Response to reviewers' comments for "Effectiveness of short-term air quality emission controls: A high-resolution model study of Beijing during the APEC period" by Tabish Umar Ansari et al.

Anonymous Referee #1

Ansari et al. report numerical model simulations of air quality over China, focusing on Beijing, during the APEC summit in 2014. They investigate the benefit of short-term emission reduction measurements on near-surface pollutant concentrations and investigate uncertainty in model parameters. They conclude that choices of meteorological input data, model resolution and physical parameterisations are central to model performance, that emission controls were valuable in reducing pollutant levels but resulted in meeting air quality standards only because of favourable meteorological conditions.

Thank you for providing useful and constructive comments on our manuscript. We address these points in turn below.

General comments:
The authors present their research and results in a reasonably well-written manner, and I could follow their reasoning with ease. There are, however a number of major concerns that I like to see addressed before this is published.

1) *Their manuscript is too long, especially the sections on evaluating meteorological input datasets can surely be shortened and superfluous text, tables and figures moved to the supplementary material. This will improve readability and avoid loosing readers before the interesting stuff happens.*

We appreciate this concern and have cut back the model evaluation section of the paper substantially, as suggested. Tables 2-4 have been significantly shortened and full versions have been moved to supplementary material along with additional figures. Further details of the sections shortened are included in response to specific comments below.

The modelling system is in parts outdated and incomplete for an investigation of air quality in such a complex context.

We chose to use the WRF-Chem model for this study as it is one of the very best available tools for air quality modelling at the scales considered here. We apply WRF-Chem version 3.7.1 as the most up-to-date at the time this study was started and adopted specific gas-phase and aerosol chemical mechanisms that have been well tested and evaluated for this region in previous published work (Gao et al., 2016a, 2016b; Guo et al., 2016; Chen et al., 2016). We acknowledge that the model has weaknesses, as does every other modelling tool, but we address these in the text, and argue that the model does not need to be perfect to provide useful and meaningful results for the conditions we explore here. Our concern for the model skill in representing the conditions in this period is clearly demonstrated by the extensive evaluation we have performed (which the reviewer notes, but asks us to cut back), and in our identification of key weakness that need to be addressed in future studies: the SO2-NO3-NH3 balance, SOA, and PBL mixing representation, none of which are yet completely understood let alone included reliably in the latest models. We hope that our studies guide future model development but contend that weaknesses in the model we have used do not substantially affect our conclusions, as our sensitivity studies have demonstrated.

Particulate matter pollution is a intricate combination of source and sink processes which are individual for each chemical component, and their reaction to emission changes is as well. Hence all major components need to be represented (well) by the model to make believable predictions. The lack of secondary organic aerosol (up to 25% mass) and dust aerosol, as well as a strong overestimation of NO3(-) and underestimation of SO4(2-) are serious, yet total PM 2.5 mass miraculously works well. This can only be for the wrong reasons, which then has strong implications for the reliability of the results of sensitivity studies.

We acknowledge that the treatment of particle composition remains imperfect, but argue that this does not undermine the conclusions we draw on wider emission controls. It is not necessary to represent all constituents perfectly to draw clear conclusions that emission reductions will reduce PM levels.
Chemical components are generally well-represented during the APEC (November) period (see table below) with the exception of OA which is underestimated. The biases are further reduced on correcting the components for boundary-layer mixing, and these results are now included for the full October-November period in Figure S1 and Table S5 in supplement.

	Model Avg.	**Obs. Avg.**
OC	10.12	25.17
BC	4.81	3.12
NO3	11.85	8.92
NH4	4.73	4.35
SO4	3.43	4.09

The overestimation of NO3 and underestimation of SO4 are relatively small in November, as shown above. Natural dust is not an important component of aerosol at this time of the year in Beijing, and anthropogenic sources of dust are already included. The total PM2.5 mass does not work well “miraculously”; it works reasonably well for November with an appropriate representation of composition excepting the underestimation of OA, and it doesn’t work as well for October where there is an overestimation which we investigate and present in section 4. Total PM2.5 is overestimated in October principally due to insufficient boundary layer mixing as described in section 4.2 in the manuscript (see Figure 6, and we have included detail on the effect on aerosol components in the supplement, Figure S1).

We have now added the following lines in the manuscript:
P3L13: “Currently available SOA schemes are poorly parameterized for Chinese conditions and significantly underpredict SOA (Gao et al., 2016b, 2015b). SOA contributed to 17–23% of total ground-level fine particulate matter in Beijing for the October-November period investigated here, while secondary inorganic aerosols (SIA) contribute up to 62% by mass (Sun et al., 2016b). We consider the lack of SOA formation in the model in drawing our conclusions.”

P19L13: ”Since different primary and secondary aerosol components can respond differently to emission controls (Table 8), we use component-level percentage reductions from the model runs and apply them to the observed component concentrations to find the percentage reduction in total PM. This is found to be approximately 22% for both October and November periods based on the APEC-controls and October-controls runs suggesting that this scaling is appropriate and robust to uncertainties in model aerosol composition.”

2) *Hence:Missing model components (SOA, dust) are readily available, especially for the WRF-Chem model used here, so they should be used*

Dust emissions can be included in WRF-Chem but are strongly sensitive to surface wind speeds and their variability (and hence fidelity in representing meteorological processes in dust source regions) and to settling processes. Dust representation in WRF-Chem is still an area of active development (LeGrand et al.,2019, GMD). However, natural dust is primarily a problem in Northern China in Spring time, and it is not a major contributor to PM levels during the October-November period examined here. We note also that anthropogenic primary PM2.5 emissions other than OC and BC are already included as passive dust in the model.

Formation of SOA is still relatively poorly understood and remains very challenging to represent fully in models, particularly under Chinese conditions (Gao et al.,2016b). WRF-Chem 3.7.1 has several options for representing SOA, most notably SORGAM and VBS methods. SORGAM is based on SOA formation via the absorptive partitioning of surrogate oxidation products of VOCs using SOA yields determined from smog chamber experiments and has been found to underestimate SOA by an order of magnitude in Beijing (Gao et al.,2016a). The VBS method represents multigenerational ageing of IVOCs/SVOCs but these processes need measurement constraints. Currently only 1D-VBS is available in WRF-Chem and this is not coupled with all gas-phase and aerosol mechanisms. It is not available with CBMZ-MOSAIC. It is available with SAPRC-MOSAIC but without aerosol direct and indirect effects (Zhang et al.,2015) and with MOZART-MOSAIC-4bin option which sacrifices details of aerosol growth processes (only two size bins are available to represent PM1). Even with an experimentally-constrained ageing framework built on 2D-VBS (not yet available in WRF-Chem), OA loadings are underestimated by 40% (medium yield scheme) at four long-term observational sites (Zhao et. al, 2016). Such underestimation appears to be common in most parts of China during different seasons, and is exacerbated during haze events (Chen et al., 2017). In light of these continuing uncertainties we have adopted the well-tested and relatively computationally efficient chemical mechanism CBMZ-MOSAIC which represents secondary inorganic aerosol formation along with primary organic aerosol.

3) *The SO2(g) to SO4(2-)(p) seems to be wrong and needs to be fixed*

We agree that conversion of SO_2(g) to SO_4^{2-}(s) through known pathways (photochemistry and cloud chemistry) is inadequate in the model to explain huge mass yields in sulfate in North China Plain during winter, and this has been identified in previous WRF-Chem studies (Gao et al.,2016a, ACP, Chen et al.,2016, ACP). However, the actual formation pathway is still unknown. Chen et al.,2016, ACP implemented a RH-dependent pseudo first-order reaction for sulfate formation but were unable to capture the peaks during the pre-APEC period. Sulfate production during winter haze in China is still an open scientific question. Some particle-level hypotheses involving nitrogen chemistry in aerosol-water surface (Cheng et al., 2016, Science Advances) and others (Wang et al.,2016, PNAS) have been proposed but have not been parameterized for use in regional chemical transport models. To address this issue, we added additional primary sulfate in the model from the same sources as SO2 to compensate for these missing rapid reactions. This simple approach works well for the APEC period and two out of three episodes during October which were relatively drier, but it underestimates sulfate during the 21-25 October episode when the RH was high (see table S2 in supplement). To address the reviewers concerns we have discussed the implications of this assumption in Section 5 (Page 18 Line 7) of the paper.

4) *The emission inventory needs updating, and it should be done in a consistent manner rather than reducing SO2 by 60% and not touching the rest.*

Emission inventories always need updating, particularly over China where emissions are changing rapidly. However, this is not practical, particularly when working at 3 km resolution, and we have therefore made a compromise by adopting the most widely-used and evaluated emissions inventory (MEIC 2010) and adapting it to represent 2014 conditions with a simple scaling approach. MEIC 2014 has become available very recently but is not available at the 3 km grid resolution required here. We reduced SO2 emissions by 50% (not 60%) over the North China Plain in this study to reflect recent emission controls. This reduction was not arbitrary but based on the best information available and has been corroborated by recent studies. SO2 emission reduction over Eastern China between 2010-2015 has been estimated to be 48% through OMI satellite columns (Krotkov et al.,2016) and 45% through top-down emission estimates (Zheng et al., 2018). While there has been an increase in emissions for many species since 2010, recent clean air actions have reduced the emissions of key pollutants like NOx in 2014 to levels very similar to those in 2010 (Zheng et al., 2018). Therefore, our emission inventory provides a reasonably good representation of 2014 conditions and this is clear from our evaluation against observed pollutant levels.

Detailed comments:
P1L19: this relationship is considered to be non-linear (e.g. Apte et al., 2015, Conibear et al., 2018a) according to recent findings - especially for high PM, benefits are much smaller. I suggest avoiding to give an exact number if this is merely the tangent at an (arbitrary?) point of a (now known-to-be) non-linear relation.

This is a good point, and the statement has now been changed to: "It is estimated that outdoor air pollution, mostly by PM 2.5, leads to 3.3 million premature deaths per year worldwide, predominantly in Asia (Lelieveld et al., 2015)."

P2l5: comma missing after "Independent observational (. . .), modeling"

Now added

P2L20: this is not a thorough evaluation of met conditions

We agree but have not purported to do this; we merely point out that previous studies have largely neglected the role that meteorological processes play, and we aim to address this in our study. In response to reviewer 2, we have added a more detailed evaluation of meteorological conditions over the period in Tables S1 and S2.

P3L1: this sounds like an arbitrary selection of processes to investigate – reason?

We have changed P3L1 to "We present sensitivity studies to key physical and chemical processes in section 4". These processes were selected for evaluation after examining the simulation results, and the reasons for this are explained in the following section. We note at P12L5: "While the baseline model simulation with ECMWF meteorological fields reproduces observed pollutant levels reasonably well, the comparisons have highlighted uncertainties associated with resolution, vertical mixing processes, and aerosol composition. We explore the sensitivity of our results to these factors here."

P3L15: This contradicts the manuscript by Sun et al. (2016b), cited here as reference. They state in their 'Implications' section: 'We demonstrate the response of aerosol composition, size distributions, and source contributions in Beijing to emission controls during APEC based on comprehensive measurements at both ground level and at a height of 260m in urban Beijing. We observed large reductions of secondary aerosols during APEC, of 61–67% and 51–57% for SIA, and of 55% and 37% for SOA at 260m and the ground site, respectively, whereas primary aerosols at ground level did not change in the same way. This large reduction of secondary aerosol is closely linked to the corresponding reduction of precursors over a regional scale, which suppresses the formation and growth of secondary aerosol by a factor of 2–3. Our results demonstrate that the achievement of "APEC Blue" is largely a result of significant reductions of secondary aerosol due to emission controls, although the mountain-valley breeze circulation also played a role.' (Sun et al., 2016b) How do you reconcile these seemingly contradictory statements? (Especially given that several of the authors of that publication are also co-authors here) Just because your "emphasisis largely on these components here" (p3l17) does not excuse missing the rest.

Sun et al.,2016 were first to report the observations made at the IAP tower and based their claim of secondary aerosol suppression due to emission controls on a simplified theoretical framework and a direct comparison of the two periods without assessment of the substantial meteorological differences between them. However, in this study we explore the same period in a meteorologically-resolved way with a regional atmospheric chemical transport model. It is clear that the reductions were largely due to meteorological changes (see table S2) and PM2.5 levels would not have been as high as during the pre-APEC period even without emission controls (figure 8). This is one of the key messages of our study and is highlighted in table 7 and in the conclusions at page 20 line 18. To address the reviewers concern we have changed the statement on page 3 to the following:

“SOA contributed to 17–23% of total ground-level fine particulate matter in Beijing for the October-November period investigated here, while secondary inorganic aerosols (SIA) contribute up to 62% by mass (Sun et al., 2016b).”

P3l30: only reducing SO2 emissions to account for the fact that the inventory is for 2010, whilst you are simulation 2014, is arbitrary - there are projections of Asian emissions available that allow to consistently project the whole dataset.

The reduction is based on best available evidence and is not arbitrary. Please see our response to point 4 above which cites relevant papers in support of our choice.

P3L33-34: again an indication that your model might be insufficient for the task at hand!

This fully addressed in our response to point 3 above.

P4L2: MEGAN has been developed for North American conditions - can you be confident that it is applicable in China?

This is indeed correct, and previous studies using MEGAN have pointed to inaccuracies in isoprene emissions in China (Situ et al., 2013, 2014; Han et al., 2013). However, biogenic emissions are relatively low in Beijing at this time of year and are more important for ozone than for PM2.5, and we are therefore confident that this bias does not affect the conclusions of our study.

P5L20 It is confusing to conflate comparisons over different spatial areas (domains D1, 2, 3) with different sets of observations given that you do 2-way nesting in your model. In D1, meteorology and chemistry over the area covered by D2 are calculated on the D2 grid and then averaged back onto D1. Same for the region D3 in D2. So basically you are evaluating observations against model results where low pass filters of different strength were applied - no new information. At the same time you add new stations in the area not covered by inner domains. Weird.

The aim of this section was to provide a broad overview of model performance over each geographical domain. We acknowledge that nesting influences results over the parent domain but note that the domains vary substantially in size and we respectfully point out that representation bias affects comparisons over different scales despite the nesting. However, we appreciate that our comparison could appear confusing, and have simplified Table 2 by focussing on the inner domain only. We separately investigate the benefits due to nesting by sampling Beijing stations only from all domains and we present this in a simplified version of Table 4.

P5L20 Section 3.1 is overly detailed for a manuscript submitted to ACP, as it provides no further scientific insight beyond showing that meteorological variables can be simulated with good quality (known since 20 years), near-surface observations are difficult to match with a coarse grid (still quite some averaging to do at 3 km!) model, and that ECMWF IFS data seems to be a bit better than NCEP FNL (seemingly the case for 5+ years now). Hence I suggest: Shorten this paragraph to ∼10 lines, move Figure 2 and Table 2 and the rest of the paragraph into Supplementary material.

We have significantly shortened Section 3.1 as suggested, shortened Table 2 and moved the more detailed version into the supplementary material (Table S1). However, we have retained some discussion here so that the reader is clear on the depth of our evaluation. While it is known that the ECMWF data is a little better than NCEP FNL, this is not true everywhere, and it is important that we characterise this for the study location. It is also important to demonstrate how well meteorological features are captured at IAP and to show how they vary over the period to allow interpretation of the pollutant measurements available.

P8L12: so you do overestimate PM 2.5, but you don’t have SOA and dust - what makes up for this missing component, so PM2.5 mass matches observations? Why? Does the replacement have the same formation

pathways as SOA and dust? How can you pretend your model will react realistically to a change in precursor emissions given you are using different species (and formation pathways) to make PM 2.5 mass?

$PM_{2.5}$ is overestimated over certain periods due to insufficient PBL mixing (Figure 6, S1). We do not have component-level observations for $PM_{1-2.5}$. Within PM1, the extra SIA produced in the model compensates for SOA mass but only amounts to 17-23% of PM1 mass. The response to emission changes for up to 83% of PM1 mass is reliable as it is properly represented in the model. The reduction in remaining (17-23%) PM1 mass has some uncertainty (discussed in section 5) however, SOA and SIA often show simultaneous build-up and clean-up, and have similar size distributions as seen in measurements in Beijing for different periods (Zhao et al., 2017; Sun et al., 2016a,b).

P8L20: This is roughly the reduction in NO2 expected from APEC emission cuts, no? So your error is roughly the magnitude of your signal, leading to quite a low signal-to-noise ratio that needs to be discussed.

The bias in NO2 is small over Beijing, less than 10%, suggesting that the emissions are appropriate. The magnitude of the APEC emission cuts is much larger than this, so there is no problem with signal-to-noise ratio. The bias over the China domain referred to here is larger, but this reflects biases in other parts of China, outside the region affected by emission cuts, and includes the effect of representation biases; grid cells are large, while monitoring sites are principally in cities, so the model may underestimate short-lived pollutants.

P8L22: As you still have this large overestimation of SO2 in Beijing, how did you come up with the 60% reduction in emissions on P3L30? Why not more? There should be more up-to-date emission estimates for Beijing than MEIC 2010.

SO2 was reduced by 50%. Please see point 4 above for a detailed explanation.

P8L25: O3 surface obs are notoriously difficult to interpret against model simulations due to the strong titration effects during the night, especially over urban areas. How does the maximum 8 hour O3 look like?

Ozone is represented well during both daytime and nighttime as shown in Figure 4, suggesting that production and titration processes are both captured well. However, ozone is not a major pollutant at this time of year and therefore we do not devote further analysis to it in this paper.

P8L27: by chance any ammonia measurements? HNO3?

Unfortunately neither NH3 or HNO3 measurements were available during this period.

P10L20: so here you go. PM2.5 and PM10 mass is right for the wrong reasons. Will a scenario simulation give the right answers, then?

We do not have component level information for aerosol larger than PM1. Even within PM1, most of the underestimation of NO3, NH4 and BC happens during the October period and these components are much better captured during the November period. Results from scenario simulations are subject to uncertainties to physical and chemical processes investigated in section 4 but are tested for robustness in section 5 where we apportion component-level percentage reductions to observed component proportions.
Also, these component-level discrepancies are minor during the APEC (November) period (see response to point 1).

P11L3: given that you underestimate SO3, NH3 will happily bind to NO3 to neutralize and form NH4NO3.

Thank you for your comment. We have now added this reason in the manuscript at P11L13:
“Some overestimation of NO3 can also be due to this underestimation of SO4 as sulfate decrease frees up ammonia to react with nitric acid and transfers it to the aerosol phase (Seinfeld and Pandis, 2006).”

P11L6: Could technically be reasoned due to the fact that SO2 -> SO4 conversion takes some time, so most of your local SO4 might be imported. Given that you are underestimating SO2 outside of Beijing, this would make sense. But: it happens during stagnant conditions, so I would suggest that something seems seriously wrong with your model for secondary inorganic aerosols / SO2-SO4 conversion.

We address this in our response to point 3 above.

P12L3: see my previous comments on lacking model SOA.

We address this in our response to point 2 above.

P14Table4: a candidate to be put in the Supplemental Material

We have moved the full table to supplementary material as suggested but have retained PM2.5 comparisons to illustrate the importance of resolution for the major pollutants of interest here.

P15L1: it is unclear to me what you have done here - how could you mix modelled PM 2.5 up to simulated and observed PBL heights? Did you do additional simulations assimilating PBLH? Explain better!

At the model grid cell representing the IAP site we identified the model levels corresponding to the simulated and observed boundary layer height each hour. We vertically averaged the simulated PM2.5 up to these model levels to create two new time-series. This averaging was mass-weighted based on the thickness of each model level to conserve mass. We then compare these time-series with surface observations and simulated values at the surface to diagnose the effect of mixing on PM2.5 levels in the model. The analysis here involves post-processing and no additional simulations were performed. Figure 6 has been made clearer now and the legend has been updated. By "mixed" we mean vertical averaging up to the corresponding model level of the simulated or observed PBLH. We have also made this clear in the text now.

P16L12: It should be made clear that 3 km average simulations over densely urbanized areas (think high-rise buildings) cannot realistically be expected to match an observation within that area due to the strong local topographical effects.

Yes. We have acknowledged this on Page 6 Line 8 in section 3.1

P15L21: SO4 is mainly formed through liquid-phase oxidation of SO2 in cloud droplets to H2SO4 and subsequent salt formation with NH3. Hence SO2→SO4 formation is typically not limited by aerosol surface area.

Thank you for your comment. We have now changed this sentence to: "However, reduction in SO4 concentrations is negligible (1 μg m^{-3}) because sulfate formation is only indirectly associated with NH3 availability (Tsimpidi et al., 2007)"

P15L21: NH3 preferably combines with SO4 to form (NH4)2SO4, only after most SO4 is depleted, the remaining NH3 forms NH4NO3 (e.g. Seinfeld and Pandis, Atmospheric Chemistry and Physics, Wiley Interscience, 2012). Your SO4 is too low from the beginning, this "sensitivity study" hence does not take place in the right chemical regime. How can you expect your results to be meaningful?

The aim of conducting this sensitivity study was to understand the response of NO3 and NH4 to ammonia emissions in the model to explore why it differs from the real world. We have added a sentence to explain this but have moved the figure into the supplement in response to the request to shorten this section.

P15L30: We finally come to the topic of this manuscript. After 15 pages. This is too long. See my previous suggestions on how to reduce the extent of this work.

We have substantially cut back on the earlier sections as detailed above..

P19L1: I think it is an oversimplification that dust episodes only affect PM10, but not PM2.5. Apparently the APEC summit took place right in a slight dust episode, but you also do not simulate that component. WRF-Chem has multiple, easy to use dust schemes - why don't you just use them?

We note that there is a brief spike in PM10 that occurs across all Beijing measurement stations on the evening of 11 November and which is not accompanied by a similar enhancement in PM2.5. In the absence of reliable evidence to the contrary, we attributed this observed feature to dust, and this remains the most likely source. However, we appreciate that it is difficult to explore this without more detailed evidence and have amended the text to downplay this as it does not have direct relevance to the conclusions of our study.

Anonymous Referee #2
Major comments:
1. The APEC emission control analysis (Section 5) is a bit confusing in terms of writing and additional modeling analysis is needed to support the authors' conclusion that meteorology played a more important role for good air quality during APEC. First, I suggest the authors put a summary at the beginning of the section to state their overall strategies to separate the relative role of emission control vs. meteorology. Second, to put this analysis in the context of previously published ones, I suggest the authors conduct a sensitivity run in which the emission reductions are implemented over the whole study period (Oct – Nov). The resulting changes in PM2.5 concentrations should be compared to the 22% change the authors estimated. If the comparison is satisfactory, it can demonstrate the authors' simplified method is justified and such a method can be adopted by others.

Thank you for these helpful suggestions. We have revised Section 5 as suggested to make the message clearer. We have already highlighted the role of meteorology in reducing pollutant levels in our statement "The difference in baseline PM 2.5 concentrations between the October and November periods without emission controls, 279 vs. 39 μg m^{-3}, highlights the dominant role played by meteorology in bringing clean air during APEC" (Page 17 line 11) but the revisions suggested have made this clearer.

In response to these comments, we have now conducted an additional 39-day sensitivity run with APEC2 controls implemented throughout the run. Considering all 39 days, we found a daily reduction of 26±6%, and a reduction of 23±4% for days with daily mean PM2.5 >75 μg m^{-3}. This is very close to the 22% reduction that we used in our study which also accounted for the temporal application of emission controls (3 days of mild APEC1 controls followed by 7 days of more stringent APEC2 controls) and this gives us increased confidence in the approach we have taken. We have now updated the paragraph in section 5 that discusses the controls and no-controls scenario (Page 19 Line 29 to Page 20 Line 11) describing this new run and making other points clearer.

2. On the evaluation of model meteorology (Section 3), I suggest the authors add a paragraph or two to state what meteorological factors/conditions are most different between the APEC and non-APEC period and to what extent the WRF model can reproduce such differences.

Thank you for this valuable suggestion. We have added an analysis of the meteorological conditions before and during the APEC period and have presented this in table S2 in the supplement. We have added a few sentences at the end of section 3.1 to summarise this: "There are some marked differences in meteorological conditions between the non-APEC period (Episode 1: 15–20 Oct, Episode 2: 21–25 Oct, Episode 3: 26 Oct–1 Nov) and the APEC period (3–12 Nov). There is a seasonal temperature drop (7 ℃) from October to November accompanied by a drop in relative humidity, increase in wind speed and a general. change in wind direction from SE to SW which are well captured by the model. For a more detailed meteorological comparison of the pre-APEC and APEC period see table S2 in supplement". We have not added more details as the paper is already long and because reviewer 1 indicated that this section needed to be cut back.

3. I concur with the first reviewer that the manuscript is too long and particularly the tables are tedious and do not add substantial values to the manuscript. I suggest Table 2-4 can be shortened (e.g. showing only the inner domain) and put the rest in the supplementary.

We appreciate this concern and have cut back the model evaluation section of the paper substantially, as suggested by both reviewers. Tables 2-4 have been significantly shortened as suggested and full versions have been moved to supplementary material.

Minor comments:
1. The first line of the abstract: add "short-term" before emission controls.

Now done.

2. Pg 3, line 15: the statement on little SOA response to emission changes is too assertive with only one reference as support. In fact, I don't agree with this statement because (1) emission controls can affect the biogenic-anthropogenic interactions (NOx-BVOC) which affect SOA and (2) there is considerable uncertainty surrounding the role of anthropogenic VOC emissions on SOA in China. Thus, I suggest the authors change the tone of the statement and acknowledge the uncertainty in their modeling exercise due to omitting of SOA.

We have changed the statement to: "Secondary organic aerosol (SOA) formation is not included in the chemical mechanism used here. Currently available SOA schemes are poorly parameterized for Chinese conditions and significantly underpredict SOA (Gao et al., 2016b, 2015b). SOA contributed to 17–23% of total ground-level fine particulate matter in Beijing for the October-November period investigated here, while secondary inorganic aerosols (SIA) contribute up to 62% by mass (Sun et al., 2016b). We consider the lack of SOA formation in the model in drawing our conclusions"

We have also included another sentence in Section 5 at Page 18 line 9:
"percentage reduction in OC maybe overestimated because all OC is primary in the model"

3. Figure 2 and Figure 4: (1) label the APEC period; (2) add the month on the x-axis

Now done.

4. Pg 9, line 9: the November period should be the October period.

Do you mean Pg8, line9? Emission controls were applied during the November period and so we show results from October only in the figure.

5. All the time series figures should have the month on the x-axis.

Now done.

Grammar:
1. Pg 2, line 6: add comma before modeling.

Now done.

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Effectiveness of short term air quality emission controls: A high-resolution model study of Beijing during the APEC period

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Abstract. We explore the impacts of short-term emission controls on haze events in Beijing in October–November 2014 using high resolution WRF-Chem simulations. The model reproduces surface temperature and relative humidity profiles over the period well and captures the observed variations in key atmospheric pollutants. We highlight the sensitivity of simulated pollutant levels to meteorological variables and model resolution, and in particular to treatment of turbulent mixing in the planetary boundary layer. We note that simulating particle composition in the region remains a challenge, and we overpredict NH_4 and NO_3 at the expense of SO_4. We find that the emission controls implemented for the APEC Summit period made a relatively small contribution to improved air quality (20–26%), highlighting the important role played by favourable meteorological conditions over this period. We demonstrate that the same controls applied under less favourable meteorological conditions would have been insufficient to reduce pollutant levels to meet the required standards. Continued application of these controls over the 6-week period considered would only have reduced the number of haze days ~~where~~ when daily-mean fine particulate matter exceeds 75 ~~m^{-3}~~ $\mu g\,m^{-3}$ from 15 to 13 days. Our study highlights the limitations of current emission controls and the need for more stringent measures over a wider region during meteorologically stagnant weather.

1 Introduction

Air pollution poses serious health risks to urban residents and is one of the most important environmental problems facing cities around the world (Liang et al., 2017). Fine particulate matter with a diameter less than 2.5 µm ($PM_{2.5}$) is a major air pollutant that often exceeds safe limits during haze episodes which are a common occurrence in many developing megacities over the past decade. ~~It is estimated that a 10 m^{-3} decrease in~~ It has been estimated that outdoor air pollution, mostly by $PM_{2.5}$ ~~concentration is related to an increase in mean life expectancy of as much as 0.6~~, leads to 3.3 ~~years (Pope et al., 2009)~~ million premature deaths per year worldwide, predominantly in Asia (Lelieveld et al., 2015). $PM_{2.5}$ also reduces visibility

and has important impacts on regional climate (Westervelt et al., 2016). Beijing is the capital, political and cultural center of China and is among the most polluted cities in the country (Batterman et al., 2016). The population of Beijing municipality increased from 14.2 million in 2002 to 21.2 million in 2013 (Ma et al., 2014) and this has been accompanied by an increase in anthropogenic emissions across the region. High $PM_{2.5}$ concentrations are frequently reported in city clusters in the Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River Delta regions in China. Haze episodes are particularly common during winter months and have attracted substantial scientific attention (Gao et al., 2017). Independent observational (Gao et al., 2016a; Zhong et al., 2018; Shang et al., 2018; Chen et al., 2015b; Sun et al., 2016a), modelling (Matsui et al., 2009; Kajino et al., 2017; Gao et al., 2015a; Chen et al., 2016a) and long-term data analysis studies (Chen et al., 2016b; Liu et al., 2016b; Chen et al., 2015a; Yan et al., 2018) have investigated the sources, evolution and fate of $PM_{2.5}$ in Beijing, but many uncertainties remain, and improved understanding is required in order to inform sound, evidence-based emission control policies. Strict short-term emission controls have been applied effectively to improve air quality in Beijing during the Beijing Olympics in 2008 (Gao et al., 2011; Yang et al., 2011) and more recently for major events such as the Asia-Pacific Economic Cooperation (APEC) summit in November 2014 (Li et al., 2017b; Wang et al., 2016b) and the Victory Parade in 2015 (Liang et al., 2017; Liu et al., 2016a; Zhao et al., 2017). Real-world emission controls provide an ideal opportunity to test current scientific understanding of the sources and processing of air pollution as represented in models in a robust way. With improved confidence in model performance over a focus region we can explore the impact of alternative control options to aid formulation of more effective policies for emission reduction.

A number of previous studies have investigated the effect of emission controls during the APEC period in November 2014 using surface observations (Sun et al., 2016b; Xu et al., 2015; Wang et al., 2016b; Li et al., 2017b; Zhou et al., 2017) and atmospheric chemical transport models (Zhang et al., 2016; Guo et al., 2016; Wang et al., 2017; Gao et al., 2017) and have found that $PM_{2.5}$ concentrations were much lower than during the preceding weeks. Many of these studies have attributed this improved air quality largely to the emission controls that were applied without thoroughly evaluating the role of meteorological variations. Comparison with observations in preceding weeks or over similar time periods in earlier years does not adequately account for the role of meteorology in governing haze episodes. Model studies with and without emission controls are insufficient to evaluate the contribution of meteorological processes if they focus on the control period alone, without evaluating the model performance outside the control period. Gao et al. (2017) found that the emission controls reduced $PM_{2.5}$ levels by about 18 ~~m^{-3}~~ $\mu g\,m^{-3}$ during APEC with about half the reduction due to emission controls in surrounding districts outside Beijing. However, the study involved coarse resolution (27 km) model simulations which may be insufficient to capture regional and city-level atmospheric events well, and lacked component level analysis of aerosols. Other studies have noted the role of meteorology during the period but have not quantified it, attributing the benefits mostly to emission controls.

In this study we investigate the effectiveness of short-term emission controls and how meteorological processes influence this, using the APEC period as an example. We use a nested version of the Weather Research and Forecasting model with Chemistry (WRF-Chem) over China with a specific focus on the Beijing-Tianjin-Hebei region. WRF-Chem has been used successfully at coarser resolution in previous studies investigating haze formation over Beijing (Matsui et al., 2009; Tie et al., 2014; Zhang et al., 2015; Chen et al., 2016a). We describe the model setup, emissions and observations in Section 2. In

~~Section~~section 3 we present a thorough meteorological and chemical evaluation of the model simulations against surface observations and tower measurements, including aerosol composition, and we assess the strengths and weaknesses of the model. We ~~also test different meteorological inputs to drive the model. We~~ present sensitivity studies to ~~model resolution, uncertainties in ammonia emissions and boundary layer processes in Section~~key physical and chemical processes in section 4. In ~~Section~~section 5 we investigate the impact of emissions controls over the APEC period and compare these with the same controls over a period two weeks earlier to demonstrate the important role of meteorological conditions in governing their effectiveness.

2 Model configuration and the APEC period

We use the WRF-Chem model (Grell et al., 2005; Fast et al., 2006) version 3.7.1 to simulate the meteorology and air quality over Northern China. Previous studies have shown that WRF-Chem is capable of reproducing air quality in China relatively well (Gao et al., 2015a, 2016b; Guo et al., 2016; Chen et al., 2016a). We use the Carbon Bond Mechanism version-Z (CBMZ) chemistry scheme coupled with the Model of Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol module (Zaveri et al., 2008). CBMZ explicitly treats 67 species with 164 gas-phase, heterogeneous and aqueous reactions, and provides a suitable compromise between chemical complexity and computational efficiency. MOSAIC uses a sectional approach with eight aerosol size bins and treats the key aerosol species, including sulfate, nitrate, chloride, ammonium, sodium, black carbon (BC), primary organic mass, liquid water and other inorganic mass. Secondary organic aerosol (SOA) formation is not included in the chemical mechanism used here. ~~While SOA may contribute as much as~~ Current SOA schemes are poorly parameterized for Chinese conditions and significantly underpredict SOA (Gao et al., 2016b, 2015b). SOA contributed only 17–23% ~~to aerosol composition in the October–November~~ of total ground-level fine particulate matter in Beijing during the period investigated here, ~~it does not respond strongly to emission controls (Sun et al., 2016b). In contrast~~ while secondary inorganic aerosols ~~contribute~~ contributed up to 62% by mass ~~of total fine particulate matter over the North China Plain, and show a significant decrease in response to emission controls (Sun et al., 2016b), and our emphasis is largely on these components here.~~

(Sun et al., 2016b). We consider the lack of SOA formation in the model in drawing our conclusions. Further details of the model configuration used in this study are given in Table 1.

We perform two-way coupled simulations with three nested domains that include China as the parent domain (D01) at 27 km horizontal resolution, Northern China as a nest (D02) at 9 km resolution and the North China Plain as an innermost nest (D03) at 3 km resolution, as shown in Fig. 1. The model is nudged to meteorological reanalysis data above the boundary layer every six hours for winds, temperature and moisture to permit direct comparison of the simulations with observed pollutant concentrations under comparable conditions.

We use anthropogenic emissions from the Multi-resolution Emission Inventory for China (MEIC) for the year 2010 (Li et al., 2017c). This provides emissions of major air pollutants including NO_x, CO, NMVOC, SO_2, NH_3, $PM_{2.5}$, PM_{10}, BC and OC from five major emission sectors that include residential, traffic, industry, power and agricultural sources, and has

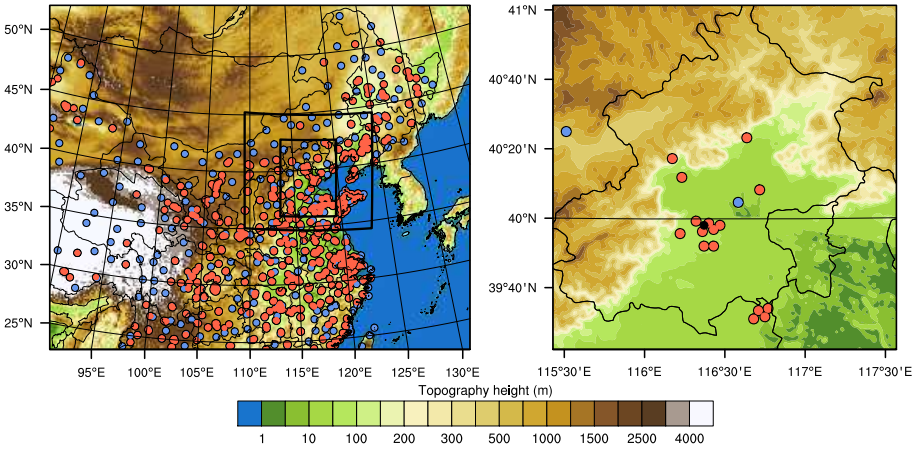

Figure 1. Map of ~~topography over~~ the model domain (left) showing nests over Northern China and the North China Plain, and map of Beijing municipality (right) showing the location of IAP (black) and measurement stations for meteorology (blue) and air quality (red).

Table 1. Model configuration used in this study

Configuration	Description
Horizontal resolution	27 km, 9 km, 3 km (3 domains)
Vertical levels	31 with model top at 50 hPa
Aerosol scheme	MOSAIC (8 bins) (Zaveri et al., 2008)
Photolysis scheme	Fast-J photolysis (Wild et al., 2000)
Gas-phase chemistry	CBMZ (Zaveri and Peters, 1999)
Cumulus parameterization	Grell 3-D scheme
Shortwave radiation	RRTMG shortwave scheme (Clough et al., 2005)
Longwave radiation	RRTMG longwave scheme (Mlawer et al., 1997)
Cloud ~~Microphysics~~ microphysics	Lin scheme (Lin et al., 1983)
Land surface scheme	NOAH LSM (Chen and Dudhia, 2001)
Land-use data	MODIS 20 category at 30 arcseconds
Surface layer scheme	Monin-Obukhov scheme (Monin and Obukhov, 1954)
Boundary layer scheme	YSU (Hong et al., 2006)
Meteorological conditions	ECMWF 6-hourly data
Chemical boundary conditions	MOZART ~~(Emmons et al., 2010)~~

been used in a number of previous modelling studies (Li et al., 2015; Gao et al., 2015a; Zhang et al., 2015; Chen et al., 2015a, 2016a). Emissions were provided at the native resolution of each domain, i.e., at 27 km, 9 km and 3 km. We impose a vertical profile for these emissions over the lowest eight model levels to account for the effective source height distribution

for each sector based on the distribution used for EMEP emissions (Bieser et al., 2011; Mailler et al., 2013), and impose a diurnal cycle for each ~~source sectorin the MEIC inventory~~sector. SO_2 emissions over the Beijing-Tianjin-Hebei region were reduced by 50% to account for strong emission reductions between 2010 and our focus year of 2014 ~~(Zheng et al., 2018)~~ (Zheng et al., 2018; Krotkov et al., 2016). We assume that 6% by mass of SO_2 is emitted as primary SO_4 to account for the discrepancy between high observed concentrations of SO_4 and low secondary production in the model (Gao et al., 2015a; Chen et al., 2016a; Li et al., 2017a). Biogenic emissions are based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2012). These are calculated online in the model based on canopy and emission factors and factors for leaf age, soil moisture, leaf area index, light dependence and temperature responses. Hourly fire emissions are included from the Fire Emissions Inventory from NCAR (FINN, Wiedinmyer et al., 2011) to represent biomass burning, although this is not a major source in the region at this time of year.

To evaluate the model, meteorological observations were obtained from the National Climate Data Center (NCDC) hourly integrated surface database (http://www.ncdc.noaa.gov/data-access/) for all of China. These sites are shown in Fig. 1. We focus on 2 m temperature and relative humidity and 10 m wind speed and direction for model evaluation. Vertical profiles of meteorological variables were ~~also~~ obtained from the 325 m high observational tower located at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences, Beijing (39°58′28″ N, 116°22′16″ E). This provides independent measurements of temperature, relative humidity, wind speed and wind direction at 17 different height levels. Measurements of boundary layer mixing height were retrieved from aerosol lidar profiles at IAP (Yang et al., 2017), providing a valuable additional test of model meteorological processes. Hourly concentrations of NO_2, CO, SO_2, O_3, $PM_{2.5}$ and PM_{10} are available from the national monitoring network run by the China National Environmental Monitoring Center (CNEMC). In addition, ~~over the October–November 2014 period~~ detailed measurements of atmospheric pollutants and aerosol composition were made from the IAP tower over the October-November 2014 period. These include measurements of NH_4, NO_3, SO_4, and OC from an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) instrument at 260 m altitude (Sun et al., 2016b) and from a High Resolution Aerosol Mass Spectrometer (HR-AMS) instrument at the surface (Xu et al., 2015), and BC at the surface was measured with an Aethalometer. The size-segregated samples collected at the two heights were analyzed for water-soluble ions. Detailed procedures for the data analysis are described in Ng et al. (2011) and Sun et al. (2012).

3 Model Evaluation

To investigate the strengths and weaknesses of the model in representing air quality in China, the model was evaluated against meteorological and pollutant measurements across all three domains and at the IAP tower site in Beijing.

3.1 Meteorology

We test the model performance ~~with~~ using two sets of meteorological fields: Final Reanalysis data (FNL) from the National Centers for Environmental Prediction (NCEP) and ERA-Interim data from the European Centre for Medium-Range Weather Forecasts (ECMWF). Table 2 presents a ~~domain-based~~ comparison of the performance of ~~simulated meteorological variables~~

Table 2. Comparison of observed and simulated meteorological variables using FNL and ECMWF fields

~~Number of Obs . avg . Ob~~ ~~Stations~~
2-m Temperature (°C)
Beijing ~~1~~ 9.68
~~D03~~ North China Plain ~~30~~ 8.91
~~D02 77 7.87 7.53 7.55 -0.34 -0.32 2.39 2.35 0.95 0.95 D01 324 9.62 7.77 7.79 -1.85 -1.83 3.23 3.23 0.94 0.94~~ 2-m Relative Humidity (%)
Beijing ~~1~~ 54.7
~~D03~~ North China Plain ~~30~~ 54.9
~~D02 77 54.4 47.8 51.1 -6.6 -3.3 17.4 15.2 0.74 0.78D01 324 62.8 60.4 62.6 -2.4 -0.2 16.8 15.6 0.73 0.76~~ 10-m Wind Speed ($m\,s^{-1}$)
Beijing ~~1~~ 5.41
~~D03~~ North China Plain ~~30~~ 5.73
~~D02 77 6.18 3.60 3.55 -2.58 -2.63 4.52 4.55 0.67 0.66D01 324 5.67 3.38 3.36 -2.29 -2.31 4.29 4.30 0.60 0.61~~ 10-m Wind Direction (°)
Beijing ~~1~~ 197.5
~~D03~~ North China Plain ~~30~~ 215.1
~~D02 77 214.4 212.2 208.9 -2.8 -5.5 65.4 65.4 0.76 0.76 D01 324 206.5 193.4 188.4 -13.1 -18.0 71.9 72.2 0.74 0.74~~ height

Hourly values are taken from 1 station in Beijing and 30 stations over the North China Plain from 12 October to 19 November 2014. Where observation data are missing, model values were remove

~~with~~ the model against ground-based observations from the NCDC dataset ~~when the model was run using these meteorological fields. With both sets of fields the~~ for Beijing and the North China Plain. For a detailed evaluation over each model domain, please see Table S1 in the supplement. The average 2 m temperature is reproduced well over ~~domains 2 and 3, but is slightly underpredicted over the largest domain, and is overpredicted for~~ the North China Plain but is overpredicted at the single Beijing site. ~~The Beijing observations are made~~ This is located at the airport on the outskirts of the city, and may not be representative of the wider region. The ~~correlation coefficients are high over all three model domains (0.94–0.95) with both ECMWF and FNL fields. The~~ surface relative humidity is underpredicted for all domains with both sets of fields, although the biases are smaller and correlation ~~coefficients higher~~ is better with ECMWF data. The humidity is underpredicted by about 15% at the Beijing site and this may have implications for heterogeneous reactions and the hydroscopic growth of secondary aerosols. The 10 m wind speed is substantially underpredicted ~~with~~ using both sets of fields~~for all domains, and performance is least good~~ , and this is most notable for the Beijing site~~where the bias is greater than 50% with both fields~~. However, the correlation ~~coefficient~~ at this site is ~~slightly better than over the three model domains~~ reasonably good suggesting that the hourly variability in wind speeds is captured ~~moderately well. If the underprediction of wind speeds extends above the surface, then this may lead to the build-up of gas-phase and aerosol species in the simulations and to overproduction of secondary aerosols due to unrealistic stagnation. The model captures the~~ adequately. The 10 m wind direction ~~reasonably well with both sets of data and the correlation coefficient is close to 0.80 for Beijing. It is notable that the correlation coefficient improves for most variables between domain 1 and domain 3, as the model resolution increases from 27 km to 3 km.~~ and its variability are also reproduced relatively well. Based

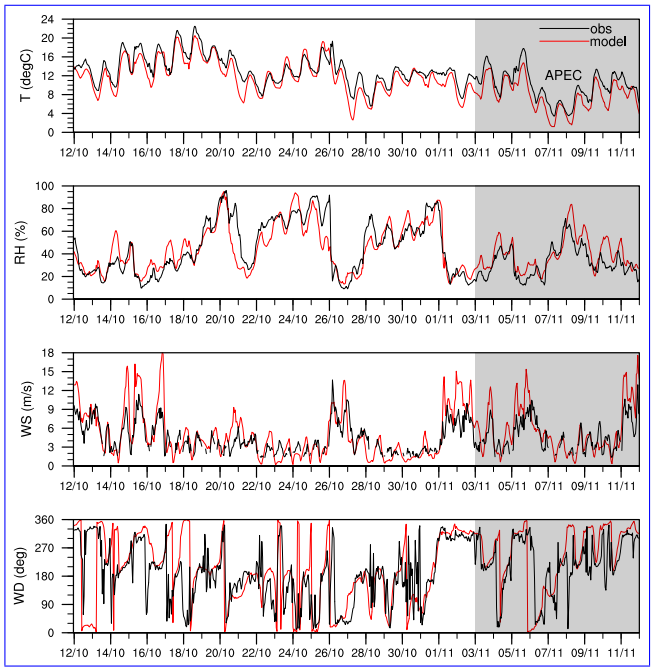

Figure 2. Comparison of meteorological measurements at 190–310 m on the IAP tower in Beijing with model simulations using ECMWF meteorological fields between 12 October and 12 November 2014. The period with emission controls is shaded.

on these comparisons~~with meteorological observations~~, and on subsequent comparison of pollutant concentrations, we find that the model performs marginally better using the ECMWF meteorological fields. With these fields the model captures the timing of pollution episodes better, leading to more realistic pollutant behaviour, and we have therefore chosen ~~ECMWF fields over FNL fields~~ to use ECMWF fields for our model studies.

Figure 2 presents an evaluation of meteorological variables with measurements from the IAP tower. We evaluate the model against measurements at 190–310 m (model level 4) to minimize the effects of buildings surrounding the site, which are not adequately resolved in the model. The daily maxima and minima in temperature are reproduced reasonably well with a small underestimation that averages less than 2 °C. The diurnal variations and averages for relative humidity, wind speed and wind direction are also captured well. The mean bias in relative humidity is 0.9% and the large underprediction seen at the airport meteorological station ~~evident in Table 2 is not seen~~ is not evident here, suggesting that it may be a surface level feature or reflect ~~the~~ overestimation of temperature at that location. Over the height of the tower (5 model levels, Figure S2) the diurnal variation in humidity drops by more than a factor of two, very similar to the reduction seen in the observations. The wind speed is slightly overestimated during windier periods, with a mean bias of 0.54 m s^{-1} ~~Again, this~~ . This suggests that the underestimation of 10-m wind speeds at meteorological stations seen in Table 2 is a surface feature in the model, and does not represent a systematic bias throughout the boundary layer. The synoptic patterns in all four variables are captured very well, highlighting the quality of the ECMWF meteorological data, and there is only one occasion on 20–21 October when substantial

deviations in temperature and humidity are evident. There are some marked differences in meteorological conditions between the APEC period (3–12 Nov) and the period preceeding it. These include a gradual temperature drop of 7 °C associated with the changing seasons which is accompanied by a drop in relative humidity. There is an increase in the frequency of northwesterly flow with higher windspeeds, and this contrasts strongly with the lighter windspeeds and more frequent southerly flow in October. These changes are captured well by the model. A more detailed comparison of the meteorological consitions is given in Table S2 in the supplement.

3.2 Air Quality

We ran the model ~~for 41 days~~ from 10 October to 19 November 2014 using ECMWF meteorology~~. The first 40 hours~~ , and the first two days were set aside as model spin-up. A comparison of ~~modelled pollutants~~ hourly modelled pollutants for Beijing and the North China Plain against measurements from the CNEMC network is presented for October in Table 3 and the mean spatial distribution of $PM_{2.5}$ during October is shown in Fig. 3. We do not include the November period here because emission controls were implemented across Beijing and surrounding provinces from the beginning of November. ~~A full time-series of the comparison to observations is shown in Fig. 4~~ A more detailed comparison of concentrations on an hourly and daily basis over all model domains is given in Table S3.

~~Table 3 shows a comparison of the key gas-phase and particulate species for all the surface pollutant stations from the observation network over the corresponding model domains.~~ The model overpredicts average surface $PM_{2.5}$ ~~slightly (~~over the period by 5–18% ~~) across all~~ across the three model domains. The correlation ~~coefficient~~ for hourly $PM_{2.5}$ improves with resolution~~from~~ , from r=0.47 for domain 1 to 0.63 for domain 3 and 0.68 for the 12 Beijing sites. The model underestimates PM_{10}~~across all domains~~, although the biases are relatively small (<10%) over Beijing. This ~~underestimation~~ may be attributed to neglect of mineral dust sources in the model, which play a relatively small role over Beijing at this time of year. CO is ~~significantly underestimated for domain 1~~ underestimated over much of China, suggesting that the emissions in the inventory are too low, but the biases ~~reduce with increasing resolution and are smallest for the Beijingsites. The underestimation of CO for coarser domains may reflect the heterogeneity of sources, although the consistency of this bias and the relatively high levels of observed CO suggest an underestimate of CO sources across much of China in the emissions inventory~~are relatively small over Beijing, and the variability is captured well. A similar effect is seen for NO_2, which is underestimated by as much as 45% over ~~the outer model domain~~parts of China, but by ~~a much smaller margin over Northern China, and averages~~ only 8% over the Beijing sites. ~~While this may reflect an underestimate in emissions, the improvement is partly due to~~ This may partly reflect better representation of the emissions distribution for this shorter-lived pollutant on a finer grid. SO_2 is underestimated by 13% over ~~domain 1~~ most of China but is overestimated over Beijing by a factor of three. ~~This~~ The large overestimation for Beijing can be attributed to the recent rapid reduction in emissions in the region between 2010 and 2014 that are not represented in the 2010 inventory (Zheng et al., 2018). Ozone ~~shows a contrasting trend, with an overestimate of 50% for domain 1 reducing to 5% for domain 3 and a 3% underestimation over Beijing. This~~ is reproduced well over Beijing, but is overestimated over much of China; this may reflect the bias in NO_2 concentrations, and is likely to be ~~heavily~~ influenced by the urban ~~characteristics~~ locations of most of the air quality stations.

Table 3. Comparison of hourly pollutant concentrations with network measurements over the period 12–31 October 2014

~~Stations hourly/daily hourly/daily hourly/daily~~ height$PM_{2.5}$ (~~m^{-3}~~µg m^{-3})	~~Number of Sta~~
Beijing stations	12
~~D03~~ North China Plain	137
~~D02 375 75.8 87.9 12.1 63.9/48.6 0.60/0.69 0.65/0.71 D01 1312 71.1 74.8 3.7 61.1/50.2 0.47/0.53 0.48/0.54~~ PM_{10} (~~m^{-3}~~µg m^{-3})	
Beijing stations	12
~~D03~~ North China Plain	137
~~D02 375 138.0 98.6 -39.4 94.3/75.8 0.54/0.65 0.44/0.52 D01 1312 121.0 82.2 -38.8 89.0/76.7 0.42/0.47 0.32/0.37~~ CO (ppm)	
Beijing stations	12
~~D03~~ North China Plain	137
~~D02 375 1.14 0.66 -0.48 0.88/0.79 0.33/0.37 0.20/0.20 D01 1312 1.00 0.50 -0.50 0.79/0.73 0.32/0.34 0.13/0.14~~ NO_2 (ppb)	
Beijing stations	12
~~D03~~ North China Plain	137
~~D02 375 24.86 19.45 -5.41 16.99/13.21 0.49/0.55 0.44/0.50 D01 1312 22.73 12.45 -10.28 18.33/15.44 0.42/0.47 0.30/0.36~~ SO_2 (ppb)	
Beijing stations	12
~~D03~~ North China Plain	137
~~D02 375 12.23 13.21 0.98 13.19/9.01 0.24/0.34 0.26/0.28 D01 1312 10.27 8.93 -1.34 11.17/8.54 0.19/0.28 0.18/0.24~~ O_3 (ppb)	
Beijing stations	12
~~D03~~ North China Plain	137
~~D02 375 21.23 23.08 1.85 17.19/12.80 0.42/0.43 0.37/0.40 D01 1312 21.44 32.29 10.85 22.44/17.03 0.29/0.27 0.27/0.25~~ height	

Where observation data are missing, model values were removed to ensure consistent sampling.

For most pollutants, the correlation ~~coefficient~~ and slope improve substantially with resolution, and are better on a daily mean basis than at hourly resolution. This suggests that the day to day variability driven largely by regional meteorological processes is captured better than the diurnal variations driven by chemistry and local boundary layer mixing, as expected. This is particularly noticeable for ozone, although concentrations of this pollutant remain low at this time of year. Daily mean concentrations are typically used for most metrics of pollutant impacts on human health, and the reasonable model performance for daily averaged data suggests that it is suitable for assessment of these policy-relevant metrics.

The spatial distribution of mean $PM_{2.5}$ concentrations over 12–31 October is shown in Fig. 3. The distribution is captured reasonably well by the model, with the western parts of China showing clean air with concentrations less than 10 ~~m^{-3}~~µg m^{-3}) while the eastern, more populous parts of the country show average concentrations of 70–150 ~~m^{-3}~~µg m^{-3}. Key hot-spots over the North China Plain, Central China and the Sichuan Basin are reproduced, and concentrations in coastal regions are notably lower, matching observations. The North China Plain is one of the most densely populated parts of the country, incorporating major cities such as Beijing, Tianjin, and Shijiazhuang, and frequently experiences heavy haze episodes with high levels of

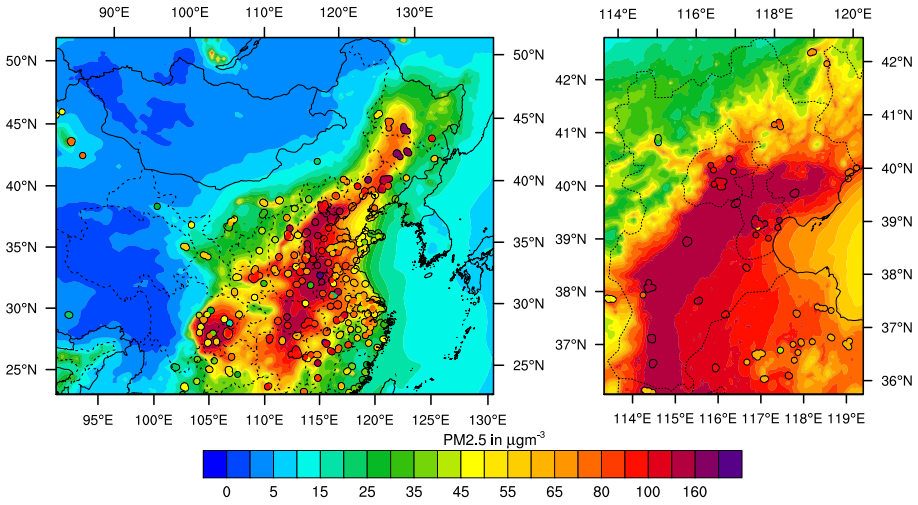

Figure 3. Average spatial distribution of $PM_{2.5}$ over the period 12–31 October 2014 for model ~~domain~~Domain 1 (China, left) and ~~domain~~Domain 3 (North China Plain, right) along with observations shown in circles.

particulate matter (Wang et al., 2014; Gao et al., 2015a). Highest concentrations of $PM_{2.5}$ occur on the western side of the North China Plain, where they are trapped by southeasterly winds against the Taihang mountains, and this is reproduced well by the model. There is a notable east-west gradient as concentrations drop off eastwards towards the coast. Over the mountains to the northwest of Beijing concentrations are much lower, typically less than 40 ~~m^{-3}~~ $\mu g\ m^{-3}$.

Figure 4 shows the time-series of key gas-phase and particulate pollutants averaged over the 12 network sites in Beijing. The general synoptic and diurnal patterns of $PM_{2.5}$, PM_{10}, CO, NO_2 and O_3 are reproduced well by the model, including the magnitude of daily maxima and minima. SO_2 is greatly overestimated in October, reflecting recent rapid emission reductions in Beijing (Zheng et al., 2018), and this is consistent with the findings of previous studies (Chen et al., 2016a; Gao et al., 2015a; Guo et al., 2016). However, we note that SO_2 is reproduced much better from 15 November onwards, following the start of the heating season, highlighting the continuing major importance of this source. The observations show that the region experiences clear synoptic patterns of pollutant build-up over 4–5 days followed by sudden clean-out which is typically associated with frontal passage from the northwest (Guo et al., 2014). These synoptic patterns are seen more clearly for particulate matter than for gas-phase pollutants like NO_2 and CO which exhibit a stronger diurnal signal reflecting chemical and dynamical processes. With the exception of SO_2, key pollutants and their variation over this period are reproduced well.

~~A more critical test of model performance is made by comparison of~~ Comparison of aerosol composition with measurements at IAP over this period provides a more critical test of model performance, see Fig. 5. ~~For all three episodes in October the~~ The model overestimates BC, NO_3 and NH_4 and underpredicts OC and SO_4 ~~. The overestimation~~ during the three episodes in October. Overestimation of BC likely reflects ~~use of emissions for 2010, highlighting reductions~~ the reduction in emissions between 2010 and 2014, but may also indicate insufficient removal in the model. The overprediction of NO_3 and NH_4 ~~could~~ may be due to uncertainty in NO_2 and NH_3 emissions or to overestimated gas to particle conversion in the model. In particular,

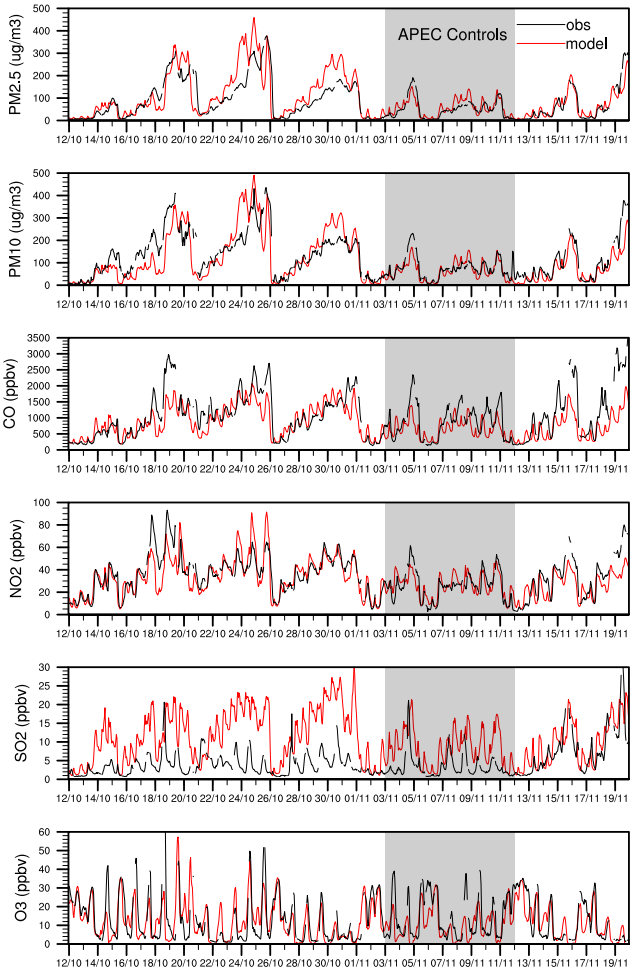

Figure 4. Mean time-series of surface pollutants over the 12 air quality stations in Beijing. Model values are with baseline emissions at all times including during the APEC period (shown shaded).

~~the model may overestimate~~ secondary production of NO_3 and NH_4 may be overestimated during stagnant conditions ~~such as those occurring during the three October episodes(note the low wind speeds shown in Fig. 2)~~during pollution episodes, but matches better ~~during the first half of November ,~~ in November when conditions are less stagnant. The underestimation of SO_4 occurs despite an overestimation of gas-phase SO_2, highlighting insufficient formation of SO_4 in the model. This may also contribute to the overestimation of NO_3 as a decrease in SO_4 frees up ammonia to react with nitric acid and transfers it into the aerosol phase (Seinfeld and Pandis, 2006). The underestimation of OC can be explained by the absence of secondary organic aerosol in ~~the chemical mechanism we have used~~our studies.

The ~~relative proportions of and to other components are similar at the surface and 260 m, while the proportion of OC is lower at 260 m and that of~~ model captures the vertical gradients of NO_3 ~~is higher. The model concentrations of~~ and SO_4 ~~are~~ well,

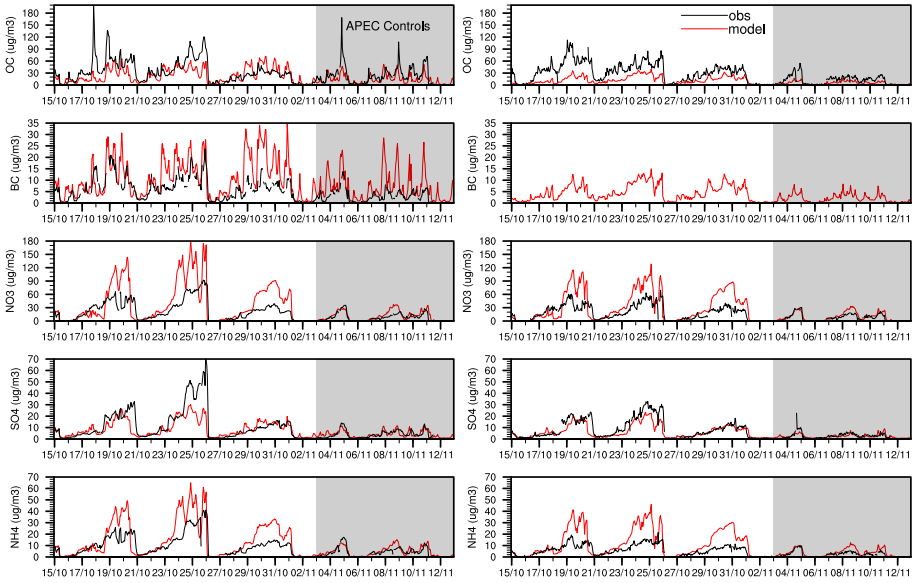

Figure 5. Measured and simulated aerosol components at the surface (left) and 260 m (right) on the IAP tower in Beijing. Model values are with baseline emissions at all times including during the APEC period (shown shaded).

with drops of 10–15% and 30% ~~lower at~~ between the surface and 260 m, respectively, ~~which is very~~ similar to the ~~observed reduction. The modelled is~~ drops seen in the observations. However, the model shows a weaker vertical gradient than observed (22~~% lower at 260 m, less than the observed reduction of~~ % vs. 33% ~~, and this may~~ drop) for NH_4 which can be attributed to ~~an overproduction of secondary~~ higher secondary production of NH_4 in the model~~which reduces the vertical concentration gradient . Modelled OC falls off more quickly with altitude than in the observations~~ . For OC, the model shows a stronger vertical gradient than observed (53% vs. 12% ~~) and this is likely to be because the OC in the model is primary and therefore its vertical distribution reflects the surface source, while there is more secondary OC in the observations (Sun et al., 2016b) which leads to a weaker gradient with altitude~~drop) which reflects the lack of secondary production at elevated levels in the model.

4 Investigating model ~~sensitivity~~weaknesses

While the baseline model simulation with ECMWF meteorological fields reproduces observed pollutant levels reasonably well, the comparisons have highlighted uncertainties associated with resolution, vertical mixing processes, and aerosol composition. We explore the sensitivity of our results to these factors here.

4.1 Model resolution

~~Running the model at high resolution comes at a substantial cost in computing resources and simulation time.~~ To investigate the ~~gains that increased horizontal resolution provides~~benefits of high spatial resolution, we sample all three model domains at

Table 4. Impacts of model resolution on simulation of hourly ~~pollutant~~ $PM_{2.5}$ concentrations in ($\mu g\,m^{-3}$) in Beijing over 12–31 October 2014

~~(m^{-3})~~	Mean Obs	Mean Mod	Bias	RMSE	NMB	NME	Correlation	Slope
D03 (3-km)	155.4	141.5	-13.9	96.5	0.65	0.79	D02	2670
D02 (9-km)	155.4	143.6	-11.8	96.6	0.65	0.80	D01	2670
D01 (27-km)	155.4	137.9	-17.5	96.6	0.65	0.79	D01 (no nest)	

~~(m^{-3}) D03 2670 155.4 141.5 -13.9 96.5 0.65 0.79 D02 2670 155.4 143.6 -11.8 96.6 0.65 0.80 D01 2670 155.4 137.9 -17.5 96.6 0.65 0.79 D01 (no nest~~

the 12 Beijing stations and compare the results with observations. ~~We use~~ To eliminate the influence of two-way nesting, ~~so results from the~~ where results from nested domains feed back to the parent domain~~. To eliminate this effect~~, we perform an additional simulation at 27 km resolution over the parent domain only. Table 4 shows a comparison ~~with measurements over Beijing in October~~ of modelled $PM_{2.5}$ over Beijing for the different resolutions with measurements in October. In the nested simulation, $PM_{2.5}$ is overestimated by 14% for domain 1, 19% for domain 2 and 16% for domain 3, but is underestimated by 8% for the domain 1 simulation without nesting. Although the mean biases do not improve with higher resolution, reflecting the two-way nesting, there is a substantial improvement in the correlation coefficient (0.59 to 0.68) and slope (0.55 to 0.83) for $PM_{2.5}$ when nesting is used, and this occurs for other pollutants too (see Table S4). For many variables the results sampled at 9-km resolution (D02) are slightly better than those sampled at 3-km resolution (D03)~~, although it should be noted that results at D02 are influenced by the higher-resolution simulation at D03 through the two-way nesting.~~. Results at 27-km resolution without nesting are substantially less good than those with two-way nesting, highlighting the important contribution of the coupling. We conclude that it is worth performing simulations at higher horizontal resolution as it gives a better representation of urban pollution levels.

4.2 Boundary layer mixing

Representing turbulent mixing processes in the boundary layer well is critical for simulating surface air quality. The nighttime boundary layer under stable meteorological conditions is particularly difficult to model, and we find that the mixing height is often severely underpredicted (~~and is~~ as low as 20 m on ~~several~~ some occasions) causing pollutant concentrations to reach unrealistically high levels. Nudging meteorological fields to ECMWF reanalysis data reduces this bias but does not remove it. After testing a number of different boundary layer algorithms we selected the Yonsei University (YSU) scheme (Hong et al., 2006) as it provides the best overall match to lidar-derived observations of boundary layer height. However, stable conditions remain a challenge for this scheme, and we therefore explore the sensitivity of simulated surface concentrations to boundary layer mixing under these conditions.

Figure 6 shows the time-series of simulated and observed planetary boundary layer (PBL) height. The observed PBL height was derived from lidar ~~data~~ extinction profiles at IAP using the cubic root gradient method of Yang et al. (2017). The simulated

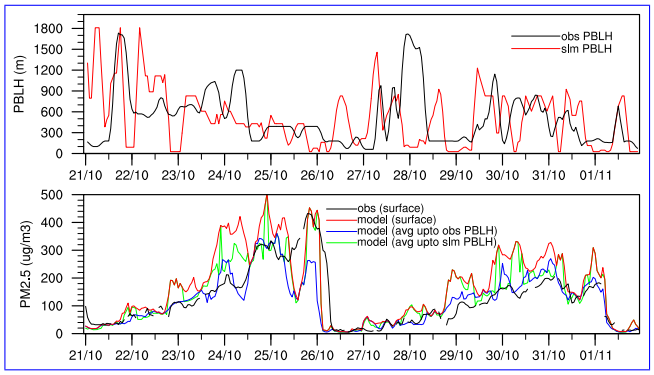

Figure 6. Simulated and observed boundary layer mixing height in metres (top) and simulated and observed $PM_{2.5}$ in ~~m^{-3}~~ $\mu g\,m^{-3}$ showing the effect of mixing up to the PBL height in the model (bottom) between ~~20~~21 October and ~~2~~1 November 2014.

PBL height was diagnosed using the maximum decrease in the modelled $PM_{2.5}$ profile to ensure a consistent definition. We compare the observed PBL height with the simulated height at IAP, and use $PM_{2.5}$ measurements from the surface pollutant station at Aotizhongxin, the closest station to the IAP site (within 2 km) to assess the effect on $PM_{2.5}$ concentrations. The PBL height shows highly variable behaviour over the day and from day to day. While the ~~model average~~ average model PBL height (514 m) is similar to the ~~observed average height~~ average observed (509 m) over the haze episodes shown, the ~~model severely underpredicts the~~ nighttime PBL height is severely underpredicted on a number of occasions. Assuming that the PBL height reflects the efficiency of mixing in the boundary layer, we expect the model to overpredict surface pollutant concentrations under these stable nighttime conditions, and this is seen in the time-series of $PM_{2.5}$ ~~at Aotizhongxin shown~~ in Fig. 6. To account for misrepresentation of local boundary layer mixing, we ~~show the modelled~~ post-process the model results by vertically mixing $PM_{2.5}$ ~~vertically-averaged~~ up to the simulated mixing height, to ~~minimise~~ eliminate the effect of underestimated mixing, and up to the observed mixing height, to provide a ~~clearer~~ direct comparison against $PM_{2.5}$ observations. ~~Mixing~~ Averaging up to the observed PBL height gives a substantial improvement in $PM_{2.5}$ levels compared to observations, particularly for the episodes of 21–25 October and 27 October–1 November when the model ~~significantly~~ underestimates the PBL height. The simulated mean surface $PM_{2.5}$ concentration during the period is reduced from 169 to ~~130~~118 ~~m^{-3}~~ $\mu g\,m^{-3}$ (the observed mean is 129 ~~m^{-3}~~ $\mu g\,m^{-3}$) and the RMSE is reduced from 94 to 65 ~~m^{-3}~~ $\mu g\,m^{-3}$, and the biases in NO_3, NH_4 and BC are significantly reduced. For a more detailed analysis of component-level sensitivity to boundary layer mixing see Figure S1 and Table S5 in the supplement.

These results highlight ~~that~~ the importance of representing PBL mixing well for accurate reproduction of surface pollutant levels~~with the model is tightly linked to how well it can reproduce PBL mixing~~. We note ~~that the PBL shows~~ a steady decline in PBL height over the pollution episode during 21–25 October, and $PM_{2.5}$ shows a consistent build-up over ~~the same~~ this period. This provides ~~some~~ observational evidence for the radiative feedback between aerosol concentrations and mixing height, and this appears to be captured relatively well by the model, as shown in previous studies (Gao et al., 2015b). ~~To further improve~~ Further improvement in simulation of surface pollutant concentrations ~~, additional research is needed to accurately~~

Table 5. Mean concentrations (in ~~m^{-3}~~ $\mu g\,m^{-3}$) at IAP during 21–25 October 2014

Species	Control run	Reduced NH_3 run	Observations
$PM_{2.5}$	210.8	154.9	157.5
NO_3	61.28	36.60	33.81
NH_4	23.11	15.24	15.03
SO_4	12.70	11.59	20.40

~~model~~ requires additional research on representaton of PBL mixing processes in urban environments. Profiles of aerosol and meteorological variables from high-resolution lidar measurements provide an important aid to such investigations.

4.3 Regional NH_3 emissions

The aerosol components NO_3 and NH_4 are overestimated in these simulations, as shown in Fig. 5. These components are governed by secondary production from their gaseous precursors NO_2 and NH_3. Since the concentration of NO_2 is close to that observed, we perform a short sensitivity study over the pollution episode from 21–25 October with NH_3 emissions over the North China Plain reduced by 50% to ~~investigate the effect on aerosol composition~~explore the response of NO_3 and NH_4 to ammonia emissions in the model. We find that the reduction in NH_3 emissions ~~not only~~ reduces NH_4 ~~concentrations but also~~ and ~~, to a lesser degree, concentrations, see Fig. ??~~NO_3 concentrations substantially and brings them closer to observations (see Table 5 and Figure S3). This is likely to be because NH_3 is the limiting reactant in the formation of NH_4NO_3 that directly controls the concentration of both NH_4 and NO_3 aerosols ~~(Gao et al., 2016b) and consequently a reduction in may suppress the secondary formation of~~ in the North China Plain (Gao et al., 2016b; Chen et al., 2016a). However, reduction in SO_4 ~~due to reduced aerosol surface area on which it can form. These reductions in aerosol components reduce total~~ concentrations is small ($1\,\mu g\,m^{-3}$) because SO_4 formation is only indirectly associated with NH_3 availability (Tsimpidi et al., 2007). Total $PM_{2.5}$ ~~concentration~~ concentrations are reduced by approximately 26% bringing ~~it~~ them closer to observed concentrations~~at the IAP site, see Table 5~~. Ammonia emissions were reported to be 1574 kt/yr over the Beijing-Tianjin-Hebei region in 2010 (Zhou et al., 2015) while those in the MEIC emissions inventory used here are only 540 kt/yr. Given that our NH_3 emissions are already low compared with other studies (Kang et al., 2016), we do not reduce them further in this study. However, we ~~demonstrate~~ have demonstrated that $PM_{2.5}$ concentrations during this period are highly sensitive to NH_3 emissions, consistent with the findings of other studies (Zhang et al., 2016), and highlight this issue for further investigation.

~~Time-series of aerosol components , and at the IAP site and at Aotizhongxin showing simulated concentrations (in m^{-3}) from the baseline model run and reduced emissions run compared to observations.~~

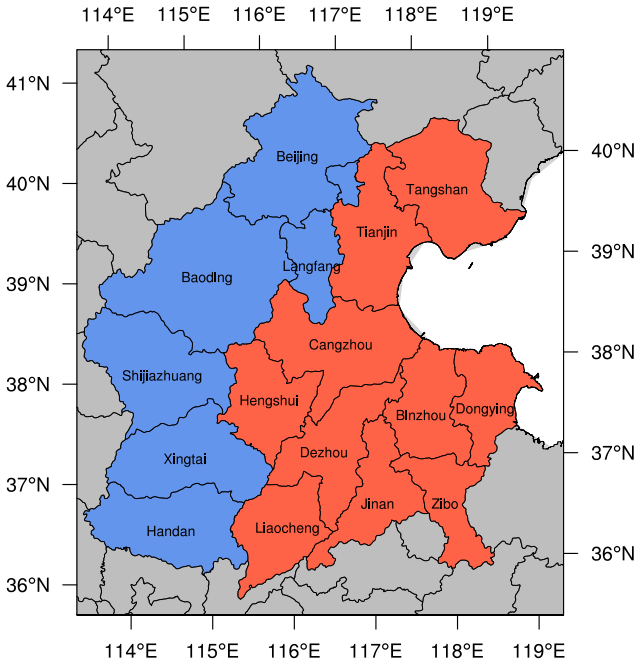

Figure 7. Map ~~showing~~ of districts where major emissions controls were implemented during the APEC period. During phase 1 emissions were ~~restricted in~~ focussed on Beijing and western Hebei (blue) and in phase 2 additional controls were ~~additionally~~ applied over other parts of the North China Plain (red).

5 APEC Emission Controls

The Asia-Pacific Economic Cooperation (APEC) summit was held from 10–12 November 2014 in Beijing, and was the focus of short-term emission controls to ensure good air quality over the period. Emission controls were applied in Beijing and surrounding regions including Tianjin city, the provinces of Hebei, Shanxi and Shandong, and Inner Mongolia Autonomous Region. More than 460 businesses with high emissions in Beijing were required to limit or stop their production during 3–12 November 2014 (Tang et al., 2015; Wang et al., 2016a; Guo et al., 2016). The number of private vehicles in operation over this period was reduced by about 50% through odd/even license-plate restrictions. Further, 9300 enterprises were suspended, 3900 enterprises were ordered to limit production, and more than 40,000 construction sites were shut down across the North China region (Wang et al., 2016b; Tang et al., 2015). The start-up of municipal winter heating systems was delayed until 15 November, after the summit. ~~Implementation~~ Previous studies report that implementation of these emission controls resulted in significant impacts on regional pollutant transport and local pollutant contributions (Meng et al., 2014; Sun et al., 2016b; Gao et al., 2017).

Previous model studies of the APEC period have adopted different estimates of the emission reductions imposed (Guo et al., 2016; Gao et al., 2017; Wen et al., 2016; Liu et al., 2017; Wang et al., 2017). The most detailed study of emission reductions considered application of controls in two distinct phases (Wen et al., 2016), and we have chosen to implement these controls in

Table 6. Emission controls during APEC period

Emission sector	Emission reduction (%)	
	Beijing	Other Districts
Industry	50	35
Power	50	35
Agriculture	40	30
Residential	40	30
Transport	40	30
PM coarse (all sectors)	80	–

APEC1: Beijing, Langfang, Baoding, Shijiazhuang, Xingtai, Handan
APEC2: APEC1 + Tangshan, Tianjin, Cangzhou, Hengshui, Dezhou, Binzhou, Dongying, Zibo, Jinan and Liaocheng

our study, as the emission reductions applied are consistent with observation-based assessments of regional emission controls (Li et al., 2017b). During the initial phase (APEC1, 3–5 November), emission controls were implemented in Beijing and the western side of the North China Plain. In a subsequent phase (APEC2, 6–12 November) controls were applied over a wider region including eastern Hebei and parts of Shandong. We represent these controls in the model over the districts shown in Fig. 7, following Li et al. (2017b), and neglect smaller changes in emissions in other districts and more distant provinces. Controls were applied across different activity sectors following Wen et al. (2016) and Li et al. (2017b), see Table 6.

Figure 8 shows the effect of these controls on key pollutants over the period 3–12 November. There is a minor pollution episode over 4–5 November, and the model underestimates $PM_{2.5}$ levels over this period ~~in the baseline run even without the~~ even without emission controls. This may partly reflect an underestimation of OC as the simulation of secondary inorganic aerosol for these two days is ~~relatively~~ good (see Fig. 5). $PM_{2.5}$ levels are very well matched in the period 6–9 November leading up to the summit when ~~applying the emission controls~~ emission controls are applied. PM_{10} levels are underestimated in the simulations, but this is ~~strongly~~ influenced by what may be a minor dust episode on 11–12 November, when coarse particles were high but $PM_{2.5}$ remained very low. Overall, the controls had a notable effect, reducing concentrations by 20–30% for all pollutants except O_3, which showed a small increase as expected ~~for lower~~ with reduced levels of NO. Over the critical 10–12 November meeting period, $PM_{2.5}$, PM_{10}, CO and NO_2 were reduced by 21%, 26%, 22% and 22% respectively, see Table 7. The reduction in $PM_{2.5}$ is very similar to the 22% found in previous studies (Gao et al., 2017). However, the absolute improvement in air quality over the meeting period was small, averaging less than 10 ~~m^{-3}~~ $\mu g\,m^{-3}$ for $PM_{2.5}$, reflecting the relatively clean conditions over the period. Average $PM_{2.5}$ in the baseline simulation was 39 ~~m^{-3}~~ $\mu g\,m^{-3}$, close to the observed 36 ~~m^{-3}~~ $\mu g\,m^{-3}$. Under these conditions the key air quality standard, a 24-hour averaged $PM_{2.5}$ of 75 ~~m^{-3}~~ $\mu g\,m^{-3}$, corresponding to a Chinese Air Quality Index (AQI) of 100, would have been met in the model simulation even without the controls.

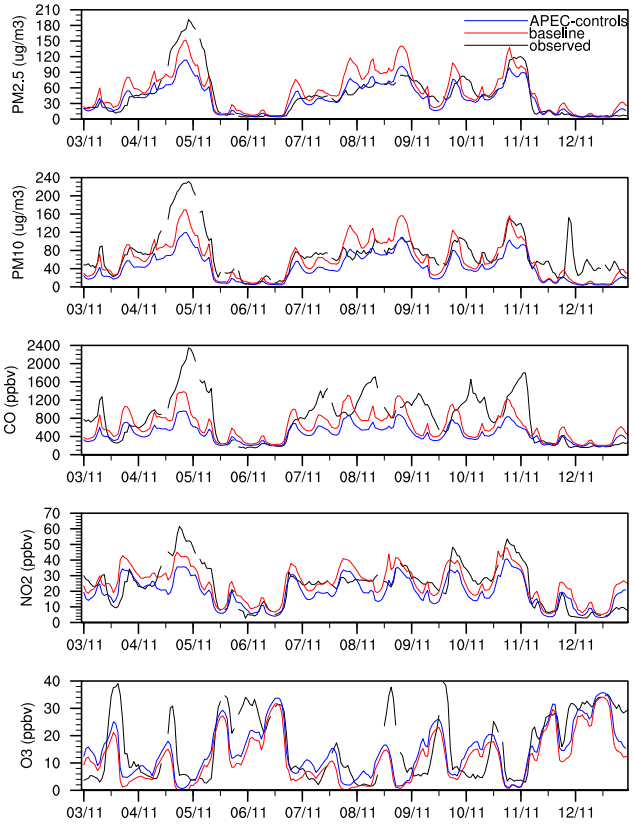

Figure 8. Time-series of surface pollutants averaged over the 12 measurement stations in Beijing during the APEC period.

To explore the importance of meteorological conditions in contributing to ~~the~~ favourable air quality during the APEC period, we apply the same magnitude, location and duration of emission controls to the major pollution episode at the end of October. Fig 9 shows the effect of these controls on key pollutants over 16–25 October. The controls reduced pollutant concentrations by a larger amount than during the APEC period, but the relative improvements of 23–38% were very similar. The absolute pollutant concentrations were much higher than in November. This can be attributed to lower wind speeds and to winds from the South and East bringing air from across the North China Plain, in contrast to the APEC period which experienced higher wind speeds and more frequent air from the clean northwest sector ~~.~~ (see Figure 2). The 3-day baseline average concentrations over 23–25 October for $PM_{2.5}$, PM_{10}, CO and NO_2 were 279 ~~m^{-3}~~ $\mu g\,m^{-3}$,310 ~~m^{-3}~~ $\mu g\,m^{-3}$, 1.48 ppm and 53 ppb respectively, substantially exceeding air quality standards. The difference in baseline $PM_{2.5}$ concentrations between the October and November periods without emission controls, 279 vs. 39 ~~m^{-3}~~ $\mu g\,m^{-3}$, highlights the dominant role played by meteorology in bringing clean air during APEC. The emission controls have a much larger absolute effect during the October episode than in the APEC period, with reductions in $PM_{2.5}$ of 65 ~~m^{-3}~~ $\mu g\,m^{-3}$ for 23–25 October, bringing average $PM_{2.5}$ levels down to 214 ~~m^{-3}~~ $\mu g\,m^{-3}$. However, this is insufficient to meet the standards needed for clean air of 75 ~~m^{-3}~~ $\mu g\,m^{-3}$. This indicates that

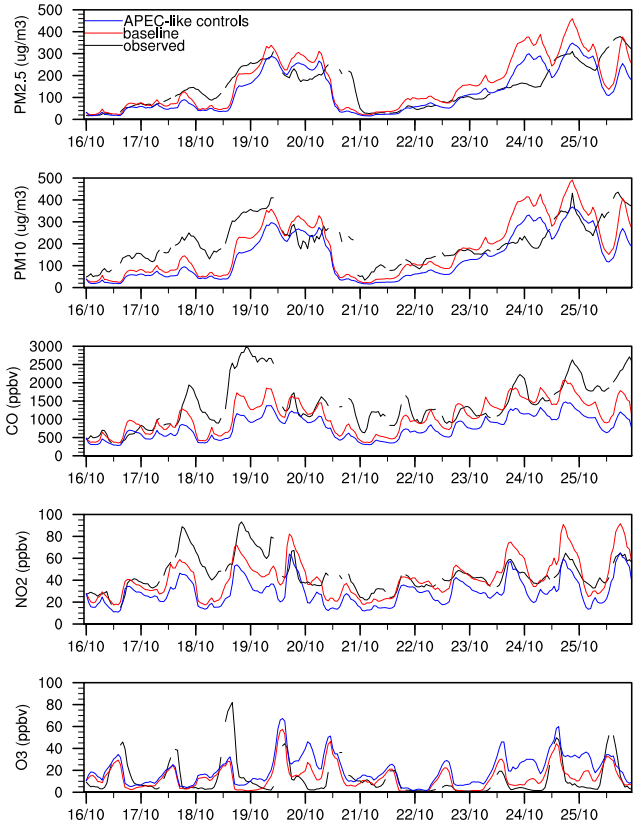

Figure 9. Time-series of surface pollutants averaged over the 12 measurement stations in Beijing during 16–25 October 2014.

the same emission control policies applied would have failed to produce the desired results if the meeting had been held at the end of October.

Table 8 presents the effect on aerosol components and gas-phase pollutants at the IAP tower. During the emission controls in both the polluted October and cleaner November periods, primary components were reduced by 31–34% while secondary components were reduced by only ~~3–18~~3–17%. This suggests that pollution episodes dominated by primary aerosols may be more easily controlled ~~. This~~ and has serious implications for winter haze episodes over the North China Plain because much of the increase in aerosol loading is contributed by regional secondary aerosols (see Sun et al., 2016b). The percentage reduction in SO_4 (14–17%) may be overestimated as some fraction of SO_4 mass for which chemical formation pathway remains unknown is treated as primary aerosol in the model. Similarly, the percentage reduction in OC may be overestimated because all OC is primary in the model.

To investigate the feasibility of meeting air quality standards during pollution episodes such as that on 21–25 October, we ran the model with all anthropogenic emissions removed over the North China Plain ~~region shown in Fig. 7~~ from 16–25 October. The 3-day average concentrations over 23–25 October showed substantial reductions: 83% for $PM_{2.5}$, 82% for PM_{10}, 79% for CO, 99% for NO_2 and 88% for SO_2. Average $PM_{2.5}$ concentrations were reduced from 279 ~~m^{-3}~~ $\mu g\,m^{-3}$ to 48 ~~m^{-3}~~ $\mu g\,m^{-3}$

, demonstrating that air quality standards can be met on highly polluted days, at least in theory, under the most stringent emission controls. From this simulation, and accounting for nonlinearity in secondary aerosol formation, we estimate that a 92% emission reduction over the 10 day period would have been needed to keep the average concentrations for 23–25th October below 75 ~~m^{-3}~~ $\mu g\,m^{-3}$. Even accounting for ~~the~~ model overestimation of average $PM_{2.5}$ during this period, driven principally by ~~a high~~ the positive bias on 24 October, we find that an 85% emission reduction would be required, substantially more than is feasible realistically. It is clear from this analysis that emissions controls would need to be applied over a much wider area over neighbouring provinces if the air quality standards in Beijing were to be met.

Finally, we analyse the full simulation period (12 October–19 November) to investigate how many days would meet the "blue-sky" criteria of 24-hour average $PM_{2.5}$ concentrations less than 75 ~~m^{-3}~~ $\mu g\,m^{-3}$ with and without ~~the controlsthat were applied. To remove any model bias we use the relative benefits of controls derived from the model simulations and apply these reductions to the daily observed~~ APEC-like controls. We conducted another simulation with APEC 2 controls implemented over the full period and found a reduction in daily average $PM_{2.5}$ ~~levels averaged over the 12 air quality stations in Beijing to generate a scenario representing the effect of emission controls. For 3–12 November, when emission controls were actually in place, we use the observed concentrations unaltered. We assume a reduction in~~ of ~~22%at other times, representing an average of the responses seen over the October and November periods discussed above. To evaluate the effect of differences between modelled and observed aerosol composition on this scaling, given that primary aerosols are reduced more efficiently than secondary aerosols (Table 8), we apply the modelled reductions for each component~~ 26±6%, and a reduction of 23±4% for haze days with daily mean $PM_{2.5}$ >75 $\mu g\,m^{-3}$. Since primary and secondary aerosol components can respond differently to emission controls, we use component-level fractional reductions from the model and apply them to the observed component concentrations to find the ~~total reduction in~~ reduction in total PM. This is ~~found to be~~ approximately 22% for both October and November periods based on our APEC control runs, suggesting that this scaling is appropriate and robust to uncertainties in model aerosol composition. ~~For the scenario with no controls , we~~ To generate an emission controls scenario over the full period, we reduce daily mean observed $PM_{2.5}$ concentrations by 22% for all days except 3–12 November when controls were actually in place. For this 3–12 November period we apply an increase of 16–33% based on the ~~November run of 16–33% for 3–12 November to estimate what the observations would have been, and use the observations on other days unaltered~~APEC controls run to represent conditions with no controls. With these scenarios we find that 15 of the 39 days considered failed to meet the blue-sky criteria of daily average $PM_{2.5}$ concentrations less than 75 ~~m^{-3}~~ $\mu g\,m^{-3}$ without controls, and this fell to 13 days when the controls were implemented, a modest decrease of 2 days, see Fig. 10. However, if we choose a higher threshold of 150 ~~m^{-3}~~ $\mu g\,m^{-3}$ (AQI of 200), the emission controls appear more effective, reducing the number of exceedances from 8 days to 5 days, and with a threshold of 200 ~~m^{-3}~~ $\mu g\,m^{-3}$ (AQI of 250) the number of exceedances falls from 4 days to 1 day.

To organize a three-day meeting such as APEC successfully, all three days must individually meet the chosen air quality criteria. We find that without emission controls, only 9 out of 37 possible three-day time slots in our simulation period meet the criteria, including only 3 out of the 8 available during the APEC period of 3–12 November. Under the emission controls, the meeting could have been organized on 14 out of the 37 slots, including all 8 during early November. This suggests that

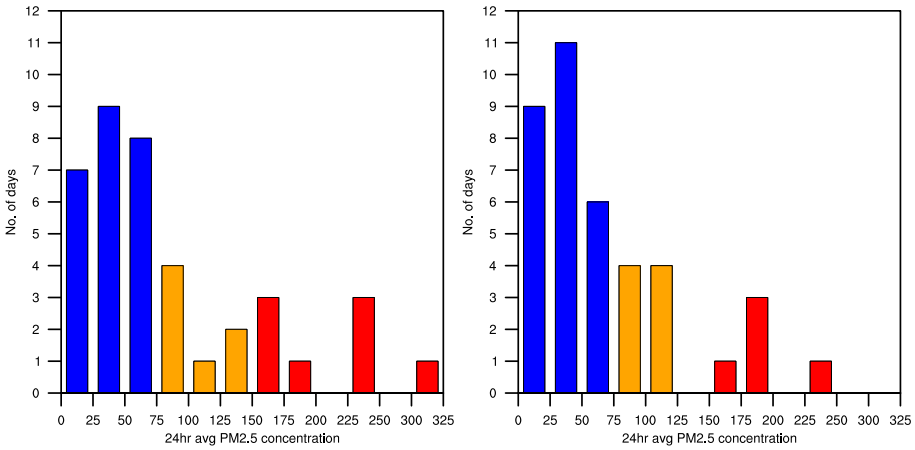

Figure 10. Frequency distribution of daily average $PM_{2.5}$ over 12 October–19 November 2014 showing the number of days meeting thresholds of 75 ~~m^{-3}~~ μg m^{-3} (blue) and 150 ~~m^{-3}~~ μg m^{-3} (blue plus orange) without (left panel) and with (right panel) emission controls.

the emission controls were only sufficient to provide an additional 5 time slots to hold a three-day event meeting the criteria. Interestingly, these all occur during the APEC period, highlighting that while favourable weather conditions were vital for meeting the air quality criteria, the emission controls provided critical support in achieving the 75 ~~m^{-3}~~ μg m^{-3} threshold needed to realise blue sky conditions. Specifically, in the absence of emission controls the first day of the APEC meeting (10 November) would have exceeded the air-quality standards. In this respect, it is reasonable to claim that the APEC emission controls were a success. However, it is clear that favourable meteorology was essential in making it possible for the emission controls to produce the marginal improvements needed to meet the air quality standards.

It should be noted that 23 out of the 37 possible three-day time periods ~~,~~(more than 60%~~,~~) would not have met the standards even under the emission controls applied. It is therefore clear that much more stringent controls are needed in future to counter the effect of unfavourable meteorological conditions. While greater reductions in the magnitude of emissions are required, it is important that these are applied over a much larger area, including in the neighbouring provinces that surround the North China Plain.

6 Conclusions

We have demonstrated that using a high-resolution nested air quality model we can reproduce the observed hourly variation of major pollutants in Beijing during October–November 2014 reasonably well. We capture the synoptic drivers of air quality well, including the build-up of pollutants during pollution episodes and the subsequent cleaning effect of winds from the northwest. The concentrations of $PM_{2.5}$, the dominant pollutant in this season, are reproduced well, and we show that where the model is biased high, typically during nighttime, underlying weaknesses in the treatment of turbulent mixing in the planetary boundary layer are often responsible. We show that use of two-way nesting to high resolution brings a substantial benefit in reproducing

Table 7. Influence of emission controls averaged over Beijing air quality stations in October and November

Species	Observed	Model		
	Mean	Baseline	Controls	Improvement
APEC period (10–12 November)				
$PM_{2.5}$ (~~m^{-3}~~ µg m^{-3})	36.1	39.3	31.1	8.2 (~~20.9~~21%)
PM_{10} (~~m^{-3}~~ µg m^{-3})	65.3	43.9	32.5	11.4 (~~26.0~~26%)
CO (ppm)	0.64	0.48	0.38	0.11 (~~22.0~~22%)
NO_2 (ppb)	19.0	20.6	16.0	4.6 (~~22.3~~22%)
SO_2 (ppb)	2.1	6.1	4.2	1.9 (~~30.8~~30%)
O_3 (ppb)	20.0	16.5	19.0	-2.5 (~~-15.3~~-15%)
October period (23–25 October)				
$PM_{2.5}$ (~~m^{-3}~~ µg m^{-3})	216.1	278.8	213.7	65.1 (~~23.3~~23%)
PM_{10} (~~m^{-3}~~ µg m^{-3})	263.8	309.6	236.4	73.2 (~~23.6~~24%)
CO (ppm)	1.77	1.48	1.05	0.44 (~~29.6~~30%)
NO_2 (ppb)	46.3	53.2	34.9	18.3 (~~34.4~~34%)
SO_2 (ppb)	4.0	18.6	11.6	7.0 (~~37.7~~38%)
O_3 (ppb)	11.4	15.2	26.7	-11.5 (~~-75.5~~-76%)

observed pollutant concentrations, even when comparing at the coarsest resolution used. Thorough evaluation against aerosol composition measurements over the period ~~highlight~~ highlights some weaknesses in representation of key aerosol components, particularly the balance between SO_4, NO_3 and NH_3 which requires more detailed analysis.

We show that short-term emission controls played a valuable role in improving air quality over the APEC period, but that their overall contribution was relatively small, with average reductions of 20–26% for key pollutants. Without the controls, average $PM_{2.5}$ levels are likely to have exceeded the national standard of 75 ~~m^{-3}~~ µg m^{-3} on 10 November, the first day of the APEC meeting, but the effects were largely incremental, highlighting the important role played by favourable meteorology during the period. If the APEC meeting had been held at a different time, particularly at the end of October, air quality standards would not have been achieved with the emission controls applied. We find that the relative effect of the controls during the pollution episodes of late October are very similar to those during the clean APEC period, averaging 23% for $PM_{2.5}$. Much greater emission reductions of at least 85% would have been needed over the North China Plain region to bring pollutant levels down to meet air quality standards. It is clear that under the stable meteorological conditions present during these pollution episodes much more stringent emission controls are needed than those that were applied, and that these need to be implemented over a much wider region of Northern China. Our study demonstrates the value of short-term emission controls, but highlights that long-term, sustained emission reductions on a regional scale are required to bring blue skies to Beijing.

Table 8. Influence of emission controls at the IAP site in October and November

Species	Observed	Model		
	Mean	Baseline	Controls	Improvement
APEC period (10–12 November)				
OC (~~m^{-3}~~μg m^{-3})	30.6	9.8	6.8	~~3.06 (31.1~~3.1 (31%)
BC (~~m^{-3}~~μg m^{-3})	3.4	4.8	3.2	~~1.63 (33.8~~1.6 (34%)
NO_3 (~~m^{-3}~~μg m^{-3})	10.9	8.6	8.3	~~0.27 (3.2~~0.3 (3%)
NH_4 (~~m^{-3}~~μg m^{-3})	5.0	3.8	3.5	~~0.30 (8.0~~0.3 (8%)
SO_4 (~~m^{-3}~~μg m^{-3})	4.8	3.5	2.9	~~0.60 (17.0~~0.6 (17%)
CO (ppm)	2.60	0.68	0.52	0.16 (~~24.0~~24%)
NO_2 (ppb)	17.2	30.2	22.9	~~7.36 (24.3~~7.4 (24%)
SO_2 (ppb)	10.4	9.4	6.3	~~3.07 (32.8~~3.1 (33%)
O_3 (ppb)	3.5	17.6	21.6	~~-4.04 (-23.0~~-4.0 (-23%)
October period (23–25 October)				
OC (~~m^{-3}~~μg m^{-3})	60.5	39.5	26.7	~~12.79 (32.4~~12.8 (32%)
BC (~~m^{-3}~~μg m^{-3})	10.2	16.5	11.0	~~5.49 (33.2~~5.5 (33%)
NO_3 (~~m^{-3}~~μg m^{-3})	51.3	95.0	79.7	~~15.32 (16.1~~15.3 (16%)
NH_4 (~~m^{-3}~~μg m^{-3})	21.1	35.2	29.9	~~5.40 (15.3~~5.4 (15%)
SO_4 (~~m^{-3}~~μg m^{-3})	31.2	18.4	15.8	~~2.53 (13.8~~2.5 (14%)
CO (ppm)	2.92	2.03	1.43	0.60 (~~29.5~~30%)
NO_2 (ppb)	44.2	78.1	57.8	~~20.30 (26.0~~20.3 (26%)
SO_2 (ppb)	18.4	26.5	16.5	~~9.93 (37.5~~9.9 (37%)
O_3 (ppb)	5.9	10.3	22.1	-11.8 (~~-114.7~~-115%)

Code and data availability. The WRF-Chem code is available from http://www2.mmm.ucar.edu/wrf/users/download/. The namelist for the model and surface pollutant distributions generated in this study are available from the Lancaster University data archive at http://dx.doi.org/10.15125/XXXXXX.

Author contributions. TA, OW and ZW designed this study, and TA performed the model simulations and analysis. JL provided emissions data and expertise on the model set-up, TY provided the lidar data and guidance on deriving PBL height, and YS, WX and ZW provided measurement data from the IAP tower. TA and OW prepared the manuscript with input from all coauthors.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. OW thanks the UK Natural Environment Research Council for support under grants NE/N006925/1 and NE/N006976/1. YS thanks the National Natural Science Foundation of China (Grant No. 91744207), and TY thanks the Strategic Priority Research Program of the Chinese Academy of Sciences (Grant No. XDA19040203).

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