

To Editor:

Sorry, I uploaded the wrong response file last time.

Thanks for these two minor corrections. We have corrected and changed the description in Table 1.

Thanks for these valuable comments to improve our manuscript. Our responses are in [blue](#).

Minor revisions:

I would like to thank the authors for their detailed and thorough response to the two reviewer comments and the short comment by Dr. Frank Dentener and for revising the manuscript accordingly. Before accepting the manuscript for final publication, I would like to see the following minor issues addressed:

1) A discussion of the author's choice to use CBM-Z/MOSAIC which lacks a treatment of SOA should be included in section 2.1 "WRF-Chem Model". The authors do discuss the lack of SOA treatment - which was noted by both reviewers – in section 3.1 and in the summary, but it would be better to include this information up front.

[Responses: Thanks for this great suggestion. We have added discussion in section 2.1. "CBMZ-MOSAIC has been proved to be capable of simulating air quality in many previous studies all over the world. Furthermore, the CBMZ-MOSAIC configuration in WRF-Chem enables us to include aerosol feedbacks with the meteorology in winter haze in a comprehensive manner. The current implementation does not include the secondary organic aerosol formation, and this limitation is discussed later in the paper"](#)

2) Response to points (2) and (3) raised by reviewer #2: I have to admit that I was also initially confused by the description of these meteorological perturbation simulations and also thought that the authors were describing counteracting feedback effects. After rereading sections 3.3 – 3.5 a few more times, I realized that this potential misunderstanding is a wording issue that has not been resolved in the revised manuscript and needs to be addressed before final publication.

Take the following sentences from page 16: "..., we decrease temperature by 2C in the initial and boundary conditions to reflect conditions more like 1960. As a result, the monthly domain mean surface temperature increases 2C (CTL – CTL_T2), ...". At first glance, these two sentences seem to suggest that decreasing temperatures in the IC/BC led to increases in simulated temperature, and this is the impression reviewer #2 and I both got. Upon re-reading the section, I realize what is meant here – temperatures were decreased from the 2010 met conditions used in CTL to represent 1960 as a sensitivity case, and consequently there is a temperature increase between CTL_T2 (representing 1960 met conditions) and CTL (representing 2010 met conditions). One way to clarify this might be to change these two sentences as follows "..., in the CTL_T2 simulation we decrease temperature by 2C in the initial and boundary conditions to reflect conditions more like those that occurred in 1960 rather than the 2010 conditions used in CTL. As a result of these changes, the monthly domain mean surface temperature increases 2C between CTL_T2 and CTL, ...".

In a sense, the root cause of the confusion is that the authors treat 1960 as a reference point for their discussion of changes in emissions and meteorology, but the “CTL” experiment does not reflect that reference point but rather relies on 2010 meteorology. This potential for confusion runs throughout sections 3.3 – 3.5 whenever the authors discuss “perturbations” since this term actually seems to refer to the approximated 1960 – 2010 meteorological change rather than the actual perturbation simulation which was a reverse meteorological change meant to “revert” the 2010 meteorological conditions in CTL to 1960 conditions. Examples: page 16, lines 13-16, page 18, lines 8-11, page 19, lines 2-6. I suggest that the author consider using wording like “Due to the approximated change in temperature/Rh/winds between 1960 and 2010” rather than “Due to the perturbation” in these instances.

Responses: Thanks for this great advice and we apologize for the misleading sentences. We have changed the sentences as you guided. We also use “Due to the approximated change in temperature/Rh/winds between 1960 and 2010” instead of “Due to the perturbation” to make it not confusing.

3) I assume that all emission and meteorological perturbations were only implemented in the innermost domain (except the boundary condition sensitivity simulation where emission were perturbed in the outermost domain). If so, please clarify this in section 2.2. If not, please clarify how the analysis nudging applied in the outermost domain was applied for the meteorological perturbation cases.

Responses: In fact, all emission and meteorological perturbations were implemented in both domains (innermost and outermost). The perturbations were applied in the analysis files, so analysis nudging process in the outermost domain includes the perturbations. We have added this clarification in section 2.2.

“In meteorological perturbation cases, the analysis nudging in outer domain includes perturbations in meteorological variables.”

4) Effect of chemical boundary conditions, page 11, lines 14 – 20. Please clarify that this test investigates changes due to boundary conditions only for the innermost domain, not the outermost domain. In other words, clarify that you are investigating the effects of regional emission changes in the 81 km domain on results in the 27 km domain and that your analysis does not account for the potential effects of changes in global atmospheric composition between 1960 and 2010 since the same 2010 MOZART simulation was used to derive boundary conditions for the outermost domain in all simulations. In addition, the boundary condition sensitivity simulation shown in Figure S2 should be added to Table 1.

Responses: Thanks for this suggestion. We added one sentence to clarify: “This investigation focuses on how emission changes in the outer domain affect results in the innermost domain, not accounting for the effects of global emission changes from (i.e., emissions from outside the outer domain) 1960 to 2010.”. We also added this sensitivity simulation in Table 1.

5) Section 3.2.4. As noted by reviewer #2 and acknowledged by the authors in the new sentence added at the end of this section, a main reason for why the results of the SO₂, NH₃ and NO_x emission sensitivity cases do not add up to the full difference between the CTL and EMI2010 cases is the change of other emissions such as VOC in the latter. Therefore, it is somewhat misleading to label this section “coupled changes in SO₂, NH₃ and NO_x emissions” – a better

choice might be “Comparison of individual changes in SO₂, NH₃ and NO_x emissions to simultaneous changes in all emissions” and to state up front that the latter case includes changes in emissions besides SO₂, NH₃ and NO_x.

Responses: Thanks for this great suggestion. We made changes in the title and added statement: “Here, we compare changes in major inorganic aerosols (i.e., sulfate, nitrate and ammonium) when SO₂, NH₃ and NO_x emissions are perturbed individually to results when all emissions (including non-methane hydrocarbon) are perturbed.”

6) As noted by reviewer #1, the PBL decreases associated with the 2C temperature increase are noteworthy. The authors responded that they agree with the reviewer’s suggestion and revised the manuscript to state that changes in vertical temperature profiles are causing the decrease in daytime PBL (“the monthly domain average daytime PBLHs decrease about 2.3% due to changes in temperature vertical profiles”). Please add more information on your analysis of how the vertical temperature profiles changed as a result of the uniform 2C increase in temperature initial and boundary conditions. Or do potential changes in wind speed induced by these temperature changes also play a role?

Responses: The 2C increase is not uniform (“temperature increases by 2.0 °C between CTL_T2 and CTL_T2, but in a non-uniform manner.”), which causes differences in PBLHs. The PBL heights highly depend on vertical structure of temperature and the temperature perturbation changes vertical structure of temperature, so PBL heights change. We have added this explanation in the revised manuscript: “PBLHs highly depend on vertical profiles of temperature, and the resulting non-uniform changes in temperature modify vertical profiles of temperature, so PBLHs change.” Yes, changes in wind speed also play a role. As we pointed out in the manuscript, “Surface horizontal winds also change (Figure 6(d)), which directly affect the distributions and magnitudes of PM_{2.5} concentrations in North China along with PBLH changes.”

7) The sensitivity simulation with the ECLIPSE_GAINS_4a emissions dataset discussed on pages 21/22 should be added to Table 1.

Responses: Thanks for mentioning this point. We have added this case in Table 1.

8) Section 3.6. If the authors have any thoughts on whether direct or indirect aerosol radiative effects are more important in driving these changes in PBL heights, it would be good to include this discussion here. This might include comparing changes in clear-sky and all-sky radiation and clouds. However, this is just a suggestion and not required since it is a relatively minor aspect of the overall study and may best be left for future work.

Responses: About the radiative effects on PBL heights, we discussed it in previous study (Gao et al., ACP, 2016). It is generally through (direct aerosol radiative effects) heating the atmosphere above the surface, cooling the air near surface, and thus inhibiting the evolution of PBL. We don’t think indirect effects play a significant role. As shown in the figure below, difference in liquid water path (integrated cloud water) is very small. In Zhang et al. (2015) also pointed that aerosol direct effect dominates over the NCP and the aerosol indirect effect is not significant. Yes, it would be interesting to compare changes in clear-sky and all-sky radiation in the future, especially the seasonal trends of these differences.

Zhang, B., Wang, Y., and Hao, J.: Simulating aerosol–radiation–cloud feedbacks on meteorology and air quality over eastern China under severe haze conditions in winter, *Atmos. Chem. Phys.*, 15, 2387–2404, doi:10.5194/acp-15-2387-2015, 2015.

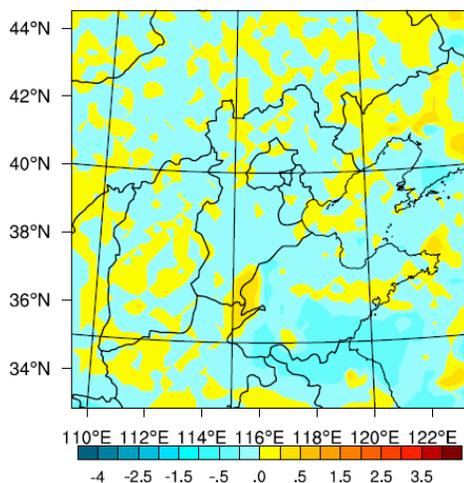


Figure Differences in LWP (g/m²) due to aerosol effects

9) While the manuscript is generally well written, it would benefit from careful proofreading for language and grammar. A few specific examples are page 7 line 22 “(i.g. SO₂-2010 NH₃-2010 NO_x-2010 cases)” which probably should be “(i.e. SO₂-2010, NH₃-2010, and NO_x-2010)”, page 8, line 1 “pointed out surface air temperature ... increased” which should be “pointed out that surface air temperature ... increased”, page 8, line 15 “for the January 2010 month” which should be “for the month of January 2010”, and page 8 line 20 “variations of surface temperature, RH” which should be “variations of surface temperature and RH”.

Responses: Thanks for pointing out these corrections. We have carefully read through the paper and made some minor corrections related to writing. For example, changed “how the PM_{2.5} concentrations change in response” to “how PM_{2.5} concentrations change in response”; changed “when modeling aerosol in China” to “when modeling aerosols in China”; changed “with MIX emission inventory” to “with the MIX emission inventory”; “changed “153% increase in BC emissions and 52% increase in OC emissions” to “154% increase in BC emissions and 54% increase in OC emissions”; etc.

1 **Response of winter fine particulate matter concentrations to**
2 **emission and meteorology changes in North China**

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1 **Abstract**

2 The winter haze is a growing problem in North China, but the causes have not been well
3 understood. The chemistry version of the Weather Research and Forecasting model (WRF-
4 Chem) was applied in North China to examine how the PM_{2.5} concentrations change in response
5 to changes in emissions (sulfur dioxide (SO₂), black carbon (BC), organic carbon (OC),
6 ammonia (NH₃), and nitrogen oxides (NO_x)), as well as meteorology (temperature, relative
7 humidity (RH), and wind speeds) changes in winter. From 1960 to 2010, the dramatic changes in
8 emissions lead to +260% increases in sulfate, +320% increases in nitrate, +300% increases in
9 ammonium, +160% increases in BC and 50% increases in OC. The responses of PM_{2.5} to
10 individual emission specie indicate that the simultaneous increases in SO₂, NH₃ and NO_x
11 emissions dominated the increases in PM_{2.5} concentrations. PM_{2.5} shows more notable increases
12 in response to changes in SO₂ and NH₃ as compared to increases in response to changes in NO_x
13 emissions. In addition, OC also accounts for a large fraction in PM_{2.5} changes. These results
14 provide some implications for haze pollution control. The responses of PM_{2.5} concentrations to
15 temperature increases are dominated by changes in wind fields and mixing heights. PM_{2.5} shows
16 relatively smaller changes in response to temperature increases and RH decreases, compared to
17 changes in response to changes in wind speed and aerosol feedbacks. From 1960 to 2010, aerosol
18 feedbacks have been significantly enhanced, due to higher aerosol loadings. The discussions in
19 this study indicate that dramatic changes in emissions are the main cause of increasing haze
20 events in North China, and long-term trends in atmospheric circulations maybe another
21 important cause since PM_{2.5} is shown to be substantially affected by wind speed and aerosol
22 feedbacks. More studies are necessary to get a better understanding of the aerosol-circulation
23 interactions.

1 **1 Introduction**

2 PM_{2.5} (particulate matter with diameter equal to or less than 2.5µm) is a main air pollution
3 concern due to its adverse effects on public health (Gao et al., 2015; Pope et al., 2009). Pope et
4 al. (2009) estimated that a decrease of 10µg PM_{2.5} is related to about 0.6 year mean life
5 expectancy increase. PM_{2.5} is also associated with visibility reduction and regional climate
6 (Cheung et al., 2005). Many cities in North China are experiencing severe haze pollution with
7 exceedingly high PM_{2.5} concentrations. In January 2010, a regional haze occurred in North China
8 and maximum hourly PM_{2.5} concentration in Tianjin was over 400µg/m³ (Zhao et al., 2013). In
9 January 2013, another unprecedented haze event happened, and the daily PM_{2.5} concentrations in
10 some areas of Beijing and Shijiazhuang reached over 500µg/m³ (L. T. Wang et al., 2014), and
11 instantaneous PM_{2.5} concentration at some urban measurement sites were over 1000µg/m³
12 (Zheng et al., 2015).

13 It is well known that particulate matter levels are strongly influenced by emissions and
14 meteorological conditions (Steiner et al., 2006). The PM in the atmosphere can be directly
15 emitted from sources like wildfires, combustion, wind-blown dust, and sea-salt, or formed from
16 emitted gases through secondary aerosol formation mechanisms. Meteorology affects PM levels
17 via changing emissions, chemical reactions, transport and deposition processes (Mu and Liao,
18 2014). For example, increasing wildfire emission in North America is mainly caused by warmer
19 temperatures and precipitation changes (Dawson et al., 2014), and increased temperature leads to
20 higher biogenic emissions, which are important precursors of secondary organic aerosols
21 (Dawson et al., 2014; Heald et al., 2008; Jacob and Winner, 2009). Increasing temperature also
22 increases sulfate concentration due to the temperature dependence of SO₂ oxidation and resulting
23 higher SO₂ oxidation rates (Aw and Kleeman, 2003; Dawson et al., 2007) and semi-volatile

1 aerosols may decrease due to evaporation under higher temperature (Sheehan and Bowman,
2 2001; Dawson et al., 2007; Tsigaridis and Kanakidou, 2007). Higher relative humidity (RH)
3 favors the formation of nitrate and increasing precipitation decreases all PM species via wet
4 scavenging (Dawson et al., 2007; Tai et al., 2010). Furthermore, increasing clouds promote in-
5 cloud sulfate production (Tai et al., 2010) and changes in wind speed and mixing height
6 determines the dilution of primary and secondary PM (Jimenez-Guerrero et al., 2012; Megaritis
7 et al., 2014; Pay et al., 2012).

8 With rapid economic and industrial developments, emissions in China have grown during the
9 past years. It is estimated that NO_x emissions in China increased by 70% from 1995 to 2004
10 (Zhang et al., 2007), Black Carbon (BC) by ~50% from 2000 to 2010 (Lu et al., 2011), Organic
11 Carbon (OC) by ~30% from 2000 to 2010 (Lu et al., 2011), and SO₂ by ~60% from 2000 to 2006
12 (Lu et al., 2011). Apart from emission changes, it was observed that the winter is warming up in
13 China, especially in the northern part (Guo et al., 2013; Hu et al., 2003; Ren et al., 2012). In
14 addition, wind speed in North China has lowered (Shi et al., 2015; Wang et al., 2004) and RH
15 has decreased in China (Song et al., 2012; Wang et al., 2004).

16 Many studies have investigated the impacts of emission changes on aerosol formation
17 (Aksoyoglu et al., 2011; Andreani-Aksoyoglu et al., 2008; Megaritis et al., 2013; Tsimpidi et al.,
18 2012a; Tsimpidi et al., 2012b) and the effects of climate/meteorology changes on PM_{2.5}
19 concentrations (Dawson et al., 2007; Megaritis et al., 2013; Megaritis et al., 2014; Tagaris et al.,
20 2007; Tai et al., 2012a; Tai et al., 2012b) in Europe and in the United States. The haze pollution
21 is growing in China, especially in North China, but the causes of the growth are not well
22 understood. For haze pollution in China, it has been reported that aerosol feedbacks that change
23 radiation and temperature can worsen pollution (Gao et al., 2016; Petřáček et al., 2016; Xing et al.,

1 2015c; Zhang et al., 2015). In addition, the connections between haze and meteorological
2 conditions have been established in many former studies (Fu et al., 2014; Jia et al., 2015; Leng et
3 al., 2015; C. Li et al., 2015; Wang and Chen, 2016; Yang et al., 2016; X. Y. Zhang et al., 2015;
4 Zhang et al., 2016). However, the roles of the large emission changes during the last 4 to 5
5 decades and the observed meteorology changes in North China are not known.

6 The main objective of this study is to investigate the responses of $PM_{2.5}$ and its major species to
7 changes in emissions, including SO_2 , BC, OC, NO_x and NH_3 , and to temperature, RH and wind
8 speed changes in North China region. Winter haze in North China has a large contribution from
9 secondary inorganic aerosols and secondary inorganic aerosols are influenced by emissions,
10 temperature and RH. The models used in previous studies of emissions and meteorology
11 perturbations referenced above are all offline models, which are not capable of considering the
12 feedbacks of changing meteorology on other meteorological variables, and the impacts of
13 aerosols on meteorology. However, as pointed by Gao et al. (2016) and J. Wang et al. (2014)
14 aerosol feedbacks should not be neglected when modeling aerosol in China. In this study, we
15 consider aerosol feedbacks using the fully online coupled WRF-Chem model.

16 This paper is organized as follows. First, the WRF-Chem model, model settings and domain
17 settings are briefly described and then in the next section, emission changes from 1960 to 2010
18 and accordingly $PM_{2.5}$ changes are discussed. After that, the responses of $PM_{2.5}$ to changes in
19 each emission species are analyzed. At last, the impacts of temperature, RH and wind speed
20 changes on $PM_{2.5}$ are analyzed and discussed.

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22

1 **2 Methodology**

2 **2.1 WRF-Chem model**

3 The WRF-Chem model is the chemistry version of the Weather Research and Forecasting model,
4 which is fully online coupled that allows gases and aerosols simulations at the same time as
5 meteorology simulations. In this study, we used a configuration that includes direct and indirect
6 feedbacks. The gas phase mechanism used in this study is the Carbon Bond Mechanism version
7 Z (CBM-Z), which includes 67 species and 164 reactions (Zaveri and Peters, 1999; Zaveri et al.,
8 2008). The gas-particle partitioning module used is the MOSAIC module, which considers all
9 important aerosol components, such as sulfate, nitrate, ammonium, BC, and OC (Zaveri et al.,
10 2008). Eight size bins version of MOSAIC was used and the aerosol sizes ranged from 0.039 μm
11 to 10 μm . CBMZ-MOSAIC has been proved to be capable of simulating air quality in many
12 previous studies all over the world. The current implementation does not include the secondary
13 organic aerosols formation. Wind-blown dust was modeled online using the AFWA scheme.
14 Two nested domains with 81km and 27km horizontal grid resolutions from outer to innermost
15 and 27 vertical grids were used (Figure S1 in supplementary material). Analysis nudging of
16 meteorology variables was used for the outer domain. In meteorological perturbation cases, the
17 analysis nudging in outer domain includes perturbations in meteorological variables. The model
18 physics configurations generally follow the settings in Gao et al. (2016). Inputs into the model
19 include meteorological boundary and initial conditions (BCs and ICs) from NCEP FNL 1° \times 1°
20 data and chemical boundary and initial conditions from MOZART model simulations (Emmons
21 et al., 2010). Chemical BCs and ICs are not changing along with the sensitivity simulations, but
22 the studied domain (innermost domain) takes boundary conditions from outer main, where
23 emissions are perturbed. The anthropogenic emission inventory used is the MACCity

1 (MACC/CityZEN EU projects) emissions dataset, which provides monthly CO, NO_x, SO₂, VOC,
2 BC, OC, and NH₃ emissions from different sectors for years between 1960 and 2020 (Granier et
3 al., 2011). We compared the MACCity emission inventory for 2010 (Granier et al., 2011) with
4 MIX emission inventory for 2010 (M. Li et al., 2015) in the China region, and the magnitudes of
5 emissions in China from these two datasets are very close. For example, the SO₂ emissions in
6 China in 2010 were estimated to be 28663 Gg in the MIX emission inventory, and were 26876.3
7 Gg in the MACCity emission inventory. Simulations for evaluating roles of emission changes
8 were conducted using emissions for year 1960 and year 2010. We assigned emissions to the first
9 6 layers from surface based on sectors. For example, emissions from large point sources (such as
10 chimneys) were assigned to higher layers. Biogenic emissions were estimated online using the
11 MEGAN model (Guenther et al., 2006). The simulation period was January 2010 and five days
12 in previous month were modeled as spin-up to overcome the influences of initial conditions.

13

14 **2.2 Sensitivity experiments**

15 We explored the sensitivities of the winter time haze event in 2010 to changes in emissions and
16 meteorology features through a series of simulations using 1960 and 2010 emission baselines.
17 Specifically, the influences of emission changes of SO₂, BC, OC, NH₃, and NO_x, and
18 meteorology (temperature, RH and wind speeds) changes on PM_{2.5} and its major species were
19 evaluated using a series of simulations. They are listed and explained in Table 1. All base
20 simulations use meteorology of January 2010. CTL case uses emissions for year 1960 and
21 EMI2010 case uses emissions for year 2010. SO₂, NH₃, and NO_x emissions were perturbed
22 separately from 1960 to 2010 (i.g., SO₂-2010, NH₃-2010, and NO_x-2010 cases). In the CTL_NF

1 and EMI2010_NF cases, aerosol-radiation interactions are excluded based on emissions for year
2 | 1960 and 2010. It was pointed out that surface air temperature in North China increased at the
3 rate of 0.36 °C per decade (Guo et al., 2013), the linear trends coefficient of relative humidity
4 anomaly in North China is about -0.60% per decade (Wang et al., 2004), and national mean wind
5 speed decreased 16% in the recent 50 years (Wang et al., 2004). To estimate the impacts of
6 changes in temperature, RH and wind speed that happened in the past several decades, we
7 decreased temperature by 2 degrees, increased RH by 10%, and increased wind speeds by 20%,
8 to reflect conditions of early decades (CTL_T2, CTL_RH10, CTL_WS20, EMI2010_T2,
9 EMI2010_RH10, and EMI2010_WS20 cases). These were conducted by perturbing the initial
10 and boundary conditions of these individual meteorological variables.

11 At different vertical heights, emission and meteorological variables were uniformly perturbed.
12 The changes of PM_{2.5} and its major components due to perturbations in emissions and
13 meteorology are analyzed for the North China region. The North China region is defined using
14 the innermost domain (shown in Figure S1) and the statistics of changes are calculated within
15 | this domain for the month of January 2010-~~month~~.

16

17 **2.3 Model Verification**

18 The WRF-Chem model performance has been evaluated using multiple observations, including
19 surface meteorological, chemical and optical data, and satellite data in Gao et al. (2016). The
20 | model was shown to capture the variations of surface temperature and, RH, while wind speed
21 was slightly overestimated (Gao et al., 2016), which has been reported as a common problem of
22 current WRF-Chem model under low wind speed conditions. The Root Mean Square Error

1 (RMSE) of temperature were all less than 3.2K and RMSEs of RH varied from 6.4 to 11.1%.
2 The RMSE of wind speeds were below the proposed criteria (2m/s) (Emery et al., 2001) at the
3 Beijing, Tianjin and Baoding stations, but larger than that criteria at the Chengde station. The
4 time series of simulated surface PM_{2.5}, NO₂, and SO₂ showed good agreement with observations
5 as did simulated aerosol optical depth (AOD) (Gao et al., 2016). Mean Fractional Bias (MFB)
6 ranged from -21.8% to 0.4% and Mean Fractional Error (MFE) ranged from 26.3% to 50.7%
7 when comparing against PM_{2.5} observations (Gao et al., 2016). In addition, the comparison
8 between model results and satellite found that the vertical distribution of aerosol and horizontal
9 distribution were captured well by the model (Gao et al., 2016). Compared with observed PM_{2.5}
10 composition, sulfate and OC were underestimated and nitrate was overestimated by the model
11 (Gao et al., 2016). The underestimation of sulfate may be due to underestimation of SO₂ gas
12 phase oxidation, errors in aqueous-phase chemistry, and/or missing heterogeneous sulfate
13 formation (Gao et al., 2016).

14

15 **3 Results and Discussion**

16 **3.1 PM_{2.5} sensitivity to emission changes from 1960 to 2010**

17 The emission changes of SO₂, NO_x, NH₃, BC and OC and resulting impacts on PM_{2.5} from 1960
18 to 2010 were examined based on the MACCity dataset for years 1960 and 2010. Figure 1(a-e)
19 displays SO₂, NO_x, NH₃, BC and OC emissions for 1960 and Figure 1(f-j) shows the changes
20 from 1960 to 2010. Populated regions of North China, such as urban Beijing, urban Tianjin, and
21 urban Shijiazhuang, exhibit large emissions of SO₂, NO_x, NH₃, BC and OC in 1960. However,
22 NH₃ emissions exhibit different spatial distribution patterns from SO₂, NO_x, BC and OC

1 emissions, because NH_3 is mainly associated with agriculture while SO_2 , NO_x , BC and OC are
2 mainly related with industrial and residential activities. From 1960 to 2010, SO_2 , NO_x , NH_3 , BC
3 and OC increased over the entire North China domain and markedly increased in the Jing-Jin-Ji
4 city cluster. In general, the domain averaged SO_2 emissions and NO_x emissions in North China
5 increased by ~220% and ~990% from 1960 to 2010, respectively. The domain averaged NH_3
6 emissions in North China increased by ~390% from 1960 to 2010, but the most significant
7 increases occurred not in the Jing-Jin-Ji city cluster, but in Inner Mongolia. Unlike NH_3
8 emissions, BC emissions increased the most in urban Beijing from 1960 to 2010. This is because
9 residential sources are the biggest contributor to BC in winter (Li et al., 2016) and the population
10 in urban Beijing sharply increased with rapid urbanization. From 1960 to 2010, the mean BC
11 emissions in North China increased by ~154%. Similar to BC emissions, OC emissions increased
12 substantially in the center of Beijing, and the domain averaged increasing ratio is about 54%
13 from 1960 to 2010. The enhancements of SO_2 , NO_x , NH_3 , BC and OC emissions in North China
14 are expected to result in substantial increase in regional $\text{PM}_{2.5}$ concentrations.

15 Figure 2 shows the simulated monthly mean concentrations of $\text{PM}_{2.5}$ and its major components
16 (sulfate, nitrate, ammonium, BC and OC) based on emissions for year 1960. As listed in Table 2,
17 the domain averaged concentrations of sulfate, nitrate, ammonium, BC, OC, and $\text{PM}_{2.5}$ are 1.9,
18 0.8, 0.8, 1.5, 4.6, and $19.2\mu\text{g}/\text{m}^3$, respectively. For year 1960, $\text{PM}_{2.5}$ concentrations are mainly
19 dominated by sulfate, OC and natural dust (the difference between $\text{PM}_{2.5}$ and the sum of sulfate,
20 nitrate, ammonium, BC, OC). Figure 3 displays the changes of sulfate, nitrate, ammonium, BC,
21 OC, and $\text{PM}_{2.5}$ due to changes in SO_2 , NO_x , BC and OC emissions from 1960 to 2010. The
22 predicted monthly mean concentrations of $\text{PM}_{2.5}$ components and $\text{PM}_{2.5}$ increase everywhere
23 over the entire domain due to emission changes resulting from the rapid urbanization and

1 industrialization from 1960 to 2010 (Figure 3(a-f)). As listed in Table 2, the predicted monthly
2 domain mean sulfate increases the largest ($5.0 \mu\text{g}/\text{m}^3$), followed by nitrate ($2.6 \mu\text{g}/\text{m}^3$) and OC
3 ($2.5 \mu\text{g}/\text{m}^3$).

4 From 1960 to 2010, the predicted BC increased by $\sim 157\%$ and OC increased by $\sim 54\%$ due to
5 153% increase in BC emissions and 52% increase in OC emissions. The nearly linear response of
6 both BC and OC aerosols to their emissions is due to the omission of a secondary organic aerosol
7 formation in the chosen CBMZ/MOSAIC mechanism. Thus, both of them were treated as
8 primary aerosols in these simulations. Our previous analyses indicate that SOA contribution in
9 this time period was small (Gao et al., 2016). The domain mean $\text{PM}_{2.5}$ concentrations increased
10 by $14.7 \mu\text{g}/\text{m}^3$ and the domain maximum increase is about $45 \mu\text{g}/\text{m}^3$ (Figure 3(f) and Table 2).

11 To quantify how much of the changes in Figure 3 are from the impacts of boundary conditions,
12 we simulated another case ([BCs 1960-2010](#)) with the innermost domain emissions fixed in 1960
13 and the outer domain emissions changed from 1960 to 2010. [This investigation focuses on how](#)
14 [emission changes in the outer domain affect results in the innermost domain, not accounting for](#)
15 [the effects of global emission changes from 1960 to 2010.](#) The impacts of boundary conditions
16 mostly occur around the south boundary and show nearly no impact on $\text{PM}_{2.5}$ in Beijing (shown
17 in Figure S2), which are consistent with the continuous weak southerly winds during the study
18 period (Gao et al., 2016). On domain average, the impacts of boundary conditions result in
19 $5.0 \mu\text{g}/\text{m}^3$ increase in the study domain, accounting for about 33.9% of the total changes in
20 $\text{PM}_{2.5}$.

21 To explore how emission changes can affect haze days, we calculated the number of haze days in
22 urban Beijing for the CTL and EMI_2010 cases, using daily mean thresholds of 35 and $75 \mu\text{g}/\text{m}^3$

1 (China National Ambient Air Quality Grade I and Grade II Standard, L. T. Wang et al., 2014). In
2 urban Beijing, there are 4 days when daily mean $PM_{2.5}$ concentrations are above $35\mu\text{g}/\text{m}^3$, and 0
3 days with daily mean $PM_{2.5}$ concentrations above $75\mu\text{g}/\text{m}^3$ for the CTL case. For the EMI_2010
4 case, these two numbers increase to 15 and 8, indicating that the large increases in emissions
5 over the past several decades have significantly affected haze occurrences in Beijing.

6

7 **3.2 Sensitivity to changes in individual emission species**

8 The results discussed above show that in the winter period, the concentrations of secondary
9 inorganic aerosols (sulfate, nitrate, and ammonium) has increased dramatically. Thus it is
10 important to explore how sensitive secondary inorganic aerosol is to perturbations in precursor
11 emissions. Three sensitivity simulations (change SO_2 , NH_3 and NH_3 emissions separately) were
12 conducted to examine how changes in emissions of each species affect aerosol concentrations.
13 The predicted changes of $PM_{2.5}$ and major $PM_{2.5}$ components at the ground-level are shown in
14 Figure 4 and monthly domain mean aerosol changes are summarized in Table 3.

15

16 **3.2.1 Changes in SO_2 emissions**

17 Due to changes in SO_2 emissions from 1960 to 2010, domain averaged sulfate increase by
18 $3.4\mu\text{g}/\text{m}^3$ (178.3%), nitrate decreases by $-0.3\mu\text{g}/\text{m}^3$ (-32.3%), and ammonium increases by
19 $0.2\mu\text{g}/\text{m}^3$ (29.4%). NH_3 reacts preferentially with SO_2 (Seinfeld and Pandis, 2006) and free NH_3
20 reacts with enhanced H_2SO_4 due to increasing SO_2 . As a result, ammonium increases and less
21 HNO_3 gas is transferred to the aerosol phase, which is consistent with the responses to increasing
22 SO_2 emissions in Kharol et al. (2013).

1

2 3.2.2 Changes in NH₃ emissions

3 As shown in Figure 4 and Table 3, changes in NH₃ emissions from 1960 to 2010 result in
4 significant increases in nitrate (1.5µg/m³, +76.0%) and ammonium (0.6µg/m³, +84.0%). The
5 domain mean changes of sulfate due to increase in NH₃ is close to zero (about 0.1µg/m³),
6 because sulfate formation is only indirectly associated with NH₃ availability (Tsimpidi et al.,
7 2007). The significant changes in nitrate and ammonium occurred in south Hebei, Shandong, and
8 Henan province, where anthropogenic NO_x emissions are very high (Figure 1). Although NH₃
9 emissions substantially increased in Inner Mongolia (Figure 1), responses of nitrate and
10 ammonium are not significant there due to trivial NO_x emissions. The substantial increases of
11 nitrate after NH₃ emission increase indicate that NH₃ limits the NH₄NO₃ formation in North
12 China region in this period.

13

14 3.2.3 Changes in NO_x emissions

15 After changing NO_x emissions from 1960 to 2010 levels, domain mean surface PM_{2.5} decreases
16 by about 0.2µg/m³, but the changes of individual PM_{2.5} inorganic components vary. The increase
17 of NO_x emissions cause 0.7µg/m³ (-39.1%) decrease in monthly domain mean sulfate and the
18 domain peak sulfate reduction is about 2.9µg/m³. The OH radical is critical in the sulfate
19 formation in the regions where SO₂ concentrations are high and there is a competition between
20 NO_x and VOCs to react with OH (Tsimpidi et al. 2012b). When the VOCs/NO_x concentration
21 ratio is close to 5.5:1, the OH reacts with NO_x and VOCs at an equal rate (Seinfeld and Pandis,
22 2006). When the concentration ratio is lower than 5.5:1, the OH primarily reacts with NO_x, and

1 the region with this concentration ratio is called VOC-limited region. In VOC-limited regions, an
2 increase of NO_x will cause a decrease of OH and ozone concentration. When the VOCs/NO_x
3 concentration ratio is higher than 5.5:1, the OH will preferentially react with VOCs, and the
4 region with this high ratio is called NO_x-limited region. In the NO_x-limited region, an increase of
5 NO_x will increase OH and ozone concentrations. In the simulated winter month, biogenic
6 emissions are low and NO_x emissions in North China are very high, leading to lower VOCs to
7 NO_x ratios, and it can be considered as VOC-limited region. Fu et al. (2012) pointed out that
8 north East Asia is VOC-limited in January and urban areas of Beijing are VOC-limited in both
9 January and July. The model averaged VOCs/NO_x ratio changes from 4.2 to 1.2 due to emission
10 perturbations from 1960 to 2010 (shown in Figure S3). As a result, the large increases in NO_x
11 emissions from 1960 to 2010 result in a 47.9% decrease in daytime surface ozone concentration
12 and 55.6% decrease in daytime surface OH concentration, which are shown in Figure 5. Over the
13 entire domain, ozone and OH decrease due to NO_x emission increases (Figure 5). Consequently,
14 sulfate aerosol decrease over the entire domain, as shown in Figure 4(i). Decreases in sulfate
15 might also be related to changes in thermodynamics of the ammonium-sulfate-nitrate system.
16 Although OH decreases, nitrate still rises (0.6μg/m³, +76.0%) due to the increase in NO_x
17 emissions. The domain mean ammonium decreases by about 5.1% (-0.04μg/m³). The net effects
18 of NO_x emission increases bring about 0.2μg/m³ decrease in monthly domain mean PM_{2.5}
19 concentration and the domain peak decrease is about 1.1μg/m³ (Table 3).

20

21 3.2.4 Comparison of individual changes in SO₂, NH₃ and NO_x emissions to simultaneous
22 changes in all emissions ~~Coupled changes in SO₂, NH₃ and NO_x emissions~~

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1 In this paragraph, we compare changes in major inorganic aerosols (i.e., sulfate, nitrate and
2 ammonium) resulted from individual changes in SO₂, NH₃ and NO_x emissions to changes
3 resulted from changes in all emissions. As shown above, increasing SO₂ emissions significantly
4 increases PM_{2.5} concentrations in North China region, increasing NH₃ emissions also increases
5 PM_{2.5} concentrations but to a lesser extent, and increasing NO_x emissions slightly decreases
6 PM_{2.5} concentrations. The effects of coupled changes in SO₂, NH₃ and NO_x emissions are not a
7 simple addition of the effect of changing them separately. As listed in Table 3, the monthly
8 domain mean sulfate, nitrate, ammonium, and PM_{2.5} increases more than the effects of changing
9 emissions separately. Domain mean sulfate increases by 5.0µg/m³ (+264.0%), nitrate increases
10 by 2.6 µg/m³ (+322.5%), ammonium increases by 2.3µg/m³ (295.2%) and PM_{2.5} increases by
11 9.9µg/m³ due to coupled changes in SO₂, NH₃ and NO_x emissions from 1960 to 2010. The
12 simultaneous increases in SO₂, NH₃ and NO_x emissions promote dramatic increases of secondary
13 inorganic aerosols in North China, which is partially due to perturbations in VOCs and other
14 species at the same time

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15

16 3.2.5 Changes in BC and OC emissions

17 Since BC and OC are treated as primary aerosols in the chosen CBMZ/MOSAIC mechanism,
18 changes in their emissions do not show any impact on other aerosol components. As listed in
19 Table 3, monthly domain mean PM_{2.5} increases by 2.3µg/m³ and 2.5µg/m³ due to changes in their
20 emissions from 1960 to 2010, respectively.

21

22 3.3 Effects of temperature increases

1 The model used in this study is a fully online-coupled model, which simulates meteorological
2 variables and chemical variables together. Therefore, it is not possible to increase temperature
3 uniformly, as was done in previous studies using offline models (Dawson et al., 2007; Megaritis
4 et al., 2013; Megaritis et al., 2014). To examine the sensitivity of PM_{2.5} to temperature change
5 (reflecting the winter warming trends), in the CTL_T2 simulation we decrease temperature by
6 2 °C in the initial and boundary conditions to reflect conditions more like those that occurred in
7 1960 rather than the 2010 conditions used in CTL. As a result of these changes, the monthly
8 domain mean surface temperature increases by 2.0 °C (between CTL_T2 and -CTL_T2), but in a
9 non-uniform manner. These responses in domain temperature are partially due to aerosol
10 feedbacks. The spatial distributions of monthly mean surface temperature and temperature
11 changes are shown in Figure 6(a). The monthly mean surface temperature increases more along
12 top left domain boundaries and less over the Bohai sea. The influence of increasing temperature
13 on biogenic emissions is included using temperature-sensitive biogenic emission model MEGAN
14 (Guenther et al., 2006).

15 Due to the perturbation-approximated change in temperature between 1960 and 2010 as
16 mentioned above, sulfate, nitrate, ammonium and PM_{2.5} are predicted to increase in most areas of
17 the domain (Figure 7). Predicted monthly mean sulfate increases by 0.06µg/m³ (+3.1%), nitrate
18 increases by 0.03µg/m³ (+4.2%), and ammonium increases by 0.02µg/m³ (+2.8%). The increases
19 of sulfate, nitrate and ammonium are mostly attributed to the increasing OH radicals, as shown in
20 Figure 6(b). After the temperature-perturbation-approximated change in temperature between
21 1960 and 2010, daytime OH increases by about 3.6% on domain average. It was found that
22 higher temperature increased volatilization of ammonium nitrate and partitioned it to the gas
23 phase (Megaritis et al., 2014), but it is not significant here due to the low temperature in winter.

1 In addition, the increase of sulfate, nitrate, and ammonium could be partially due to accelerated
2 gas-phase reaction rate at higher temperature (Dawson et al., 2007; Megaritis et al., 2014). It may
3 also due to enhanced photolysis caused by decreases in cloudiness after approximated change in
4 temperature between 1960 and 2010~~temperature perturbations~~ (as shown in Figure S4: changes
5 in liquid water path).

6 As shown in Figure 7 (d-e), the concentrations of primary aerosols (BC and OC) also increase
7 after the approximated change in temperature between 1960 and 2010~~temperature perturbations~~.
8 This is due to changes in other physical parameter, such as wind direction, wind speed, and
9 PBLHs, which are key factors in the diffusion of air pollutants. Figure 6(c) shows that monthly
10 PBLHs in most North China areas decrease after the approximated change in temperature
11 between 1960 and 2010~~temperature perturbation~~, and PBLHs over the Bohai sea decrease the
12 most, with monthly mean decrease over 50 meters. The monthly domain average daytime
13 PBLHs decrease about 2.3% due to changes in temperature vertical profiles. PBLHs highly
14 depend on vertical profiles of temperature, and the resulting non-uniform changes in temperature
15 modify vertical profiles of temperature, so PBLHs change. Surface horizontal winds also change
16 (Figure 6(d)), which directly affect the distributions and magnitudes of PM_{2.5} concentrations in
17 North China along with PBLH changes.

18
19 The responses of PM_{2.5} concentrations to approximated change in temperature between 1960 and
20 2010~~temperature perturbation~~ are different from the responses of sulfate, nitrate, ammonium,
21 BC and OC (Figure 7), with decreases in northwestern regions and increases in most areas of the
22 North China Plain. This is because natural dust is dominant in northwestern regions (as shown in
23 Figure 2(f)), and the concentrations of natural dust decrease under lower horizontal wind speeds

1 (Figure 6(d)). The monthly $PM_{2.5}$ concentration decreases by $0.01\mu\text{g}/\text{m}^3$ on domain average due
2 to ~~the approximated change in temperature between 1960 and 2010~~~~temperature perturbation~~.
3 Because of temperature increase, the numbers of haze days (defined using the daily mean
4 threshold 35 and $75\mu\text{g}/\text{m}^3$) in urban Beijing do not change.
5 The discussions shown above are based on emission levels in 1960. The responses to ~~the~~
6 ~~approximated change in temperature between 1960 and 2010~~~~temperature perturbations~~ were also
7 investigated based on emission levels in 2010, and the results are shown in Figure S5, S6 and
8 Table 3. The spatial distributions of the changes are similar to the results shown above, but with
9 larger magnitudes. The domain mean PBL heights decreases slightly more (-8.6 compared to -8.3
10 meters). The domain mean $PM_{2.5}$ concentrations and $PM_{2.5}$ components exhibit larger increases
11 in North China, although daytime OH concentrations increases less (2.6×10^{-9} compared to
12 3.3×10^{-9} ppmv), suggesting that the responses of $PM_{2.5}$ concentrations are mostly due to changes
13 in PBL heights and wind fields.

14

15 **3.4 Effects of RH decreases**

16 The RH was enhanced by 10% in model initial and boundary conditions ~~in CTL_RH10~~ to
17 represent RH for the previous decades. As a result, the simulated monthly mean RH decreases by
18 9.3% on domain average ~~between CTL_RH10 and CTL~~. Due to ~~RH perturbation~~~~the~~
19 ~~approximated change in RH between 1960 and 2010~~, domain mean $PM_{2.5}$ concentration
20 decreases by $0.7\mu\text{g}/\text{m}^3$. As shown Figure 8(a), $PM_{2.5}$ concentrations decrease in the Jing-Jin-Ji
21 region but increase in southern areas of the domain. The ammonium nitrate formation
22 equilibrium depends on RH (Tai et al., 2010), so HNO_3 may be shifted to the gas phase under
23 lower RH. In addition, the changes in RH can also affect the wet deposition rate. The increases in

1 southern areas of the domain are mainly due to suppressed in-cloud scavenging, as the decreases
2 in RH inhibit the formation of clouds. As shown in Figure 8(b), liquid water path (LWP)
3 decreases by 75.0%. As a result, the in-cloud scavenging loss rate decreases. The changes of
4 predicted aerosol optical depth at 600nm are shown in Figure 8(c). In most regions, visibility
5 decreases due to lower RH. Because of RH decreases, the numbers of haze days (defined using
6 the daily mean threshold 35 and 75 $\mu\text{g}/\text{m}^3$) in urban Beijing do not change. The responses to the
7 approximated change in RH between 1960 and 2010~~RH-perturbations~~ were also investigated
8 based on emission levels in 2010, and the results are shown in Figure S7 and Table 3. The
9 responses are also similar to changes based on emission levels in 1960, but with larger
10 magnitudes.

11

12 **3.5 Effects of wind speed decreases**

13 Simulations were also carried out when wind speeds in initial and boundary conditions were
14 increased in CTL WS20 to estimate the wind speeds for the previous decades. The predicted
15 domain averaged monthly mean wind speed decreases by about 0.7 m/s between CTL WS20
16 and CTL. As shown in Figure 9(a), the monthly mean near surface horizontal winds are
17 pronounced in mountainous areas (northwest areas of the domain) and relatively smaller in other
18 areas. Figure 9(b) shows the changes of wind speeds (CTL-WS20) due to model perturbations.
19 The predicted monthly mean $\text{PM}_{2.5}$ concentrations decrease by 2.3 $\mu\text{g}/\text{m}^3$ on domain average, but
20 the responses of $\text{PM}_{2.5}$ vary within the domain. As shown in Figure 9(c), $\text{PM}_{2.5}$ concentrations
21 decrease in the northwestern areas because of lower production of natural dust under lower
22 horizontal wind speeds. However, in most areas of the North China Plain, $\text{PM}_{2.5}$ concentrations
23 increase under lower wind speeds (Figure 9(c)). The domain peak increase is about 2.4 $\mu\text{g}/\text{m}^3$,

1 which is based on low predicted $PM_{2.5}$ concentrations using emissions for year 1960. If the
2 concentration in base case is higher, the responses will be enhanced. As shown in Figure 9(d),
3 the domain maximum increases in $PM_{2.5}$ increases from 2.4 to $9.4 \mu\text{g}/\text{m}^3$. Because of wind speed
4 decreases, number of haze days that daily mean $PM_{2.5}$ concentrations are above $35 \mu\text{g}/\text{m}^3$
5 increases by 1.

6

7 **3.6 Effects of changes in aerosol feedbacks**

8 As mentioned in Gao et al. (2016), high concentrations of aerosol enhance stability of boundary
9 layer and increase $PM_{2.5}$ concentrations. Due to dramatic changes in emissions from 1960 to
10 2010, the strength of aerosol feedbacks may also have changed. To quantify these changes, we
11 simulated four cases (i.e., CTL, CTL_NF, EMI2010, and EMI2010_NF). CTL-CTL_NF and
12 EMI2010-EMI2010_NF are used to represent the contributions of aerosol radiative effects in
13 1960 and 2010. The changes in monthly mean daytime PBL heights and $PM_{2.5}$ concentrations are
14 shown in Figure 10. In 1960, the domain averaged PBL height decreases by 6.7 meters due to
15 aerosol radiative effects, and the domain maximum decrease is 25.4 meters. Correspondingly, the
16 domain averaged $PM_{2.5}$ increases by $0.1 \mu\text{g}/\text{m}^3$ and the domain maximum increase is $0.9 \mu\text{g}/\text{m}^3$. In
17 2010, the domain averaged PBL height decreases by 13.8 meters and the domain maximum
18 decrease is 55.2 meters (more than two times compared to 1960). Correspondingly, the domain
19 averaged $PM_{2.5}$ increases by $0.7 \mu\text{g}/\text{m}^3$ and the domain maximum increase is $5.1 \mu\text{g}/\text{m}^3$. The
20 enhanced strength of aerosol feedbacks is another important cause of degraded aerosol pollution.
21 Thus, controlling emissions will have a co-benefit of reducing strength of aerosol feedbacks.

22

1 **3.7 Implications for the effects of emission and meteorology changes on PM_{2.5}**
2 **concentrations**

3 The simulated responses of PM_{2.5} concentrations to emission changes and meteorology changes
4 presented here, along with the previous presented effects of aerosol feedbacks (Gao et al. 2016),
5 provide important implications for the causes of the dramatic increases in winter PM_{2.5}
6 concentrations.

7 We calculated domain maximum changes in PM_{2.5} concentration averaged over four stagnant
8 days (January 16-19) owing to emission changes from 1960-2010 (EMI2010-CTL), temperature
9 increases (CTL-CTL_T2), RH decreases (CTL-CTL_RH10), wind speed decreases (CTL-
10 CTL_RH20), and aerosol feedbacks (CTL-CTL_NF). The values are 137.7, 2.0, 2.6, 7.5 and
11 4.0µg/m³, respectively. When the perturbations are based on emission levels in 2010, domain
12 maximum changes in PM_{2.5} concentration due to temperature increases (EMI2010-
13 EMI2010_T2), RH decreases (EMI2010-EMI2010_RH10), wind speed decreases (EMI2010-
14 EMI2010_WS20), and aerosol feedbacks (EMI2010-EMI2010_NF) are 4.8, 4.7, 26.4 and
15 25.5µg/m³. The effects of emission changes on haze formation are dominant and the effects of
16 aerosol feedbacks are comparable to the effects of wind speed decreases.

17 The comprehensive comparisons of these factors are also summarized in Table 3. Based on the
18 monthly domain mean responses of PM_{2.5} concentrations to these factors, dramatic emission
19 changes due to urbanization and industrialization are the main causes of degraded air quality and
20 frequent haze occurrences in in North China. PM_{2.5} shows significant responses to changes in
21 SO₂, NH₃, NO_x emissions than BC and OC (about 106.3% higher). In addition, PM_{2.5} shows
22 significant increases in response to changes in SO₂ and NH₃ emissions, as compared to increases
23 in response to changes in NO_x emissions. This region is relatively ammonia-poor in winter, so

1 reducing NH₃ emissions might be effective, which is consistent with previous findings in Europe
2 (Megaritis et al., 2013). SO₂ is the precursor of sulfate, which accounts for a large fraction of
3 PM in this region. Thus, they should be preferentially controlled in order to reduce PM_{2.5} levels.
4 To control SO₂ emissions, the usage of natural gas or other clean energy should be promoted to
5 reduce the usage of coal. NH₃ emissions in China are mainly from agriculture sources (about
6 90%), including livestock, fertilizer, and agricultural soil (Huang et al., 2012). Lelieveld et al.
7 (2015) found that agricultural emissions make the largest relative role in PM_{2.5} concentration in
8 eastern USA, Europe, Russia and East Asia. To control NH₃ emissions from agriculture sources,
9 some animal feeding and animal housing strategies should be taken. In addition, controlling
10 emissions will also have a co-benefit of reducing strength of aerosol feedbacks.

11 According to the ECLIPSE_GAINS_4a emission dataset, SO₂ emissions in China will decrease
12 by -26%, NO_x emissions in China will increase by 19%, and NH₃ emissions in China will
13 increase by 14% from 2010 to 2030. We predicted ([EMI_2030](#): by perturbing SO₂, NO_x and
14 NH₃ emissions by -26%, 19% and 14%) that these changes will lead to large decreases in winter
15 sulfate (-2.3μg/m³ on domain average). Nitrate will increase by 1.5μg/m³ and ammonium will
16 slightly decrease (-0.05μg/m³) on domain average. The net change of domain averaged PM_{2.5}
17 concentration is not significant (-0.8μg/m³), so more efforts are needed to control these important
18 gaseous precursors.

19 From the information listed in Table 3, the responses of PM_{2.5} concentrations to [approximated](#)
20 [changes in](#) temperature and RH [between 1960 and 2010](#)~~perturbations~~ are not as significant as to
21 [approximated change in](#) wind speed [between 1960 and 2010](#)~~perturbations~~. From Sect. 3.3, we
22 also found that the effects of [approximated changes in temperature between 1960 and 2010](#)
23 ~~temperature perturbation~~ on PM_{2.5} concentration are dominant by changes in PBLH and wind

1 fields. Previous studies have pointed out the occurrences of haze events are highly associated
2 with atmospheric circulation anomalies (Chen and Wang, 2015; Zhang et al., 2016). Thus,
3 changes in atmospheric circulations may be another important cause of growing haze pollution,
4 in addition to emission changes. Furthermore, aerosol can also change atmospheric circulation,
5 especially in severely polluted East Asia. Thus, controlling emission may have co-benefits of
6 mitigate aerosol effects on atmospheric circulation.
7 The effects of changing atmospheric circulations on winter haze pollution in China is beyond the
8 scope of this paper, but should be investigated in future studies.

9

10 **4 Summary**

11 A fully online coupled meteorological and chemical transport model, WRF-Chem was used to
12 study responses of winter PM_{2.5} concentrations to changes in emissions of SO₂, BC, OC, NH₃,
13 and NO_x and to meteorology (temperature, RH, and wind speeds) changes in North China region,
14 where people are suffering from severe winter haze pollution.

15 The detailed historical emissions dataset MACCity for year 1960 and 2010 were used to evaluate
16 the impacts of changes in emissions of SO₂, BC, and OC. From 1960 to 2010, the dramatic
17 changes in emissions lead to +264.0% increases in sulfate, +322.5% increases in nitrate,
18 +295.2% increases in ammonium, +157.0% increases in BC and 54% increases in OC. The
19 domain mean PM_{2.5} concentrations increase by 14.7µg/m³ and the domain maximum increase is
20 about 45µg/m³. The responses of PM_{2.5} to individual emission species indicate that the
21 simultaneous increases in SO₂, NH₃ and NO_x emissions dominated the increases in PM_{2.5}
22 concentrations. PM_{2.5} shows significant increases in response to SO₂ and NH₃ emission changes.

1 The increases in NO_x emissions may decrease surface ozone concentration and surface OH
2 radical concentrations, because North China region is VOC-limited in the winter. In addition, OC
3 accounts for a large fraction in $\text{PM}_{2.5}$ changes.

4 The sensitivities of $\text{PM}_{2.5}$ to emission changes of its precursors provide some implications for
5 haze pollution control. SO_2 , NH_3 and OC should be preferentially controlled. In China, the
6 residential sector, particularly biofuel usage is the primary sources of OC (Lu et al., 2011). The
7 usage of natural gas or other clean energy should be promoted to reduce the usage of coal and
8 biofuel to reduce SO_2 and OC. To control NH_3 emissions from agriculture sources, some animal
9 feeding and animal housing strategies should be taken.

10 The effects of changes in winter time meteorology conditions were also studied. Emission
11 changes from 1960 to 2010 substantially increase numbers of haze days, but meteorology
12 perturbations do not show any significant impacts. The ~~perturbations-approximated changes~~ in
13 temperature and RH between 1960 and 2010 do change $\text{PM}_{2.5}$ concentrations, but the strength is
14 not as significant as the effects of wind speed and emission changes. The effects of the
15 approximated changes in temperature between 1960 and 2010~~temperature-perturbation~~ are
16 dominated by the changes in surface wind fields and PBLHs. The effect of aerosol feedbacks is
17 comparable to the effect of decreasing wind speeds and the strength of aerosol feedbacks
18 significantly increased from 1960 to 2010.

19 The above discussions indicate that aerosol concentrations are mainly controlled by atmospheric
20 circulations, except emission changes. Thus, long-term trends in atmospheric circulations maybe
21 another important cause of winter haze events in North China. More studies are necessary to get
22 a better understanding of the aerosol-circulation interactions.

1 In our previous modeling study of the same period (January 2010), we found that SOA
2 contribution was small, so we did not include SOA in this study. But this indication might be
3 problematic due to current poorly parameterized SOA scheme. In the future, how changes in
4 emissions and meteorology variables affect productions of SOA during winter should be further
5 studied using more advanced SOA schemes. In addition, we did not consider primary PM except
6 BC and OC in the model because there is no information in the MACCity emission inventory,
7 which is another direction for improvements in future studies.

8

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14 gao@uiowa.edu) or G.R. Carmichael (gcarmich@engineering.uiowa.edu) for data requests.

15

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Table 1. Simulation cases and descriptions

Cases	Descriptions
CTL	Base case, anthropogenic emissions are from MACCity dataset for year 1960, <u>meteorological conditions are for January 2010</u>
EMI2010	Anthropogenic emissions are from MACCity dataset for year 2010
SO ₂ -2010	Same as CTL case except SO ₂ emissions are for year 2010
NH ₃ -2010	Same as CTL case except NH ₃ emissions are for year 2010
NO _x -2010	Same as CTL case except NO _x emissions are for year 2010
CTL_T2	Same as CTL case except temperature BCs and ICs are decreased by 2K
CTL_RH10	Same as CTL case except RH BCs and ICs are increased by 10%
CTL_WS20	Same as CTL case except wind speed BCs and ICs are increased by 20%
CTL_NF	Same as CTL case except aerosol-radiation interactions are excluded
EMI2010_T2	Same as EMI2010 case except temperature BCs and ICs are decreased by 2K
EMI2010_RH10	Same as EMI2010 case except RH BCs and ICs are increased by 10%
EMI2010_WS20	Same as EMI2010 case except wind speed BCs and ICs are increased by 20%
EMI2010_NF	Same as EMI2010 case except aerosol-radiation interactions are excluded
	<u>BCs 1960-2010, innermost domain emissions fixed in 1960 and the outer domain emissions changed from 1960 to 2010</u>

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EMI 2030; perturbate SO₂, NO_x and NH₃ emissions by -26%, 19% and 14%

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Table 2. Monthly domain mean concentrations of PM_{2.5} and its major components for year 1960, and domain maximum and mean concentrations for changes from 1960 to 2010 due to emission changes (µg/m³)

Years		SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	BC	OC	PM _{2.5}
1960	Domain	1.9	0.8	0.8	1.5	4.6	19.2
	mean						
1960-2010	Domain	18.9	7.8	6.8	9.9	11.1	45.0
	maximum						
	Domain	5.0	2.6	2.3	2.3	2.5	14.7
	mean	(264.0%)	(322.5%)	(295.2%)	(156.6%)	(54.0%)	(76.4%)

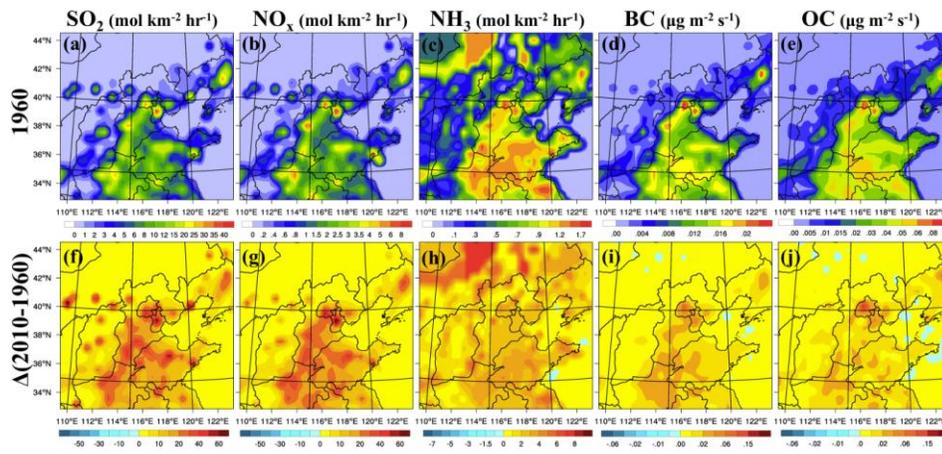
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Table 3. Monthly domain mean changes of sulfate, nitrate, ammonium and PM_{2.5} concentrations (µg/m³) due to emission and meteorology perturbations, and aerosol feedbacks (the two values of PM_{2.5} changes are for meteorology perturbations and aerosol feedbacks based on 1960 and 2010 emission levels, respectively)

	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	PM _{2.5}
Changes in SO ₂ emissions	3.4(178.3%)	-0.3 (-32.3%)	0.2 (29.4%)	3.4
Changes in NH ₃ emissions	0.1 (5.3%)	1.5 (189.6%)	0.6 (84.0%)	2.3
Changes in NO _x emissions	-0.7 (-39.1%)	0.6 (76.0%)	-0.04 (-5.1%)	-0.2
Changes in SO ₂ , NH ₃ , NO _x emissions	5.0 (264.0%)	2.6 (322.5%)	2.3 (295.2%)	9.9
Changes in BC emissions	-	-	-	2.3
Changes in OC emissions	-	-	-	2.5
Temperature perturbations	-	-	-	-0.01/0.3
RH perturbations	-	-	-	-0.7/-1.1
Wind speed perturbations	-	-	-	-2.3/-0.5

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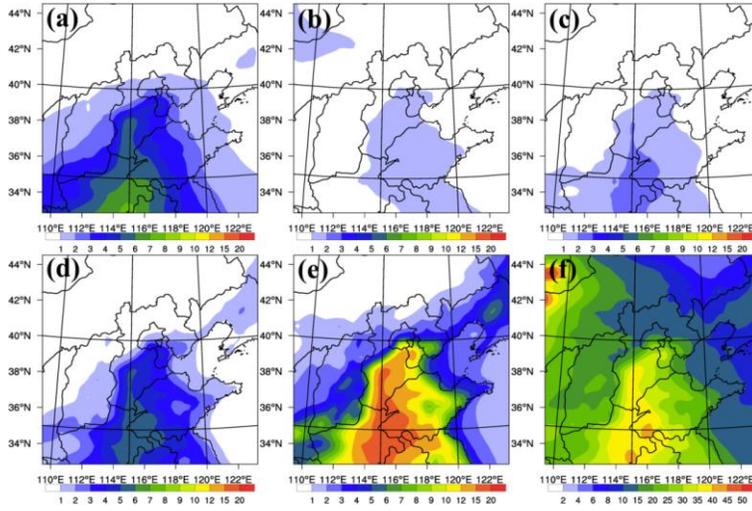
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4 Figure 1. SO₂, NO_x, NH₃, BC and OC emissions for year 1960 (a-e), and the changes of them from

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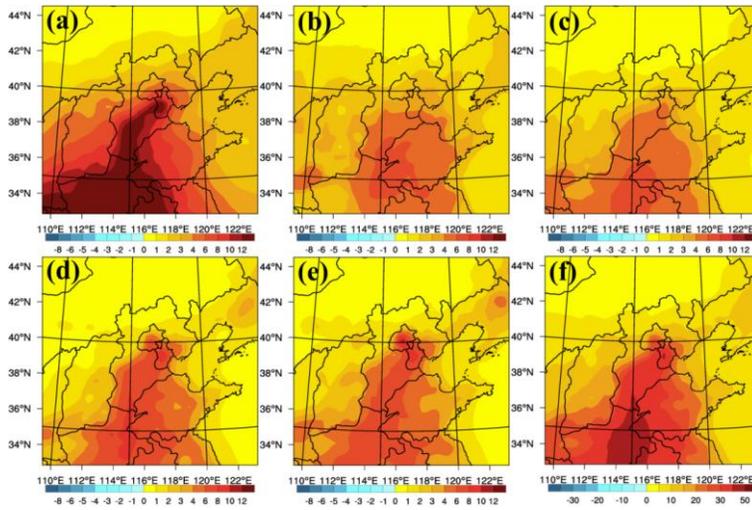
1960 to 2010 (f-j)

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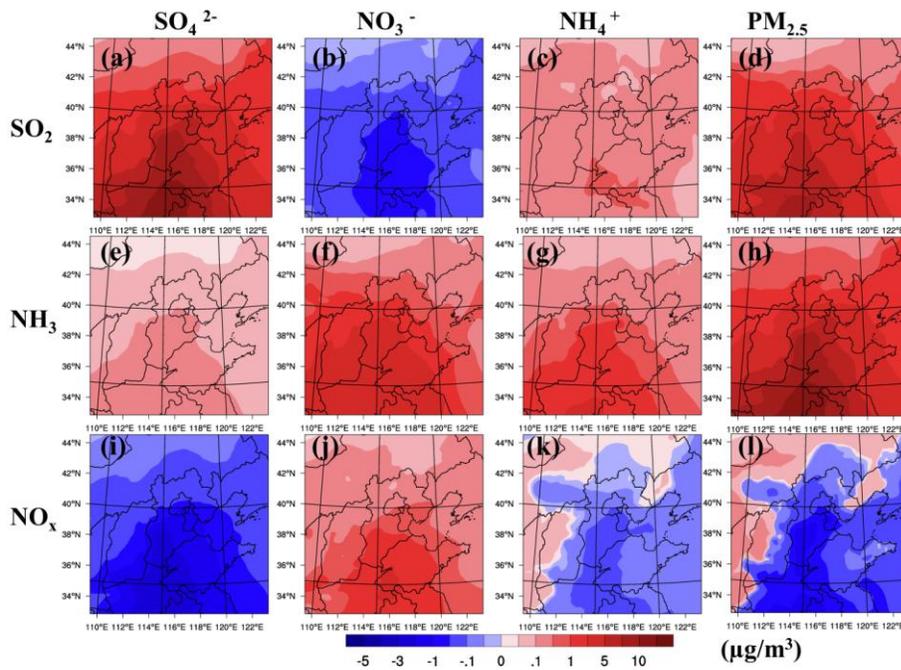
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Figure 2. Predicted monthly mean sulfate (a), nitrate (b), ammonium (c), BC (d), OC (e) and $PM_{2.5}$ (f) concentrations based on emissions for year 1960



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Figure 3. Predicted monthly mean changes of sulfate (a), nitrate (b), ammonium (c), BC (d), OC (e) and $PM_{2.5}$ (f) due to emission changes from 1960 to 2010



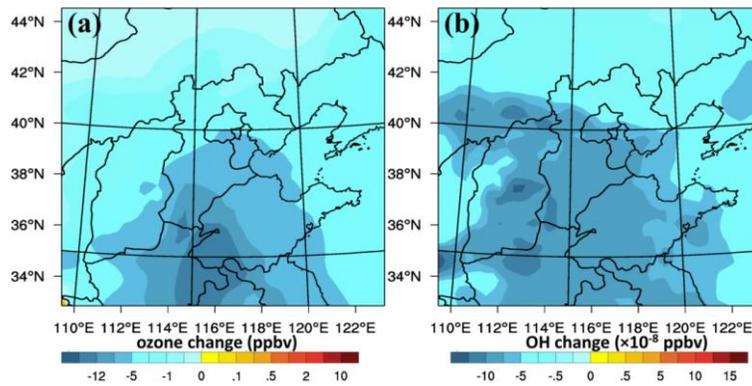
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Figure 4. Responses of PM_{2.5} and major PM_{2.5} inorganic species (sulfate, nitrate, and ammonium) to individual changes in SO₂, NH₃ and NO_x emissions from 1960 to 2010

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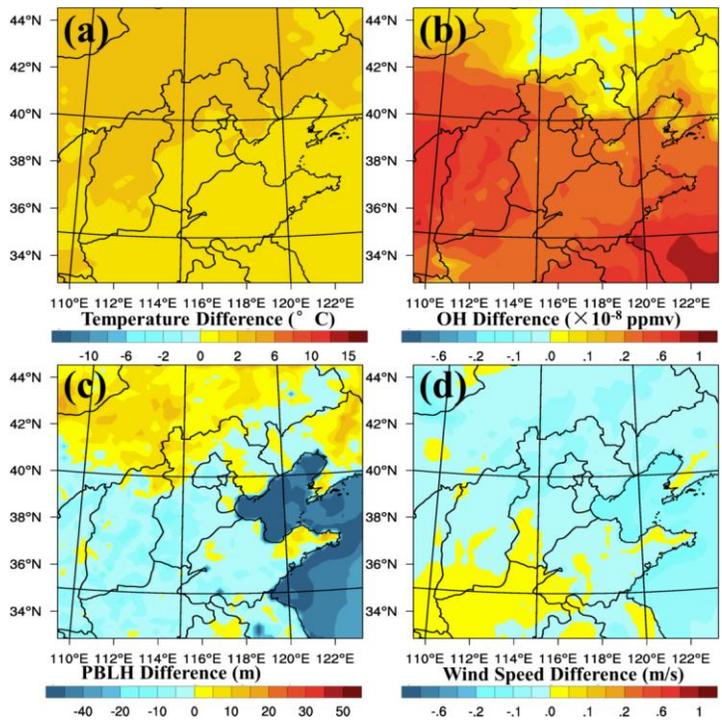


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Figure 5. Daytime ozone (a) and daytime OH (b) changes due to NO_x emission increases

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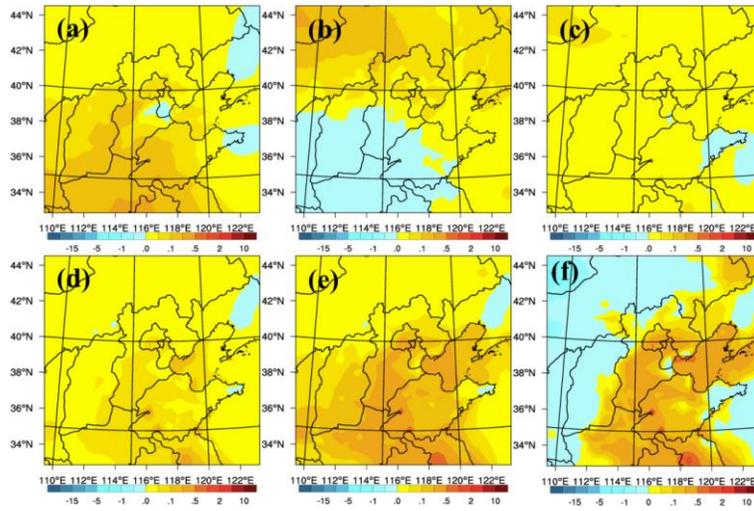


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3 Figure 6. Monthly mean temperature difference due to perturbation in initial and boundary conditions

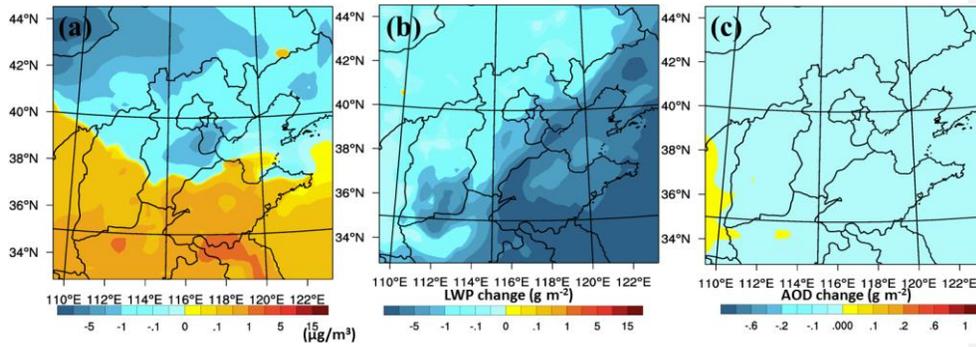
4 (a), and daily mean OH (b), mean PBLH (c) and mean near surface wind speed changes (d) due to

5 temperature increase



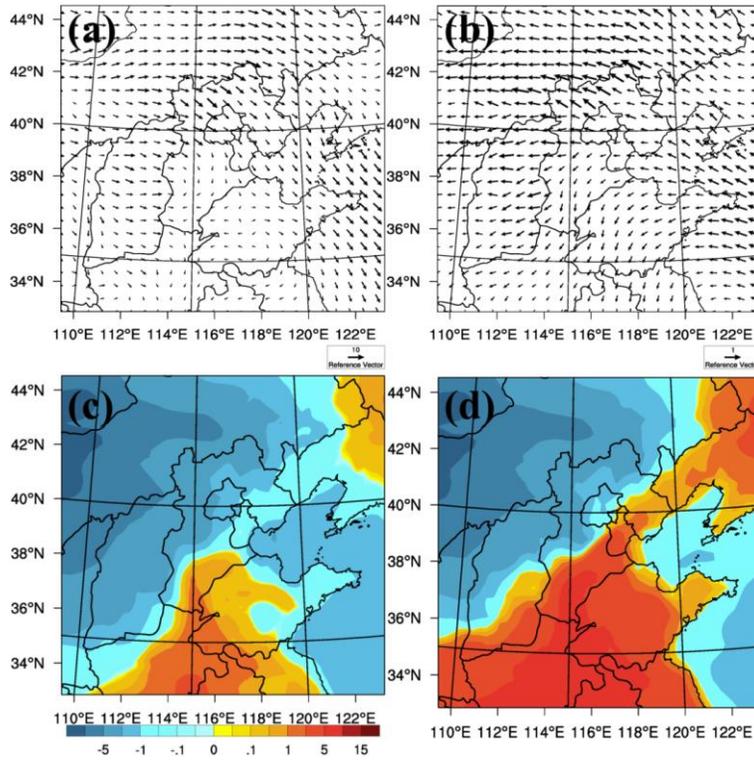
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Figure 7. Monthly mean changes of sulfate (a), nitrate (b), ammonium (c), BC (d), OC (e), and PM_{2.5} (f) and due to temperature increase



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Figure 8. Monthly mean changes of PM_{2.5} (a), LWP (b), and AOD at 600nm (c) due to RH decrease



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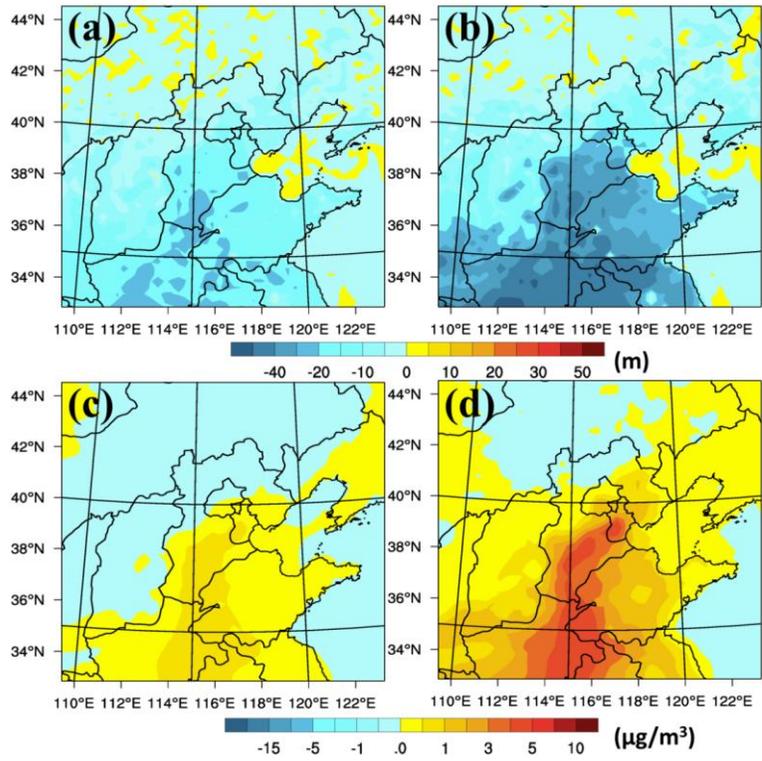
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Figure 9. Monthly mean wind fields for WS20 case (a) and changes of wind speeds (CTL-
 CTL_WS20) (b), and mean changes of $PM_{2.5}$ concentrations based on 1960 emission levels (c) and 2010
 emission levels (d)



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Figure 10. Monthly mean changes of daytime PBL heights for year 1960 (a) and 2010 (b), and of daytime PM_{2.5} concentrations for year 1960 (c) and 2010 (d) due to aerosol-radiation interactions