NMHC and $PM_{2.5}$ emission factors from SPEW are distributed by fuel type (wood, dung, agriculture residue, or LPG) (Table 2). Given the low $PM_{2.5}$ emission rate of LPG, (Shen et al., 2018), emissions from LPG are assumed to be negligible. To further speciate NMHCs, we employed HC species-specific emission factors (Fleming et al. 2018b), differentiated by fuel and stove type (i.e. traditional stove, or *chulha*, with wood or dung, and simmering stove, or *angithi*, with dung). We assume that all NMHC emissions in each computational grid cell are produced by either wood or dung, whichever contributes the greater fraction of total $PM_{2.5}$ emissions in that cell (Figure 3). The NMHC emission profile of dung was assumed to be the average of measurements from chulha and angithi stoves. The emission profile for agricultural residue is similar to that of wood; therefore, wood speciation profiles are applied in cells where agricultural residue dominates.

Particle-phase speciation of total $PM_{2.5}$ was based on PM mass emissions from wood- and dung-fueled cooking fires as reported by Jayarathne et al. (2018), and primary cooking fuel type distribution data from the 2011 census (Tables 2 and 3). A single $PM_{2.5}$ speciation profile, defined as the average of that of wood and that of the wood-dung mixture, was applied in all cells for lack of information on pure dung emissions (Table 3). Non-carbon organic particulate matter (PNCOM) and particulate water (PH_2O) were assumed to be negligible owing to lack of information on these species. Emissions of remaining particle-phase species (i.e. Al, Ca, Fe, Mg, Mn, Si, and Ti) are also assumed to be negligible for lack of information. Unspeciated fine particulate matter (PM_{othr}) is defined in CMAQ as the portion of total $PM_{2.5}$ unassigned to any other species:

$$\mathrm{PM_{othr}} = \mathrm{PM_{2.5}} - \left(\mathrm{P_{EC}} + \mathrm{P_{OC}} + \mathrm{P_{Na}} + \mathrm{P_{NH_4}} + \mathrm{P_K} + \mathrm{P_{Cl}} + \mathrm{P_{NO_3}} + \mathrm{P_{SO_4}}\right) \qquad (2)$$

Tables 4 and 5 summarize emission rates for the study domain.

3 Atmospheric Modeling

To study the impact of household emissions on ambient air pollution, we simulated two emission scenarios each for three time periods which coincide with available INCLEN observation data (Tables 6 and 7). A “total” emission scenario represents the overall atmospheric environment by including emissions from all source-sectors in the inventory. A “non-residential” emission scenario represents zeroing-out or “turning-off” all household emissions. By considering these scenarios independently, we can isolate the effect of the residential sector on the ambient atmosphere. Each scenario was simulated over a region in northern India (Figure 1) for those periods when measurements were carried out in the region of interest. Figure 1 shows the 600 km by 600 km domain with 4 km grid resolution. The domain is centered over the Palwal District and the SOMAARTH DDESS and includes New Delhi and portions of surrounding states.

Simulation of regional air quality was carried out using the U.S. Environmental Protection Agency Community Multiscale Air Quality modeling system (CMAQ), version 5.2 (Appel et al., 2017; US EPA, 2017). CMAQ is a three-dimensional chemical transport model (CTM) that predicts the dynamic concentrations of airborne species. CMAQ includes modules of radiative processes, aerosol microphysics, cloud processes, wet and dry deposition, and atmospheric transport. Required input to the model includes emissions inventories, initial and boundary conditions, and meteorological fields. The domain-specific, gridded emissions inventory provides hourly-resolved total emission rates for each species (not differentiated by source) by cell, timestep, and vertical layer. Initial conditions (ICs) and boundary conditions (BCs) are necessary to define the atmospheric chemical concentrations in the domain at the first time step and at the domain edges, respectively. Simulations operating with nested domains require two groups of initial conditions and boundary conditions. The present study uses the global chemical transport model GEOS-Chem v11-02c (acmg.seas.harvard.edu/geos/index.html) to generate concentrations on the boundary of the computational domain and CMAQ to produce initial and boundary conditions for the inner parent domain and nested domain, respectively. Meteorological conditions (including temperature, relative humidity, wind speed and direction and land use and terrain data) drive the atmospheric processes represented in CMAQ. The Weather Research and Forecasting modeling system (WRF) – Advanced Research WRF (WRF-ARW, version 3.6.1), was used to simulate the meteorological input for CMAQ (Skamarock et al., 2008).

3.1 GEOS-Chem

We used GEOS-Chem v11-02c, a global chemical transport model driven by assimilated meteorological observations from the NASA Goddard Earth Observing System -- Fast Processing (GEOS-FP) of the Global Modeling and Assimilation Office (GMAO), to simulate the boundary conditions for the CMAQ modeling. Simulations are performed at 2˚x2.5˚ horizontal resolution with 72 vertical layers, including both the full tropospheric chemistry with complex SOA formation (Marais et al., 2016) and UCX stratospheric chemistry (Eastham et al., 2014). Emissions used the standard HEMCO configuration (Keller et al., 2014), including EDGAR v4.2 anthropogenic emissions (http://edgar.jrc.ec.europa.eu/overview.php?v=42), biogenic emissions from the MEGAN v2.1 inventory (Guenther et al., 2012), and GFED biomass burning emissions (http://www.globalfiredata.org). Simulations were run for 1 year, after which hourly time series diagnostics were compiled for the CMAQ modeling period. Using the PseudoNetCDF processor, we remapped a subset of the 616 GEOS-Chem-produced species to CMAQ species (https://github.com/barronh/pseudonetcdf). The resulting ICs and BCs include 119 gas- and particle-phase species, 80 adapted from GEOS-Chem and the remaining 39 (including OH, HO_2, ROOH, oligomerized secondary aerosols, coarse aerosol, and aerosol number concentration distributions) from the CMAQ default initial and boundary conditions data (which were developed to represent typical clean-air pollutant concentrations in the United States).

3.2 Weather Research and Forecasting (WRF) Model

Three monthly WRF version 3.6.1 simulations were conducted in the absence of nudging or data assimilation. The large-scale forcing to generate initial and boundary meteorological fields is adopted from the latest version of the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA5 released in January 2019. These reanalysis data are on a 31 km grid and resolve the atmosphere using 137 levels from the surface to a height of 80 km. WRF simulations were performed with 4 km horizontal resolution and 24 vertical layers (the lowest layer of about 50 m depth), consistent with the setup of the CMAQ model. No cumulus parameterization was used in the simulations. Meteorological outputs from WRF were prepared as inputs to CMAQ by the Meteorology-Chemistry Interface Processor (MCIP) version 4.4 (Otte et al., 2010).

3.3 Community Multiscale Air Quality (CMAQ) Modeling System

Within the chemical transport portion of CMAQ, there are two primary components: a gas-phase chemistry module and an aerosol chemistry, gas-to-particle conversion module. The present study employs a CMAQ-adapted gas-phase chemical mechanism, CB6R3 (derived from the Carbon Bond Mechanism 06) (Yarwood et al., 2010), and the aerosol-phase mechanism, AERO6, which define the gas-phase and aerosol-phase chemical resolution. The present study considers 70 non-methane hydrocarbon (NMHC) compounds lumped into 12 groups of volatile organic compounds (VOCs). The emissions inventory provides emission rates for 28 chemical species, including 18 gas-phase species and 10 particle-phase species. The CB6R3 adaptation describes atmospheric oxidant chemistry with 127 gas-phase species and 220 gas-phase reactions, including chlorine and heterogenous reactions. The CMAQ aerosol module (AERO6) describes aerosol chemistry and gas-to-particle conversion with 12 traditional SOA precursor classes, and 10 semi-volatile primary organic aerosol (POA) precursor reactions. The majority of the gas-phase organic species are apportioned to lumped groups by their carbon bond characteristics, such as single bonds, double bonds, ring structure, and number of carbons. Some organic compounds are apportioned based on reactivity, and others, like isoprene, ethene, and formaldehyde, are treated explicitly.

The secondary organic aerosol module, AERO6, developed specifically for CMAQ, interfaces with the gas-phase mechanism, predicts microphysical processes of emission, condensation, evaporation, coagulation, new particle formation, and chemistry, and produces a particle size distribution comprising the sum of the Aitken, Accumulation, and Coarse log-normal modes (Figure 4). AERO6 predicts the formation of SOA from anthropogenic and biogenic volatile organic compound (VOC) precursors (properties of which are shown in Table 8), as well as semi-volatile POA and cloud processes. CB6R3 accounts for the oxidation of the first-generation products of the anthropogenic lumped VOCs: high-yield aromatics, low-yield aromatics, benzene, PAHs, and long-chain alkanes (Pye and Pouliot, 2012).

In addition to SOA formation from traditional precursors, CMAQv5.2 accounts for the semi-volatile partitioning and gas-phase aging of POA using the volatility basis set (VBS) framework independently from the rest of AERO6 (Murphy et. al., 2017). The module distributes directly emitted POA (as the sum of primary organic carbon, POC, and noncarbon organic matter, NCOM) from the emissions inventory input into five new emitted species grouped by volatility: LVPO1, SVPO1, SVPO2, and SVPO3, and IVPO1 (where LV is low volatility, SV is semi-volatile, IV is intermediate volatility, and PO is primary organic). POA is apportioned to these lumped vapor species using an emission fraction and are oxidized in CB6R3 by OH to LVOO1, LVOO2, SVOO1, SVOO2, and SVOO3 (where OO denotes oxidized organics) with stoichiometric coefficients derived from the 2D-VBS model. AERO6 then partitions the semi-volatile primary organics and their oxidation products to the aerosol phase (Figure 4). Thus, the treatment of POA as semi-volatile products leads to an additional twenty species, a particle- and vapor-phase component for each primary organic and oxidation product (Murphy et al., 2017).

Emissions inventory modifications were required to match the most recent aerosol module, AERO6, in the CMAQ model. Initially, the lumped emissions of PAR (a lumped VOC group characterized by alkanes) and XYL (a lumped VOC group characterized by xylene) derived from grouping specific NMHCs, calculated using the University of Illinois estimation and the Fleming et al. (2018a) emission factors, accounted for characteristics of naphthalene (NAPH) and SOA-producing alkanes (SOAALK), which are not individually described by any of the sources used to construct the inventory. Moreover, only a subset of VOCs in the plume could be measured. However, CMAQv5.2 simulations incorporate a surrogate species, potential secondary organic aerosol from combustion emissions (pcSOA), to address sources of missing SOA, including unspeciated emissions of semivolatile and intermediate volatility organic compounds. AERO6 predicts the formation of SOA from NAPH and SOAALK independently as well as from XYL and PAR; these secondary aerosol precursor emission rates are calculated with:

$$\mathrm{XYLMN} = 0.998 * \mathrm{XYL} \tag{3}$$

$$\mathrm{NAPH} = 0.002 * \mathrm{XYL} \tag{4}$$

$$\mathrm{PAR_{CMAQ}} = \mathrm{PAR_{calculated}} - 0.00001 * \mathrm{NAPH} \tag{5}$$

$$\mathrm{SOAALK} = 0.108 * \mathrm{PAR_{CMAQ}} \tag{6}$$

where XYLMN, NAPH, PARCMAQ, and SOAALK are the new inventory species (Pye and Pouliot, 2012). SOA-producing alkanes are treated separately in AERO6.

4 Surface Observational Data

Gas-phase air quality data analyzed in the present study come from the Central Pollution Control Board (CPCB) of the Ministry of Environment, Forest & Climate Change, Government of India at two sites in New Delhi (one in the west, and one in the south). Particle-phase data analyzed come from the SOMAARTH Demographic, Development, and Environmental Surveillance Site (Mukhopadhyay et al., 2012; Pillarisetti et al., 2014; Balakrishnan et al., 2015) managed by the International Clinical Epidemiological Network (INCLEN). Palwal District has a population of ~ 1 million over an area of 1400 km^2. In this district, ~39% of households utilize wood burning as their primary cooking fuel, with dung (~25%) and crop residues (~7%) (Census of India, 2011). Specific sites studied are the SOMAARTH headquarters (HQ) in Aurangabad (15 km south of Palwal) and the village of Bajada Pahari (8 km northwest of SOMAARTH HQ). Ambient measurement sites are shown in Fig. 1, and Table 6 details available data for each location. We used meteorological data (hourly surface temperature and near-surface wind speed and direction) from INCLEN and CPCB at the two rural and two urban sites, respectively, to evaluate the WRF simulations performance.

5 Simulation Results

5.1 WRF Evaluation

We evaluated WRF simulated meteorology against the available surface observations at different sites during the same periods. Figure 5 shows that there is generally good agreement of surface temperature between WRF and observations for all three months. The surface wind direction is found consistent between model and observations for each site and each month (Table 9). The simulated near-surface wind speeds are overestimated in WRF, with an averaged mean-bias (MB) of about +1.5 m/s. Such a bias is partly a result of the difference in the definition of "near-surface" between the model and observations.

5.2 Particulate Matter

Figures 6–9 show measured and predicted total $PM_{2.5}$ and the average diurnal profile at each site for the periods with available measurements. The diurnal profile in these figures includes that of both emission scenarios: the total scenario with all emissions and the non-residential scenario with zeroed-out residential sector. The simulations capture the general trend well and produce significant diurnal profiles (Table 10). Rural sites show typical $PM_{2.5}$ levels are predicted between 50 µg m^{-3} and 125 µg m^{-3} in December and 25 µg m^{-3} and 75 µg m^{-3} in September months (Figures 6 and 7). On the other hand, typical values at urban sites range from 100 µg m^{-3} to 300 µg m^{-3} in December and 50 µg m^{-3} to 125 µg m^{-3} in September months (Figures 8 and 9). Observations and predictions show higher $PM_{2.5}$ levels in December than September, owing to frequent temperature inversions in winter and shallower planetary boundary layers. Two daily peaks and lows of $PM_{2.5}$ compare with ambient observations at Bajada Pahari December 2015 and September 2016, SOMAARTH HQ September 2015 and 2016, West New Delhi December 2015, and South New Delhi December and September 2015. Average daily $PM_{2.5}$ levels regularly exceed the 24-hour Indian standard of 60 µg m^{-3} in each month in both rural and urban locations, surpassing even double the standard in the village of Bajada Pahari during mealtimes in December. Afternoon minima tend to be underestimated in September 2015 and December 2015. Diurnal trends of $PM_{2.5}$ were weaker in September 2016 than the other months, with lower predictions but overestimated minima. Urban sites show greater overestimation than rural sites. This is likely due in part to the granularity of the primary emissions inventory datasets. The non-residential sector was prepared from data with a native resolution of 36 km, while the residential sector used data with ~1 km resolution. Underpredictions of peak

$PM_{2.5}$ concentrations in September could also result because the emission inventory does not account for day-to-day variations, especially in the agricultural burning sector in which emissions can change significantly on a daily basis. Observed and predicted $PM_{2.5}$ levels in New Delhi can exceed 300 μg m^{-3}, especially in winter. In this highly populated urban environment, particulate matter levels are more than double those reported in the nearby rural areas. The employed emissions inventory specifies particulate matter surface emissions, which surpass those of Bajada Pahari and SOMAARTH HQ more than 30-fold (Table 5). Biogenic emissions are predicted to be of little importance, accounting for less than 10% on average of total $PM_{2.5}$ concentrations for most stations and months (Table 10).

Figure 10 shows CMAQ predictions of secondary organic $PM_{2.5}$ (SOA). Like $PM_{2.5}$, SOA is typically predicted to be higher in New Delhi than in the rural sites, due to higher $PM_{2.5}$ and precursor VOC emissions and ambient concentrations in urban environments (Tables 5 and 6). Higher levels are similarly attained in December than in September due to longer residence times and more aging during winter. SOA has high day-to-day variability. Values range from below 20 μg m^{-3} to over 200 μg m^{-3} in December, with average peaks up to 55 μg m^{-3} at the rural sites. September months predict lower SOA, ranging from 10 μg m^{-3} to 130 μg m^{-3}. Diurnal average SOA maxima in December for the rural stations is nearly double that of September 2016, which can be attributed to temperature inversions and a shallower planetary boundary layer in winter.

The significance of household emissions on outdoor $PM_{2.5}$ concentrations is demonstrated by the diurnal profiles in Figure 11. The top row of plots shows the predicted contribution of the residential sector to anthropogenic $PM_{2.5}$, while the middle row of plots describes the predicted contribution of the residential sector to secondary organic $PM_{2.5}$, as in Equations 7 and 8 respectively:

$$\frac{\text{Residential Anthropogenic PM}_{2.5}}{\text{Total Anthropogenic PM}_{2.5}} \tag{7}$$

$$\frac{\text{Residential SOA}}{\text{Total SOA}} \tag{8}$$

The bottom row of plots shows the predicted SOA portion of residential $PM_{2.5}$, as

$$\frac{\text{Residential SOA}}{\text{Residential PM}_{2.5}} \tag{9}$$

where residential PM is calculated as the difference in predictions from the non-residential and total emission scenario and averaged over simulation durations (Table 7). Importance of household emissions to ambient PM is strongly correlated with mealtimes. Predicted maximum contributions to anthropogenic $PM_{2.5}$ in Bajada Pahari and SOMAARTH HQ are about double that of South and West New Delhi for each month. Household energy-use is estimated to account for up to 27% of anthropogenic $PM_{2.5}$ (at SOMAARTH HQ during September 2016), remaining consistently above 10% for each rural site during all months. Similar behavior is predicted for SOA (middle plots of Fig. 11). An estimated 15% to 34% of secondary organic matter is attributable to residential emissions in September and 2016. Again, the impact is smaller in West and South New Delhi (up to 19% and 21%, respectively in September 2016), where there are greater emissions of SOA precursors from other sectors. The diurnal profile of the contribution to SOA is subdued for all sites in December, suggesting that SOA generation is less efficient in winter when radiation and temperatures are lower. Aging of VOCs is captured by the phase shift of the impact on SOA daily trend, where peaks consistently occur an hour after the residential sector shows greatest importance to anthropogenic $PM_{2.5}$.

At each measurement site during all months, SOA is predicted to make up more than 40% of $PM_{2.5}$ produced by the residential sector on average (bottom row of plots of Fig. 11). SOA is least significant to residential $PM_{2.5}$ in the first half of mealtimes (~20% during breakfast and ~40% during dinner) at rural sites, when primary particulate matter is largest. Aging of precursor VOCs from cooking emissions, paired with maximum incoming radiation, lead to maximum $\frac{\text{Residential SOA}}{\text{Residential PM}_{2.5}}$ values in early afternoon, when SOA accounts for more than 75% of residential $PM_{2.5}$ at both rural and urban sites during each simulated month.

The fractional contribution of total SOA to total $PM_{2.5}$ is shown in Fig. 12. While concentrations of SOA depend significantly on the site and time period, their contribution to total $PM_{2.5}$ shows little variation. At all stations, SOA is predicted to make up to 55% of $PM_{2.5}$ in September months and to be most significant around midday. However, diurnal variation of the significance of SOA is greater in New Delhi than in Bajada Pahari or SOMAARTH HQ, owing to greater diversity of energy-use activities and emissions characteristics in the urban environment.

5.3 Ozone

The 8-hour India Central Pollution Control Board (CPCB) standard for ozone is 100 μg m^{-3} for an 8-hour average. In the alternative unit of ozone mixing ratio, a mass concentration of ozone of 100 μg m^{-3} at a temperature of 298 K at the Earth's surface equates to a mixing ratio of 51 parts-per-billion (ppb). A number of atmospheric modeling studies of ozone over India exist (Kumar et al., 2010; Chatani et al., 2014; Sharma et al., 2016).

Sharma et al. (2016) carried out baseline CMAQ simulations for 2010 and compared ozone predictions with measurements at six monitoring locations in India (Thumba, Gadanki, Pune, Anantpur, Mt. Abu, and Nainital). Also carried out were sensitivity simulations in which each emissions sector (transport, domestic, industrial, power, etc.) was systematically set to zero. The domestic sector was predicted to contribute ~60% of the non-methane volatile organic carbon emissions, followed by 12% from transportation and 20% from solvent use and the oil and gas sector. The overall NO_x-to-VOC mass ratio in the region simulated by Sharma et al. (2016) was 0.55. This exceptionally low NO_x-to-VOC ratio was attributed, in part, to the widespread use of biomass fuel for cooking (leading to high VOC emissions), coupled with relatively low NO_x emissions. (Although vehicle emissions are high in urban areas, overall vehicle ownership is relatively low at the national level. In addition, Euro equivalent norms have led to reduction of NO_x emissions.) Predicted O_3 levels at the six observation sites tended to exceed measured values, with the ratio of predicted to observed annual average O_3 being in the range of 1.04–1.37 at the six locations. Moreover, the overall low NO_x-to-VOC ratios in India lead to NO_x-sensitive O_3 formation conditions. Based on emissions inventories, the overall anthropogenic NMVOC/NO_x mass emissions ratio in India in 2010 as computed by Sharma et al. (2016) was 1.82. Considering only ground-level sources, the ratio increases to 3.68.

Ozone surface measurements and predicted mass concentrations based on the CMAQ 4 km resolution simulations at two sites in New Delhi over the periods 9/7/2015 – 9/29/2015, 12/7/2015 – 12/30/2015, and 9/7/2016 – 9/29/2016 in the present study are shown in the three panels in Fig. 13. The predicted O_3 concentrations are reproduced well at the West New Delhi and South New Delhi stations, especially in September (Table 10). However, when NO concentrations are higher due to meteorological inversion conditions, ozone concentrations are underestimated, as local $NO+O_3$ titration reactions near the monitoring site are not resolved. The performance of the model improves in prediction of higher values of ozone (as in the case of September), which are of greater importance for assessing exposures. High ozone concentrations in September are quite well reproduced by the model. This shows that, on the larger scale, the model captures photochemistry quite well; however, micro-scale titration is not well represented due to the limitations of inventory resolution. This would require further enhancement of emission inventories at even higher resolution. The results of ozone simulations in the present study are generally consistent with those of previous simulations over India. For example, also using WRF-CMAQ, Kota et al. (2018) showed that the relative bias in ozone simulation ranges from −30% to +50% in major cities of India. In South New Delhi, the bias in O_3 predictions in the present study lies between -2.67 and +7.01 µg m^{-3}, as compared to the observations of 29.28 to 62.76 µg m^{-3}.

6 Conclusions

Air quality in India is determined by a mixture of industrial and motor vehicle emissions, and anthropogenic fuel combustion, that includes residential burning of biomass for household uses, such as cooking. Average daily $PM_{2.5}$ levels frequently exceed the 24-hour standard of 60 µg m^{-3} and can exceed 200 µg m^{-3}, even in rural areas. $PM_{2.5}$ is a mixture of directly-emitted particulate matter and that formed by the atmospheric conversion of volatile organic compounds to secondary organic aerosol. Here, we assess the extent to which observed O_3 and $PM_{2.5}$ levels in India can be predicted using state-of-the-science emissions inventories and atmospheric chemical transport models. We have focused on the 308 sq km of the SOMAARTH Demographic, Development, and Environmental Surveillance Site (DDESS) in the Palwal District of Haryana, India.

Atmospheric simulation of particulate matter levels over a complex region like India tends to be demanding, owing to the combination of a wide range of primary particulate emissions and the presence of secondary organic matter from atmospheric gas-phase reactions generating low-volatility gas-phase products that condense into the particulate phase, forming secondary organic aerosol (SOA). Consequently, the main focus of the present work has been the evaluation of the extent to which ambient particulate matter levels over the current region of India can be predicted. Simulations capture the general trend of observed daily peaks and lows of particulate matter, with $PM_{2.5}$ reaching values as high as 250 µg m^{-3}. Secondary organic matter accounts for 10% to 55% of total $PM_{2.5}$ mass on average. In India, over 50% of households report use of wood, crop residues, or dung as cooking fuel; such fuels produce significant gas- and particle-phase emissions. We evaluated the fractional impact of the residential sector emissions on the formation of secondary organic aerosol, as a function of time of day, for New Delhi, SOMAARTH HQ, and Bajada Pahari. The predicted fractional contribution of residential sector emissions to secondary organic

$PM_{2.5}$ in Bajada Pahari and SOMAARTH HQ reaches values as high as 34% and, moreover, displays a distinct diurnal profile, with maxima corresponding to the morning and evening mealtimes. In both rural and urban areas, SOA is predicted to account for more than 40% of residential $PM_{2.5}$, reaching up to 80% in early afternoon in September months.

Simulations of ozone levels in New Delhi reported here are largely in agreement with ambient monitoring data, although the simulations fail to capture several one- to two-day ozone episodes that exceed predictions by a factor of two or more. The overall agreement between observed and predicted O_3 levels, also demonstrated in the study of Sharma et al. (2016), suggests that gas-phase atmospheric chemistry over India is reasonably well understood. While ozone and particulate matter were simulated for September and December months, we employed a single emissions inventory, regardless of season. Thus, the inventory does not capture December-specific characteristics, including heating combustion. Furthermore, information regarding household solvent use, emissions profiles by fuel type, and speciation of certain emissions (such as semi volatile organic compounds and intermediate volatility organic compounds) is lacking. Variation in the resolution of specific input data additionally contributes to uncertainty.

Air quality studies such as the present one provide a quantification of the elements of atmospheric composition in India, especially that owing to household sources. The importance of replacing traditional household combustion devices with modern technology is evident in studies such as the present one.

Author Contributions

BR carried out the simulations, and wrote the paper; RZ, YW and KB assisted with the simulations; AP carried out field measurements; SS, SK, TB, NL, BO LX, and VG helped formulate the emissions inventory; LF, RW, SM, and DB designed and carried out measurements; SN, RE, AY, and NA performed data analysis; KS designed the research; JS designed the research and wrote the paper.

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Table 1. Residential Emissions Inventory Sources by Species

	CMAQ Required Species[1]	**Source**		**Solely Emitted by Residential Sector**
Gas	NO	T. Bond (University of Illinois) NO_x using Sharma et al. (2015) $NO:NO_2 = 10:1$		No
	NO_2			No
	SO_2	Sharma et al. (2015)		No
	NH_3	Sharma et al. (2015), assumed to be negligible		
	CO	T. Bond (University of Illinois)		No
NMHC	ALD_2	Speciation from T. Bond (University of Illinois) NMHC using Fleming et al. (2018a,b) emission factors		No
	ALD_X			Yes
	ETH			No
	ETHA			No
	ETOH			No
	FORM			No
	MEOH			No
	OLE			No
	$PAR_{calculated}$[3]			No
	TOL			No
	XYL[3]			No
CMAQ AERO6 Species	**ISOP[2]**	All-sector total ISOP emission from GEOS-Chem daily average and subtracted non-residential ISOP emission from Sharma et al. (2015)		No
	TERP[2]	Assumed to be negligible		
	XYLMN	$XYLMN = 0.998 * XYL$	Pye and Pouliot (2012)	No
	NAPH	$NAPH = 0.002 * XYL$		No
	PAR_{CMAQ}	$PAR_{CMAQ} = PAR_{calculated} - 0.00001 * NAPH$		No
	SOAALK	$SOAALK = 0.108*PAR_{CMAQ}$		No
PM	**P_{EC}**	T. Bond (University of Illinois)		No
	P_{OC}			No
	P_{NA}	Speciation of $PM_{2.5}$ from T. Bond (University of Illinois) using Jayarathne et al. (2018) mass percentage		Yes
	P_{CL}			Yes
	P_K			Yes
	P_{NH4}			Yes
	P_{NO3}			No
	P_{SO4}			No
	PM_{OTHR}	$PM_{OTHR} = PM_{2.5} - (P_{EC}+ P_{OC}+P_{NA}+P_{NH4}+P_K+P_{CL}+P_{NO3}+P_{SO4})$		No
	PM_C	Sharma et al. (2015)		No
	P_{NCOM}	Unknown, assumed to be 0		
	P_{H2O}			
	P_{AL}	Assumed to be negligible		
	P_{CA}			
	P_{FE}			
	P_{MG}			
	P_{MN}			
	P_{SI}			
	P_{TI}			

[1] Bolded species contribute to SOA production via the AERO6 module. [2] Total isoprene and terpene emissions from all sectors are taken from GEOS-Chem and were included only in the O_3 simulations. [3] $PAR_{calculated}$ and XYL are excluded from CMAQ and replaced with PAR_{CMAQ}, XYLMN, NAPH, and SOAALK.

Table 2. Residential $PM_{2.5}$ and NMHC Emissions Speciation

Emitted Species	**Fuel-Specific Data**	**Use**
$PM_{2.5}$ (Bond et al., 2004)	wood, dung, agricultural residue, LPG	Total $PM_{2.5}$ emission rate distributed by wood, dung, and agricultural residue. LPG emissions assumed negligible.
Speciated $PM_{2.5}$ (Jayarathne et al., 2018)	wood, wood/dung mix	Average profile of wood and wood/dung mix applied to all fuel type emissions.
NMHC (Bond et al., 2004)	wood, dung, agricultural residue, LPG	Total $PM_{2.5}$ emission rate distributed by wood, dung, and agricultural residue. LPG emissions assumed negligible.
Speciated HCs (Fleming et al., 2018a,b)	wood, dung	One profile applied to each cell according to which fuel type dominates emissions in that cell. Where agricultural residue dominates, wood profile is assumed.

Table 3. $PM_{2.5}$ Speciation by Fuel Type

Emitted Species[1]	**% Mass of Total Emitted $PM_{2.5}$**		
	Wood[2]	*Wood/Dung*[2]	*Average Employed*[3]
P_{EC}	14	5.10	9.55
P_{OC}	52	61	56.50
P_{NA}	0.05	0.39	0.22
P_{CL}	3.20	8.58	5.89
P_{K}	1.78	0.52	1.15
P_{NH4}	1.12	4.46	2.79
P_{NO3}	0.42	0.21	0.32
P_{SO4}	0.33	0.46	0.40
PM_{OTHR}	27.10	19.29	23.19

[1] Total $PM_{2.5}$ mass emission rates from residential combustion were estimated and distributed by fuel type (wood, dung, or agricultural residue) by University of Illinois. [2] Emitted $PM_{2.5}$ weight percent reported by Jayarathne et al. (2018). [3] An average profile applied to all cells, indiscriminate of fuel type.

Table 4. Particulate Matter Surface Emissions over Study Domain

Species		**Emission Rate**	**% Emitted by Residential Sector**
Particulate Matter (kg/day)	P_{OC}	1.48×10^6	30.78
	P_{EC}	7.18×10^5	15.89
	P_{CL}	1.69×10^3	100
	P_{K}	4.61×10^3	100
	P_{NA}	2.46×10^4	10.07
	P_{NH4}	2.11×10^5	1.47
	P_{NO3}	6.51×10^5	27.90
	P_{SO4}	1.18×10^6	61.45
	PM_{C}	9.00×10^3	100
	PM_{OTHR}	2.18×10^4	100
SOA Precursor VOCs (mol/day)	*NAPH*	6.82×10^3	2.72
	SOAALK	3.75×10^6	34.54
	TOL	1.54×10^6	27.21
	XYLMN	3.40×10^6	2.72

Table 5. Mealtime[1] Particulate Matter Surface Emissions over Corresponding 16 km^2 Grid Cell

		Bajada Pahari		SOMAARTH HQ		New Delhi	
	Species	*Total*	*% Residential*	*Total*	*% Residential*	*Total*	*% Residential*
Particulate Matter (kg/day)	P_{OC}	35.17	67.13	36.04	100	609.73	5.70
	P_{EC}	10.22	33.23	6.02	100	346.84	2.21
	P_{CL}	2.19	100	3.40	100	3.40	100
	P_{K}	0.43	100	0.66	100	0.66	100
	P_{NA}	0.08	100	0.12	100	0.12	100
	P_{NH4}	1.037	100	1.61	100	1.61	100
	P_{NO3}	0.37	32.01	0.18	100	12.59	1.45
	P_{SO4}	2.49	5.90	0.23	100	116.80	0.20
	PM_{C}	63.99	91.94	72.56	100	275.99	7.12
	PM_{OTHR}	13.92	61.94	13.37	100	276.91	4.83
SOA Precursor VOCs (mol/day)	*NAPH*	0.11	6.50	0.03	59.56	3.78	0.65
	SOAALK	112.31	50.62	113.20	77.95	1696.26	11.14
	TOL	43.88	42.28	39.38	71.05	750.00	1.04
	XYLMN	56.47	6.50	13.03	59.56	1886.15	0.65

[1]Mealtimes are assumed to be 4 am – 10 am and 4 pm – 8pm (local).

Table 6. Ambient Observation Data Availability

Location (Grid Cell)	**PM$_{2.5}$**	**O_3**
Bajada Pahari[1] **(74,74)**	12/20/15 – 12/31/15 9/19/16 – 9/30/16	n/a
SOMAARTH HQ[1] **(75,74)**	9/22/15 – 9/27/15 9/23/16 – 9/30/16	n/a
West New Delhi[2] **(71,91)**	9/7/15 – 9/30/15 12/7/15 – 12/31/15 9/7/16 – 9/30/16	9/7/15 – 9/30/15 12/7/15 – 12/31/15
South New Delhi[2] **(71,89)**	9/7/15 – 9/30/15 12/7/15 – 12/31/15 9/7/16 – 9/30/16	9/7/15 – 9/30/15 12/7/15 – 12/31/15 9/7/16 – 9/30/16

[1] Data from the International Epidemiological Clinical Network. Observations at Bajada Pahari are the average of two monitoring locations that coincide within the same grid cell. [2] Data from the Central Pollution Control Board of India at New Delhi Punjabi Bagh monitoring station.

Table 7. Simulation Durations

CMAQ[1]	09/07/15 - 09/30/15	12/07/15 - 12/31/15	09/07/16 - 09/30/16
WRF[2] (Meteorology)	09/02/15 - 09/30/15	12/02/15 - 12/31/15	09/02/16 - 09/30/16
GEOS-Chem[3] (Boundary Conditions)	09/07/15 - 09/30/15	12/07/15 - 12/31/15	09/07/16 - 09/30/16

[1] Five days prior to date shown were run and omitted from analysis as spinup. [2] One day prior to date shown was run and omitted from analysis as spinup. [3] GEOS-Chem was run for one year before extracting atmospheric diagnostics.

Table 8. Properties of anthropogenic traditional semi-volatile SOA precursors in CMAQv5.2

SOA species	Precursor	Oxidants	Semi-volatile	α (mass-based)	C* (μg/m^3)	ΔH$_{vap}$ (kJ/mol)	# of C	Molecular weight (g/mol)	OM/OC
AALK1	long-chain alkanes	OH	SV_ALK1	0.0334	0.15	53.0	12	168	1.17
AALK2	long-chain alkanes	OH	SV_ALK2	0.2164	51.9	53.0	12	168	1.17
AXYL1	XYLMN	OH,NO	SV_XYL1	0.0310	1.3	32.0	8	192	2.0
AXYL2	XYLMN	OH,NO	SV_XYL2	0.0900	34.5	32.0	8	192	2.0
AXYL3	XYLMN	OH,HO$_2$	nonvolatile	0.36	NA	NA	NA	192	2.0
ATOL1	TOL	OH,NO	SV_TOL1	0.0310	2.3	18.0	7	168	2.0
ATOL2	TOL	OH,NO	SV_TOL2	0.0900	21.3	18.0	7	168	2.0
ATOL3	TOL	OH,HO$_2$	nonvolatile	0.30	NA	NA	NA	168	2.0
ABNZ1	benzene	OH,NO	SV_BNZ1	0.0720	0.30	18	6	144	2.0
ABNZ2	benzene	OH,NO	SV_BNZ2	0.8880	111	18	6	144	2.0
ABNZ3	benzene	OH,HO$_2$	nonvolatile	0.37	NA	NA	NA	144	2.0
APAH1	naphthalene	OH,NO	SV_PAH1	0.2100	1.66	18	10	243	2.03
APAH2	naphthalene	OH,NO	SV_PAH2	1.0700	265	18	10	243	2.03
APAH3	naphthalene	OH,HO$_2$	nonvolatile	0.73	NA	NA	NA	243	2.03

The semi-volatile reaction products of “long alkanes” (SV_ALK1 and SV_ALK2) are parameterized by Presto et al. (2010). Values for “low-yield aromatics” products (SV_XYL1 and SV_XYL2) are based on xylene, with the enthalpy of vaporization (ΔH$_{vap}$) from studies of m-xylene and 1,3,5-trimethylbenzene. ΔH$_{vap}$ for products of “high-yield aromatics” (SV_TOL1 and SV_TOL2) are based on the higher end of the range for toluene. The products of benzene (SV_BNZ1 and SV_BNZ2) assume the same value for ΔH$_{vap}$. All semi-volatile aromatic products are assigned stoichiometric yield (α) and effective saturation concentration (C*) values from laboratory measurements by Ng et al. (2007). Remaining parameters for PAH reaction products (SV_PAH1 and SV_PAH2) are taken from Chan et al. (2009). Properties of semi-volatile primary organic aerosol precursors are given in Murphy et al. (2017).

Table 9. Quantification of WRF model biases in meteorological fields.

		Bajada Pahari			**SOMAARTH HQ**			**West New Delhi**			**South New Delhi**		
		Sep ‘15	**Dec ‘15**	**Sep ‘16**	**Sep ‘15**	**Dec ‘15**	**Sep ‘16**	**Sep ‘15**	**Dec ‘15**	**Sep ‘16**	**Sep ‘15**	**Dec ‘15**	**Sep ‘16**
Temperature (°C)	**PRE**	-	15.28 *(4.59)*	30.10 *(3.19)*	29.27 *(3.48)*	-	30.22 *(3.06)*	30.45 *(3.79)*	16.59 *(4.91)*	30.07 *(3.05)*	30.32 *(3.74)*	17.59 *(4.82)*	29.96 *(30.3)*
	OBS	-	15.62 *(4.91)*	30.86 *(5.67)*	32.15 *(4.12)*	-	33.26 *(5.31)*	32.80 *(3.60)*	19.04 *(3.66)*	31.46 *(2.33)*	28.48 *(4.30)*	12.58 *(5.52)*	29.22 *(4.22)*
	MB	-	-0.34	-0.76	-2.89	-	-3.04	-2.35	-2.45	-1.38	1.84	5.02	0.74
	ME	-	1.60	3.08	2.92	-	3.07	3.03	2.58	1.54	2.11	5.02	2.37
	RMSE	-	2.20	3.71	3.39	-	3.99	3.58	2.99	1.88	2.50	5.33	2.75
Wind Speed (m·s^{-1})	**PRE**	-	2.91 *(1.17)*	2.31 *(1.07)*	-	-	2.01 *(0.66)*	-	-	2.57 *(1.28)*	2.80 *(1.27)*	2.72 *(1.08)*	2.74 *(1.39)*
	OBS	-	1.18 *(0.75)*	0.73 *(0.40)*	-	-	0.55 *(0.30)*	-	-	1.03 *(0.51)*	1.26 *(0.83)*	0.94 *(0.71)*	1.18 *(0.79)*
	MB	-	1.72	1.58	-	-	1.46	-	-	1.54	1.54	1.77	1.56
	ME	-	1.75	1.62	-	-	1.50	-	-	1.58	1.61	1.82	1.62
	RMSE	-	1.96	1.85	-	-	1.66	-	-	1.88	1.85	2.01	1.84
Wind Direction (°)	**PRE**	-	247 *(111)*	116 *(45)*	272 *(70)*	-	111 *(51)*	-	-	179 *(98)*	206 *(118)*	254 *(97)*	191 *(96)*
	OBS	-	259 *(57)*	102 *(41)*	255 *(58)*	-	110 *(48)*	-	-	181 *(97)*	198 *(45)*	224 *(44)*	228 *(50)*
	MB	-	0.14	14	16	-	-0.14	-	-	-6	9	35	-34
	ME	-	51	38	44	-	32.71	-	-	49	94	74	75
	RMSE	-	66	51	64	-	47.50	-	-	64	106	87	90

PRE is mean predictions; OBS is mean observations; MB is mean bias; ME is mean error; and RMSE is root mean square error. Standard deviation of predictions and observations are noted in parentheses.

Table 10. CMAQ Model Performance and Summary Statistics.

		Bajada Pahari			**SOMAARTH HQ**			**West New Delhi**			**South New Delhi**		
		Dec ‘15	**Sep ‘15**	**Sep ‘16**	**Dec ‘15**	**Sep ‘15**	**Sep ‘16**	**Dec ‘15**	**Sep ‘15**	**Sep ‘16**	**Dec ‘15**	**Sep ‘15**	**Sep ‘16**
PM$_{2.5}$	**PRE**	133.49	54.83	59.22	131.80	32.16	63.66	212.29	101.71	106.44	191.35	92.68	92.85
		(40.66)	*(21.24)*	*(9.89)*	*(42.81)*	*(15.99)*	*(11.24)*	*(75.55)*	*(41.49)*	*(28.58)*	*(61.03)*	*(39.46)*	*(24.37)*
	OBS	136.01	-	35.55	-	75.83	58.03	120.49	81.53	-	254.15	70.24	70.97
		(28.35)	-	*(13.76)*	-	*(37.16)*	*(35.19)*	*(29.92)*	*(12.72)*	-	*(70.89)*	*(13.04)*	*(18.72)*
	MB	-2.52	-	23.67	-	-43.67	5.64	91.80	20.19	-	-62.81	22.44	21.88
	ME	35.20	-	24.66	-	43.67	25.04	91.93	41.02	-	67.67	26.42	25.71
	RMSE	40.23	-	26.35	-	56.23	27.71	115.76	48.60	-	81.02	37.50	35.37
O$_3$	**PRE**	72.76	80.72	47.24	71.83	80.75	47.22	32.59	57.14	31.66	40.90	62.76	36.29
		(39.47	*(3.87)*	*(17.56)*	*(39.99)*	*(34.06)*	*(17.60)*	*(41.34)*	*(53.36)*	*(30.16)*	*(44.87)*	*(53.52)*	*(29.89)*
	OBS	-	-	-	-	-	-	21.74	71.09	-	43.57	59.47	29.28
								(8.05)	*(42.41)*		*(37.07)*	*(36.30)*	*(20.27)*
	MB	-	-	-	-	-	-	10.93	-13.95	-	-2.67	3.29	7.01
	ME	-	-	-	-	-	-	16.83	18.74	-	12.62	24.72	19.29
	RMSE	-	-	-	-	-	-	22.96	22.10	-	14.08	27.64	23.31
SOA	**PRE**	44.60	17.89	23.30	44.81	18.06	22.95	44.22	23.76	33.28	43.95	22.44	31.78
		(7.76)	*(2.40)*	*(3.96)*	*(7.59)*	*(2.34)*	*(3.77)*	*(3.76)*	*(4.74)*	*(8.80)*	*(3.82)*	*(4.11)*	*(7.84)*
F$_{bio}$	**PRE**	0.09	0.18	0.08	0.09	0.18	0.08	0.04	0.06	0.04	0.04	0.03	0.05
		(0.03)	*(0.10)*	*(0.02)*	*(0.03)*	*(0.11)*	*(0.02)*	*(0.01)*	*(0.01)*	*(0.01)*	*(0.01)*	*(0.01)*	*(0.01)*
F$_{SOA,res}$	**PRE**	0.15	0.24	0.26	0.15	0.24	0.26	0.13	0.15	0.16	0.13	0.16	0.16
		(0.02)	*(0.05)*	*(0.04)*	*(0.02)*	*(0.05)*	*(0.04)*	*(0.03)*	*(0.02)*	*(0.03)*	*(0.03)*	*(0.02)*	*(0.03)*
F$_{an,res}$	**PRE**	0.12	0.16	0.20	0.12	0.17	0.20	0.07	0.08	0.09	0.07	0.08	0.10
		(0.03)	*(0.04)*	*(0.03)*	*(0.03)*	*(0.04)*	*(0.04)*	*(0.02)*	*(0.03)*	*(0.02)*	*(0.02)*	*(0.03)*	*(0.02)*
F$_{res,SOA}$	**PRE**	0.48	0.51	0.52	0.47	0.50	0.52	0.43	0.51	0.55	0.45	0.53	0.54
		(0.16)	*(0.20)*	*(0.18)*	*(0.16)*	*(0.21)*	*(0.18)*	*(0.18)*	*(0.21)*	*(0.17)*	*(0.18)*	*(0.22)*	*(0.17)*

Statistics are calculated for average diurnal profiles of predicted parameters. PM$_{2.5}$, O$_3$, and SOA are the mass concentrations in μg m^{-3} of total fine particulate matter, ozone, and secondary organic matter, respectively. F$_{bio}$ is the fraction of total PM$_{2.5}$ that is produced by biogenic emissions; F$_{SOA,res}$ is the fraction of total secondary organic matter attributable to the residential sector; F$_{an,res}$ is the fraction of total anthropogenic PM$_{2.5}$ attributable to the residential sector; and F$_{res,SOA}$ is the fraction of residential PM$_{2.5}$ attributable to SOA. PRE is mean predictions; OBS is mean observations; MB is mean bias; ME is mean error; and RMSE is root mean square error. Standard deviation of predictions and observations are noted in parentheses.

Figures

Fig. 1. Geographic area of simulation. The left panel shows the entirety of India, and the right panel shows a closeup of the model domain. The domain spans a 600 km by 600 km area with a grid resolution of 4 km (150 cells along each axis) and includes both New Delhi and SOMAARTH DDESS.

Fig. 2. Fraction of daily household emissions by quantifiable fuel-use activity. Red, green, blue, and purple indicates cooking, space heating, water heating, and lighting, respectively. This represents the fraction of activity-specific daily emissions at each hour. Each species obeys the same profile. While profiles for heating are shown, the inventory assumes temperatures too high for this activity to take effect.

Fig. 3. Fuel type assumed for speciation of household NMHC emissions. Study domain: 600 by 600 km at 4 km resolution. Red indicates cells where dung use dominated emissions and thus was assumed to be the sole fuel type used. Orange indicates cells where wood and agricultural residue use dominated emissions and was thus assumed to be the sole fuel type used.

Fig. 4. Treatment of anthropogenic SOA in CMAQv5.2. Predicted aerosol species are included in the black box. Species in white boxes are semi-volatile and species in gray boxes are nonvolatile. Blue indicates species and processes predicted by CB6R3. All other coloring indicates the AERO6 mechanism where green arrows are 2-product volatility distribution, orange arrows are particle- and vapor-phase partitioning, and purple arrows are oligomerization. In AERO6, anthropogenic and biogenic VOC emissions (lumped by category), are oxidized by OH, NO, and HO_2 and OH, O_3, NO, and NO_3 respectively, to semi-volatile products that undergo partitioning to the particle phase (Pye et al., 2015). Semi-volatile primary organic pathways in CMAQv5.2 are described by Murphy et al. (2017).

Fig. 5. Evaluation of WRF simulated meteorological fields versus ground observations.

Fig. 6. Measured and predicted $PM_{2.5}$ (left) and average diurnal cycle (right) in Bajada Pahari for 12/20/15 – 12/31/15 (top) and 09/20/16 – 09/30/16 (bottom). Here the yellow lines correspond to CMAQ predictions of the “total” (solid) and “non-residential” (dotted) simulations. The solid black line represents ambient observations. Standard deviations of the diurnal profiles for observations and predictions are indicated, respectively, by colored shading. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution.

Fig. 7. Measured and predicted $PM_{2.5}$ (left) and average diurnal cycle (right) at SOMAARTH HQ for 12/20/15 – 12/31/15 (top) and 09/20/16 – 09/30/16 (bottom). Here the green lines correspond to CMAQ predictions of the “total” (solid) and “non-residential” (dotted) simulations. The solid black line represents ambient observations. Standard deviations of the diurnal profiles for observations and predictions are indicated, respectively, by colored shading. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution.

Fig. 8. Measured and predicted $PM_{2.5}$ (left) and average diurnal cycle (right) in West New Delhi for 12/20/15 – 12/31/15 (top) and 09/20/16 – 09/30/16 (bottom). Here the pink lines correspond to CMAQ predictions of the “total” (solid) and “non-residential” (dotted) simulations. The solid black line represents ambient observations. Standard deviations of the diurnal profiles for observations and predictions are indicated, respectively, by colored shading. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution.

Fig. 9. Measured and predicted $PM_{2.5}$ (left) and average diurnal cycle (right) in South New Delhi for 12/20/15 – 12/31/15 (top) and 09/20/16 – 09/30/16 (bottom). Here the blue lines correspond to CMAQ predictions of the “total” (solid) and “non-residential” (dotted) simulations. The solid black line represents ambient observations. Standard deviations of the diurnal profiles for observations and predictions are indicated, respectively, by colored shading. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution.

Fig. 10. Predicted secondary organic $PM_{2.5}$ (left) and average diurnal cycle (right) for 12/20/15 – 12/31/15 (top), 09/07/09/30/15 (middle), and 09/20/16 – 09/30/16 (bottom). Bajada Pahari is shown in yellow, SOMAARTH HQ in green, West New Delhi in pink, and South New Delhi in blue. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution. Statistics are shown in Table 10.

Fig. 11. Average diurnal $\frac{\text{Residential Anthropogenic PM}_{2.5}}{\text{Total Anthropogenic PM}_{2.5}}$ (top), $\frac{\text{Residential SOA}}{\text{Total SOA}}$ (middle), and $\frac{\text{Residential SOA}}{\text{Residential PM}_{2.5}}$ (bottom). Bajada Pahari is shown in yellow, SOMAARTH HQ in green, West New Delhi in pink, and South New Delhi in blue. Shading indicates mealtimes. Residential PM is calculated as the difference in predictions from the non-residential and total emission scenario and averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution. Statistics are shown in Table 10.

Fig. 12. Predicted $\frac{\text{Total SOA}}{\text{Total PM}_{2.5}}$ (left) and average diurnal cycle (right) for 12/20/15 – 12/31/15 (top), 09/07/09/30/15 (middle), and 09/20/16 – 09/30/16 (bottom). Bajada Pahari is shown in yellow, SOMAARTH HQ in green, West New Delhi in pink, and South New Delhi in blue. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution.

Fig. 13. Predicted O_3 (left) and average diurnal cycle (right) for 12/20/15 – 12/31/15 (top), 09/07/09/30/15 (middle), and 09/20/16 – 09/30/16 (bottom) in West New Delhi (pink), and South New Delhi (blue). Standard deviations of the diurnal profiles for observations and predictions are indicated, respectively, by colored shading. Diurnal profiles were averaged over simulation durations (Table 7). Computations were carried out at 4 km resolution.

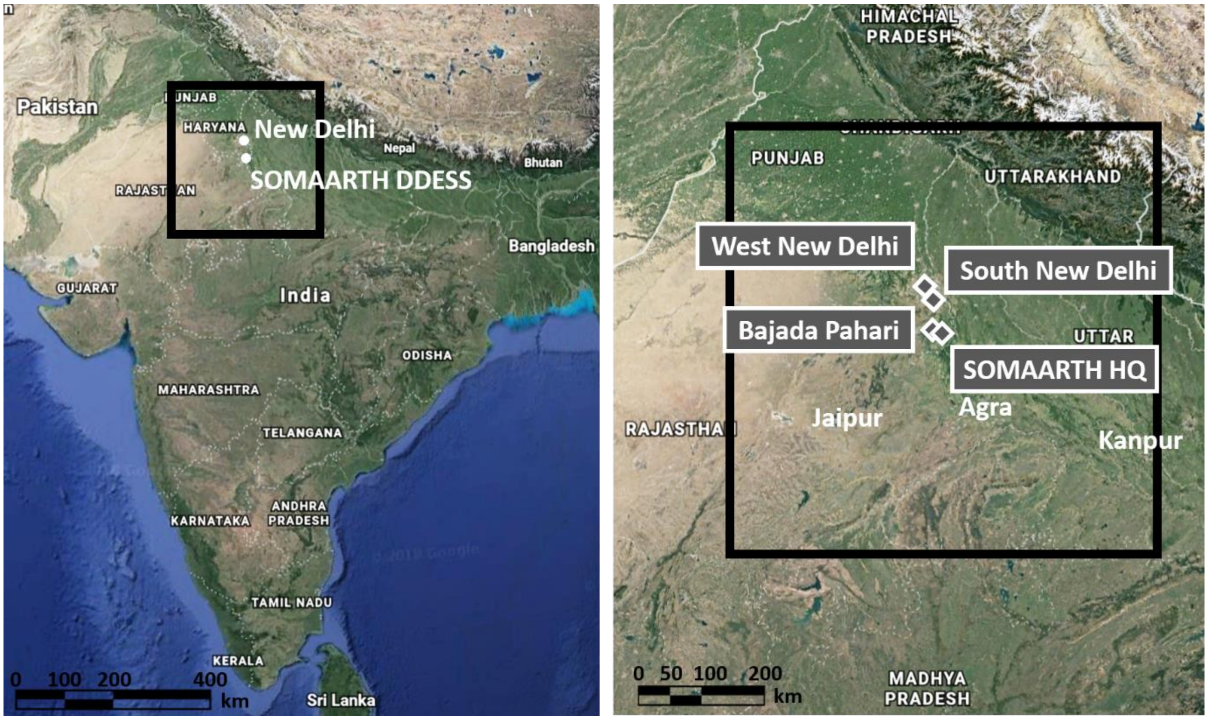
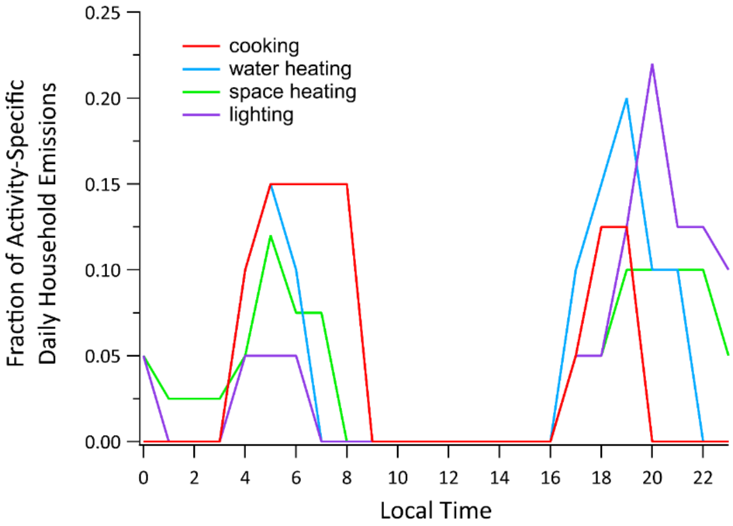

Pakistan
UNJAB
HARYANA
New Delhi
Nepal
Bhutan
RAJASTHAN
SOMAARTH DDESS
Bangladesh
GUJARAT
India
ODISHA
MAHARASHTRA
TELANGANA
KARNATAKA
ANDHRA PRADESH
TAMIL NADU
KERALA
Sri Lanka
0 100 200 400 km
HIMACHAL PRADESH
CHANDIGARH
PUNJAB
UTTARAKHAND
West New Delhi
South New Delhi
Bajada Pahari
UTTAR
SOMAARTH HQ
Agra
RAJASTHAN
Jaipur
Kanpur
0 50 100 200 km
MADHYA PRADESH

Figure 1.



Fraction of Activity-Specific Daily Household Emissions
Local Time
cooking
water heating
space heating
lighting
0.00
0.05
0.10
0.15
0.20
0.25
0
2
4
6
8
10
12
14
16
18
20
22

Figure 2.

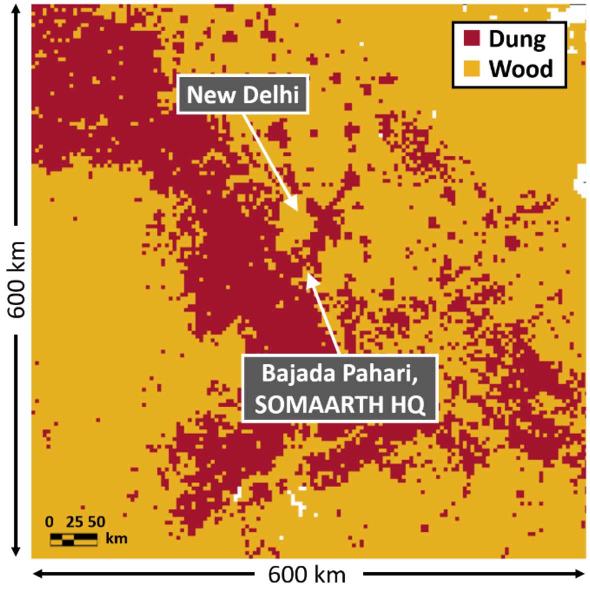

Dung
Wood
New Delhi
Bajada Pahari, SOMAARTH HQ
0 25 50
km
600 km
600 km

Figure 3.

CMAQv5.2 CB6R3 and AERO6 Anthropogenic SOA Treatment

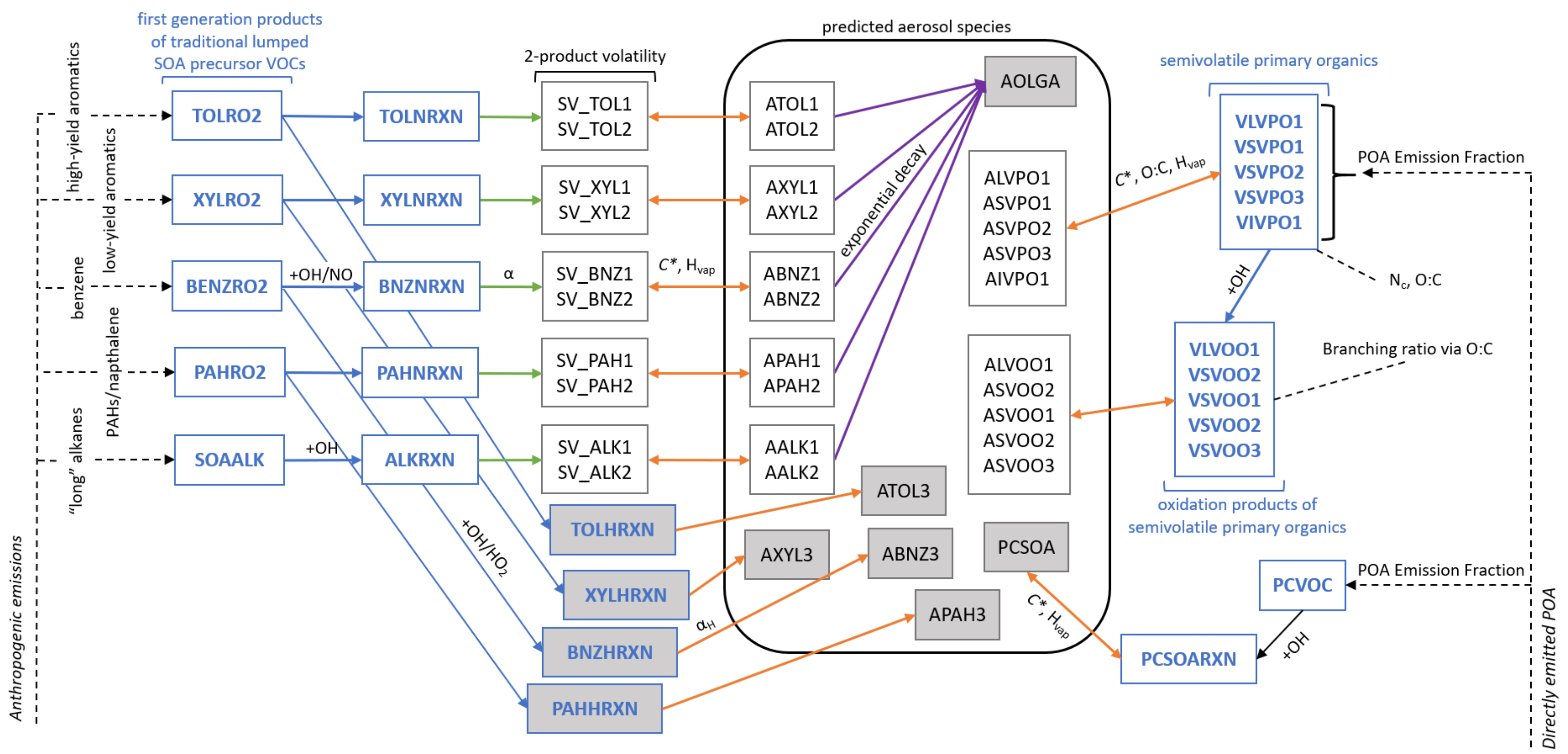
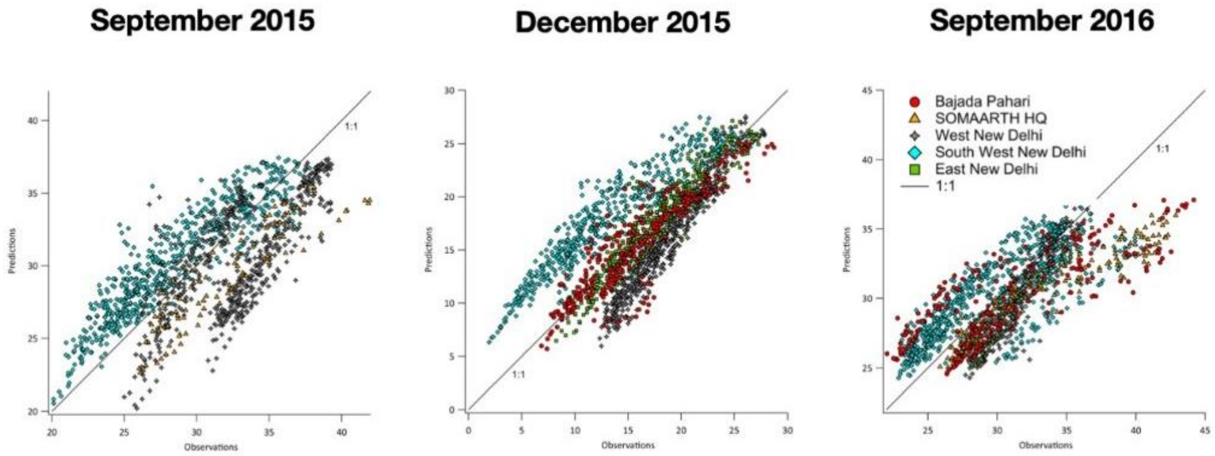


Figure 4.



September 2015

December 2015

September 2016

Figure 5.

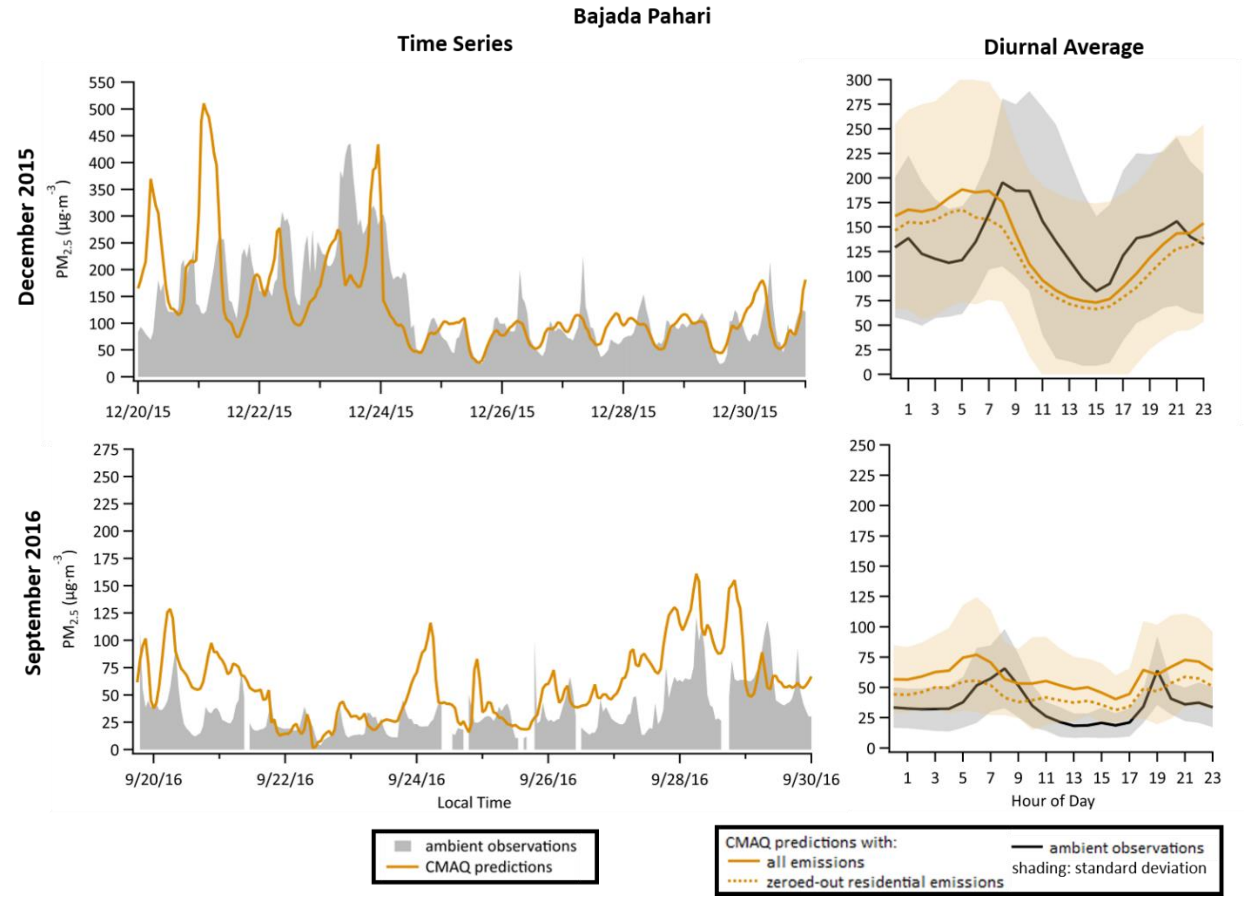

Bajada Pahari
Time Series
Diurnal Average
December 2015
PM2.5 (µg·m-3)
550
500
450
400
350
300
250
200
150
100
50
0
12/20/15
12/22/15
12/24/15
12/26/15
12/28/15
12/30/15
300
275
250
225
200
175
150
125
100
75
50
25
0
1 3 5 7 9 11 13 15 17 19 21 23
September 2016
PM2.5 (µg·m-3)
275
250
225
200
175
150
125
100
75
50
25
0
9/20/16
9/22/16
9/24/16
9/26/16
9/28/16
9/30/16
Local Time
250
225
200
175
150
125
100
75
50
25
0
1 3 5 7 9 11 13 15 17 19 21 23
Hour of Day
ambient observations
CMAQ predictions
CMAQ predictions with:
all emissions
zeroed-out residential emissions
ambient observations
shading: standard deviation

Figure 6.

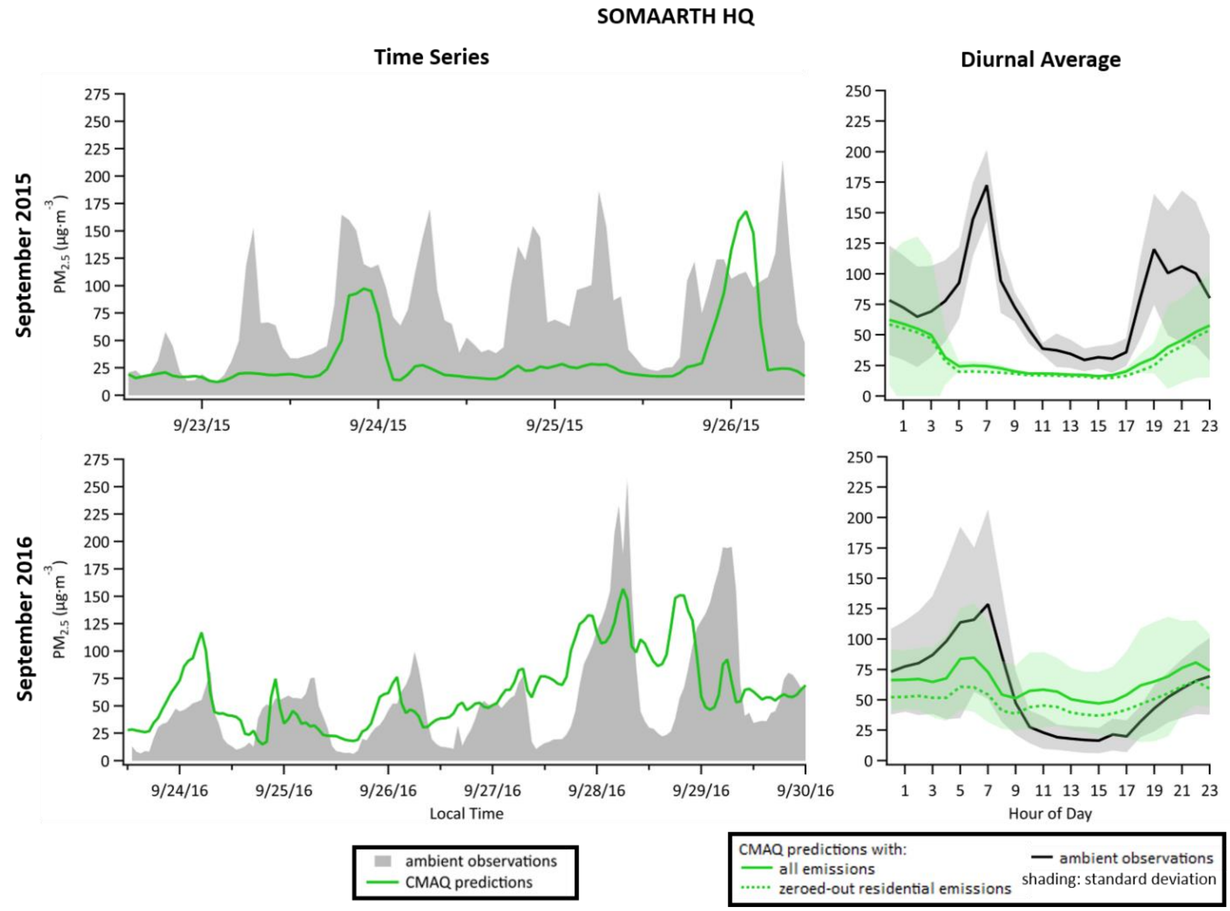

SOMAARTH HQ
Time Series
Diurnal Average
September 2015
September 2016
PM$_{2.5}$ (µg·m^{-3})
275
250
225
200
175
150
125
100
75
50
25
0
9/23/15
9/24/15
9/25/15
9/26/15
1 3 5 7 9 11 13 15 17 19 21 23
9/24/16
9/25/16
9/26/16
9/27/16
9/28/16
9/29/16
9/30/16
Local Time
Hour of Day
ambient observations
CMAQ predictions
CMAQ predictions with:
all emissions
zeroed-out residential emissions
ambient observations
shading: standard deviation

Figure 7.

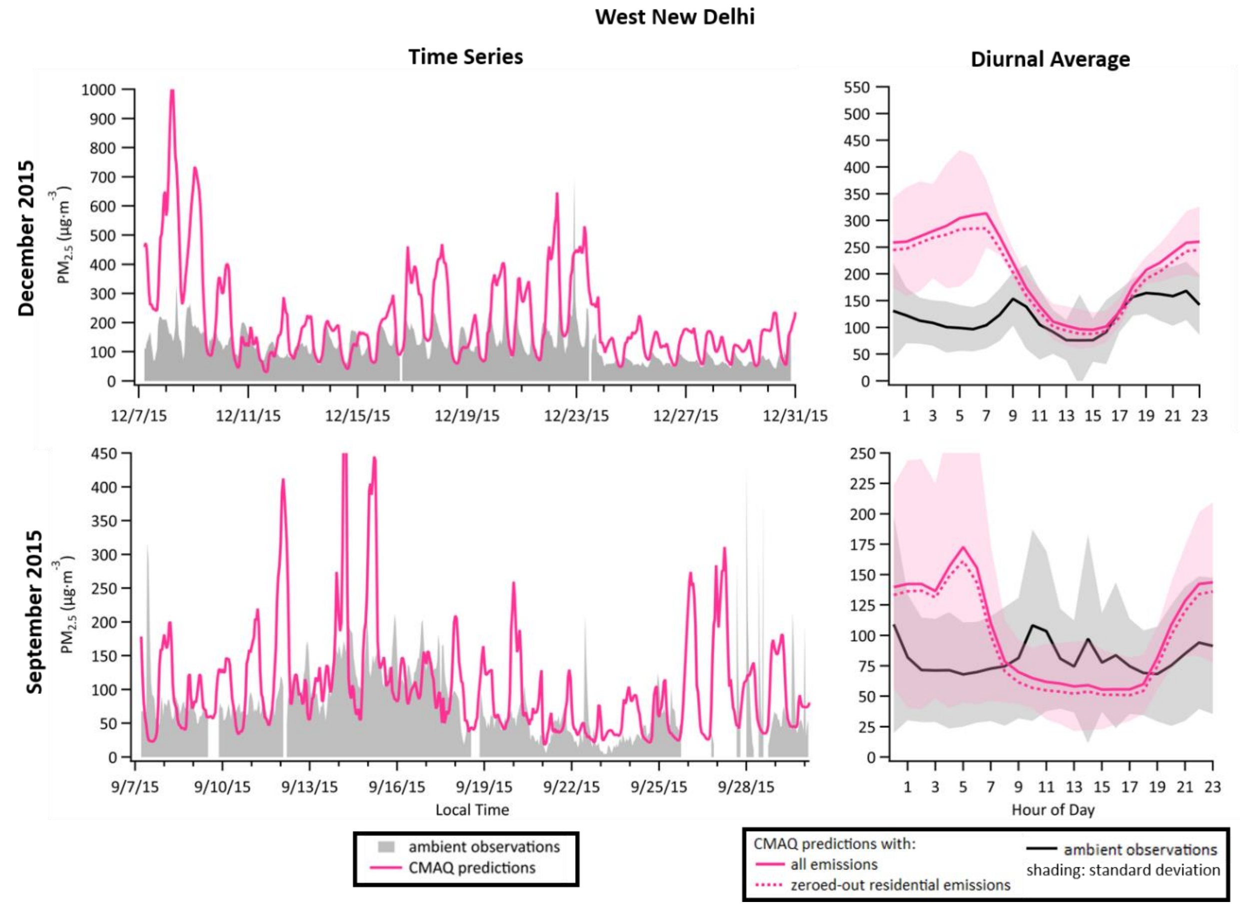

West New Delhi
Time Series
Diurnal Average
December 2015
September 2015
PM2.5 (µg·m-3)
0
100
200
300
400
500
600
700
800
900
1000
12/7/15
12/11/15
12/15/15
12/19/15
12/23/15
12/27/15
12/31/15
0
50
100
150
200
250
300
350
400
450
500
550
1
3
5
7
9
11
13
15
17
19
21
23
9/7/15
9/10/15
9/13/15
9/16/15
9/19/15
9/22/15
9/25/15
9/28/15
Local Time
0
25
50
75
100
125
150
175
200
225
250
Hour of Day
ambient observations
CMAQ predictions
CMAQ predictions with:
all emissions
zeroed-out residential emissions
ambient observations
shading: standard deviation

Figure 8.

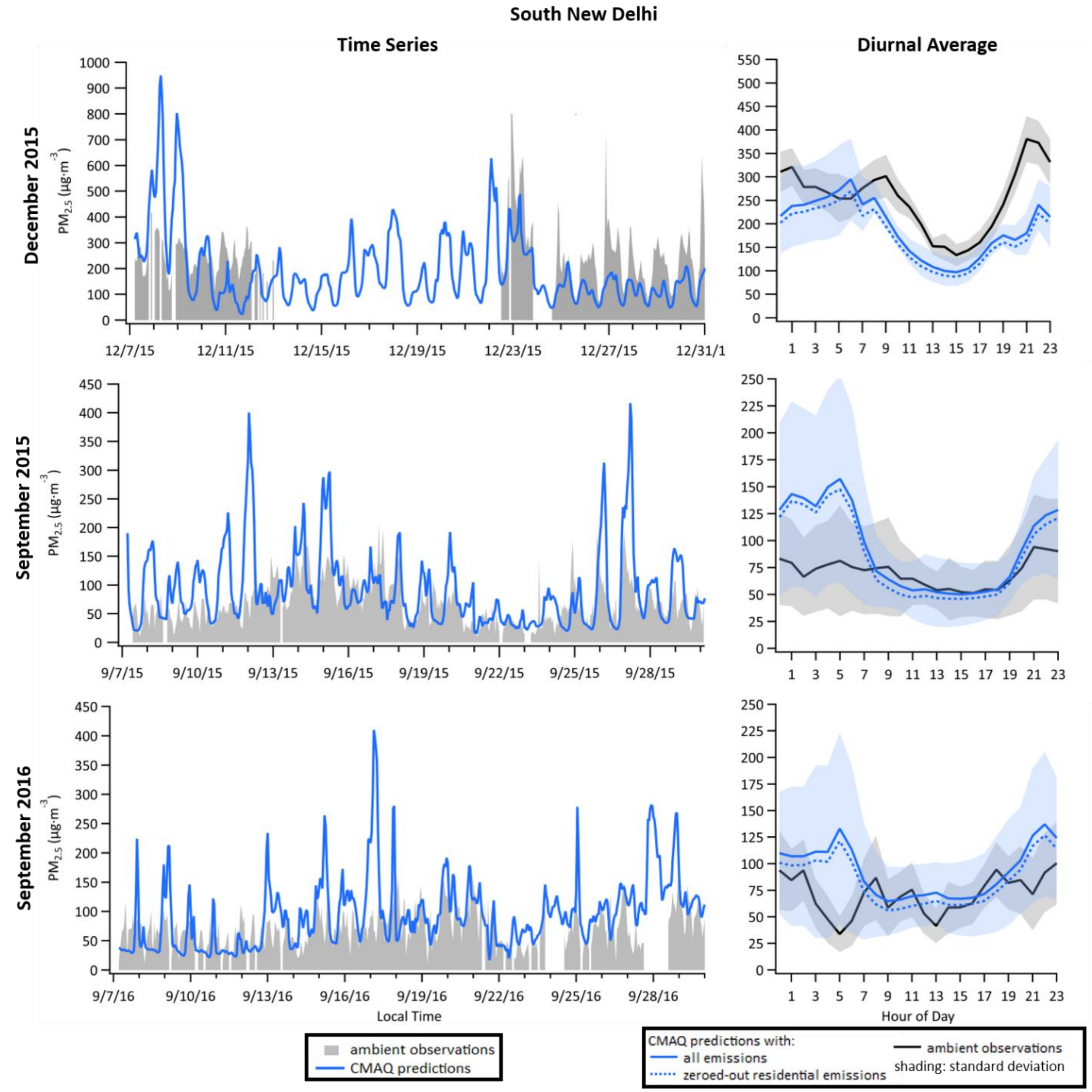

South New Delhi
Time Series
Diurnal Average
December 2015
September 2015
September 2016
PM$_{2.5}$ (µg·m^{-3})
12/7/15
12/11/15
12/15/15
12/19/15
12/23/15
12/27/15
12/31/1
9/7/15
9/10/15
9/13/15
9/16/15
9/19/15
9/22/15
9/25/15
9/28/15
9/7/16
9/10/16
9/13/16
9/16/16
9/19/16
9/22/16
9/25/16
9/28/16
Local Time
Hour of Day
ambient observations
CMAQ predictions
CMAQ predictions with:
all emissions
zeroed-out residential emissions
ambient observations
shading: standard deviation

Figure 9.

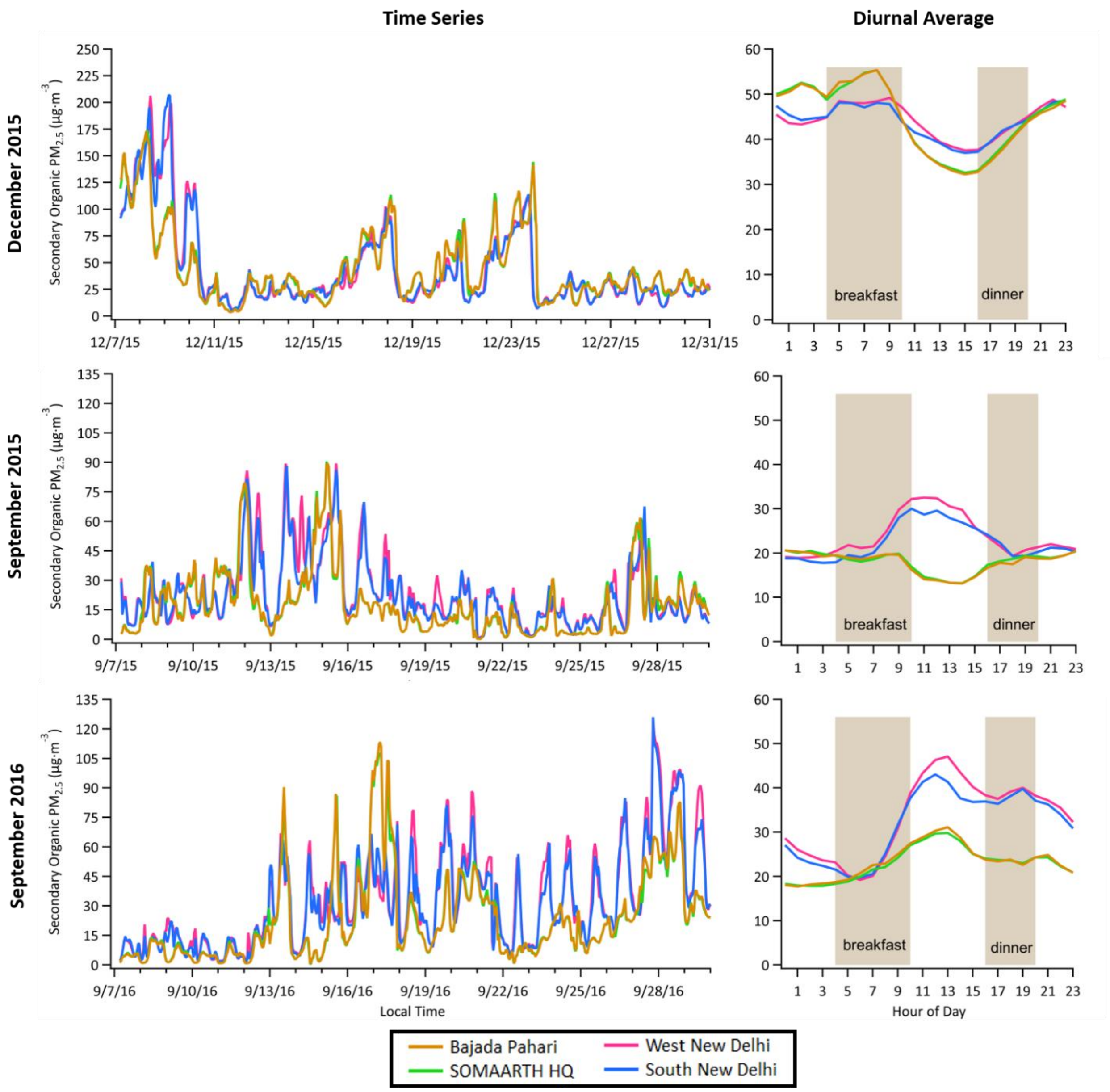

Time Series
Diurnal Average
December 2015
September 2015
September 2016
Secondary Organic PM2.5 (μg·m-3)
250
225
200
175
150
125
100
75
50
25
0
12/7/15
12/11/15
12/15/15
12/19/15
12/23/15
12/27/15
12/31/15
135
120
105
90
75
60
45
30
15
0
9/7/15
9/10/15
9/13/15
9/16/15
9/19/15
9/22/15
9/25/15
9/28/15
9/7/16
9/10/16
9/13/16
9/16/16
9/19/16
9/22/16
9/25/16
9/28/16
Local Time
60
50
40
30
20
10
0
breakfast
dinner
1 3 5 7 9 11 13 15 17 19 21 23
Hour of Day
Bajada Pahari
SOMAARTH HQ
West New Delhi
South New Delhi

Figure 10.

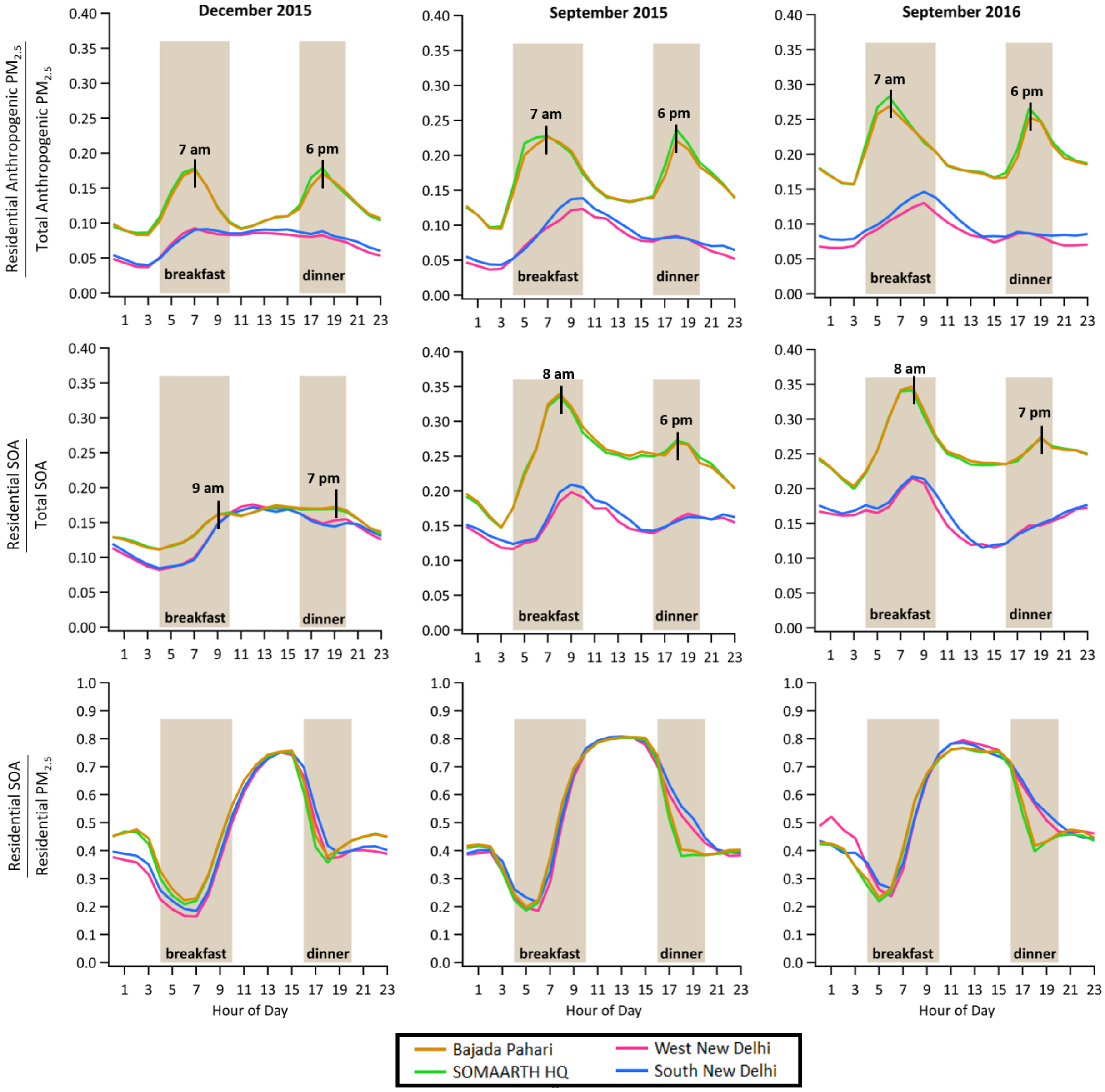

December 2015
September 2015
September 2016
Residential Anthropogenic PM$_{2.5}$ / Total Anthropogenic PM$_{2.5}$
Residential SOA / Total SOA
Residential SOA / Residential PM$_{2.5}$
0.00
0.05
0.10
0.15
0.20
0.25
0.30
0.35
0.40
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1 3 5 7 9 11 13 15 17 19 21 23
Hour of Day
breakfast
dinner
7 am
6 pm
9 am
7 pm
8 am
Bajada Pahari
SOMAARTH HQ
West New Delhi
South New Delhi

Figure 11.

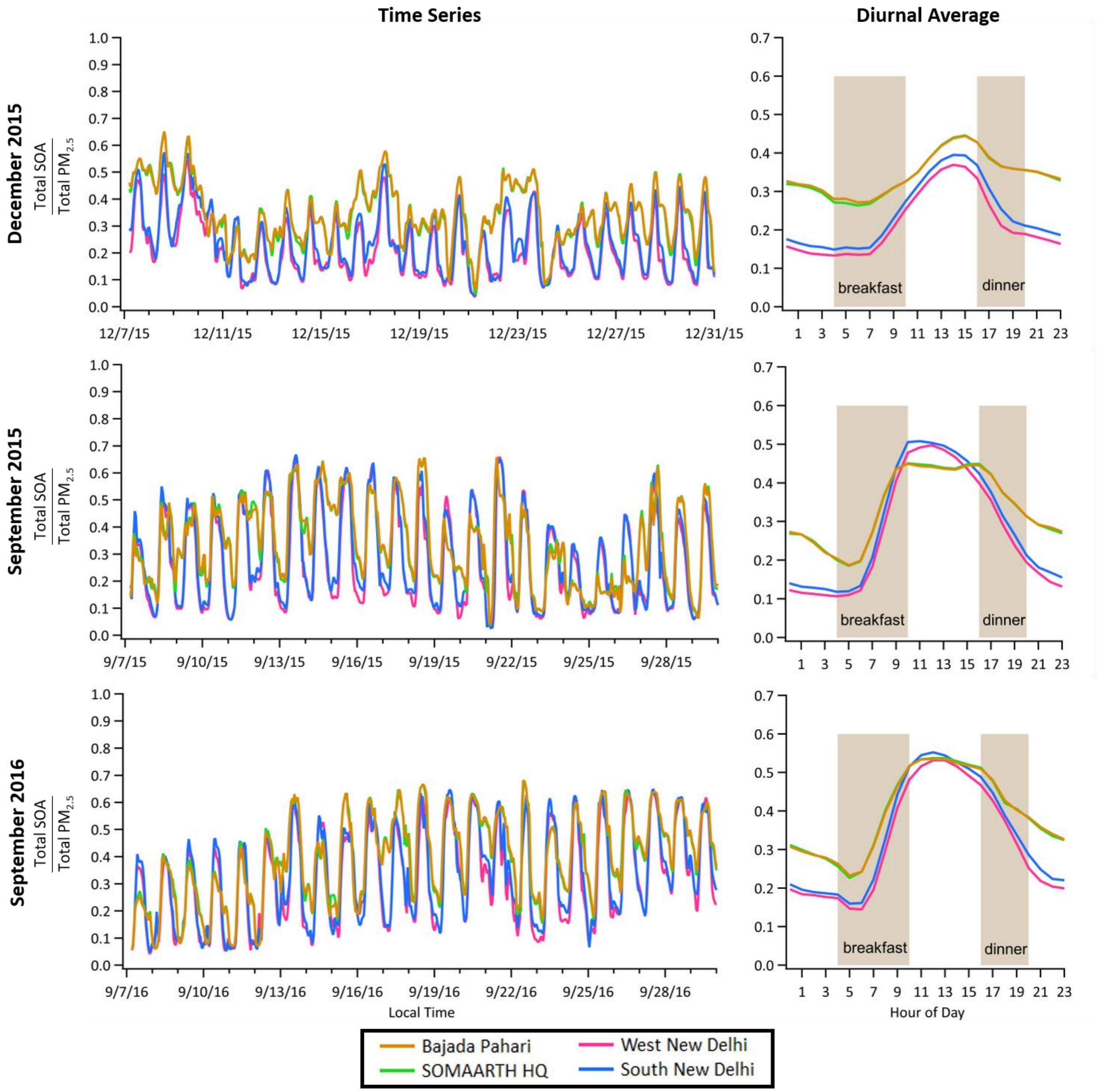

Time Series
Diurnal Average
December 2015
September 2015
September 2016
Total SOA / Total $PM_{2.5}$
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
12/7/15
12/11/15
12/15/15
12/19/15
12/23/15
12/27/15
12/31/15
9/7/15
9/10/15
9/13/15
9/16/15
9/19/15
9/22/15
9/25/15
9/28/15
9/7/16
9/10/16
9/13/16
9/16/16
9/19/16
9/22/16
9/25/16
9/28/16
Local Time
breakfast
dinner
1 3 5 7 9 11 13 15 17 19 21 23
Hour of Day
Bajada Pahari
SOMAARTH HQ
West New Delhi
South New Delhi

Figure 12.

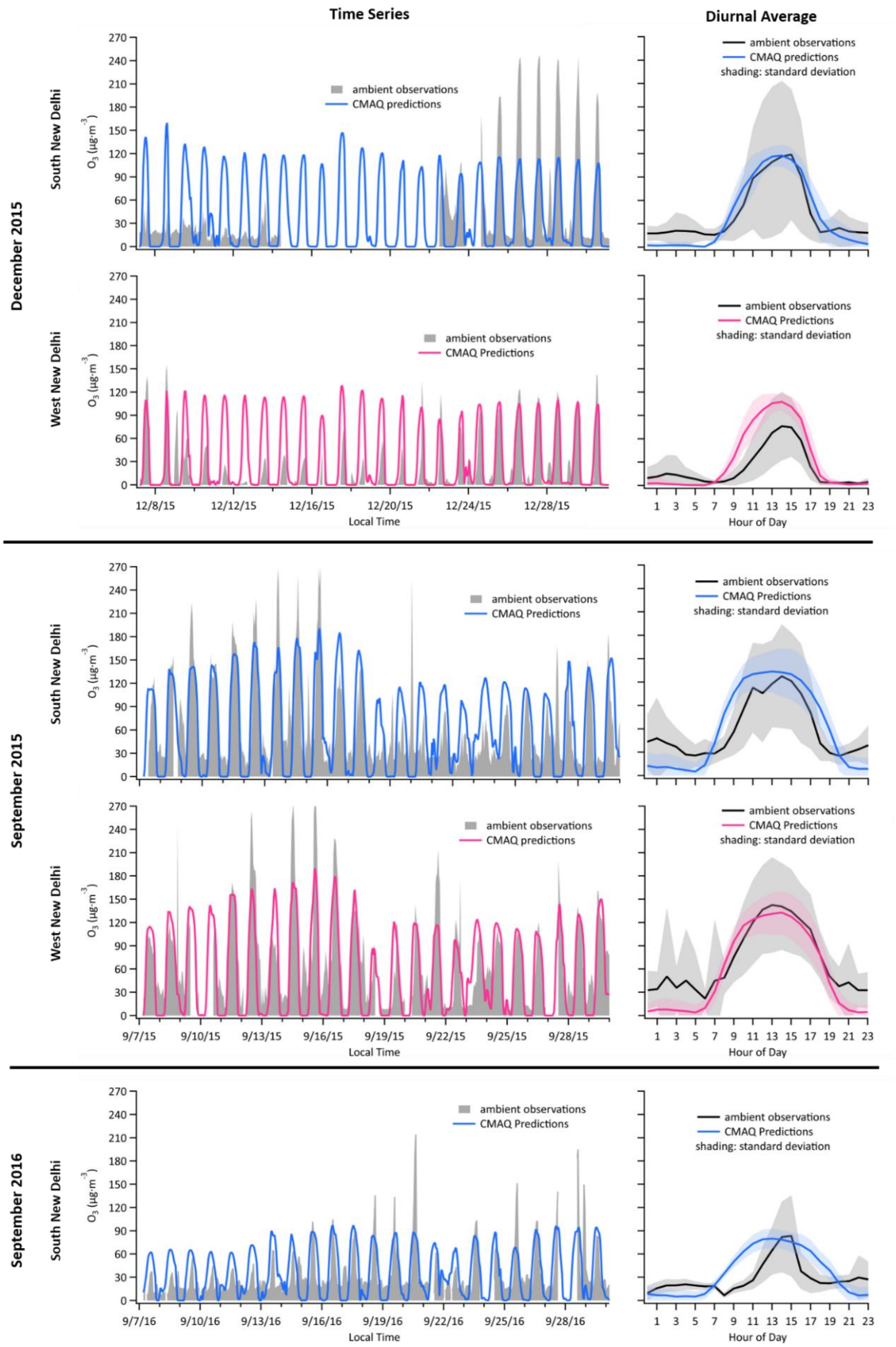

Time Series
Diurnal Average
December 2015
South New Delhi
West New Delhi
September 2015
South New Delhi
West New Delhi
September 2016
South New Delhi
O3 (µg·m-3)
270
240
210
180
150
120
90
60
30
0
ambient observations
CMAQ predictions
CMAQ Predictions
shading: standard deviation
12/8/15
12/12/15
12/16/15
12/20/15
12/24/15
12/28/15
9/7/15
9/10/15
9/13/15
9/16/15
9/19/15
9/22/15
9/25/15
9/28/15
9/7/16
9/10/16
9/13/16
9/16/16
9/19/16
9/22/16
9/25/16
9/28/16
Local Time
1 3 5 7 9 11 13 15 17 19 21 23
Hour of Day

Figure 13.