

We thank both reviewers for careful reading and thoughtful and constructive comments to improve the analysis and writing of the manuscript. The revisions/additions/edits are highlighted as red texts in the revised manuscript.

Reviewer #1

This is a sensitivity study on the efficiency of different regional SO₂ emission control scenarios in reducing sulfate pollution in China. The authors ran GEOS-Chem chemical transport model for 4 different emission reduction scenarios, all of which cut the overall SO₂ emissions from China by 8% from the 2010 level but distribute the reductions differently. In one scenario, the SO₂ emissions are cut uniformly over the entire country, while in the other three scenarios, the reductions are limited to three main source regions (North China, South China, and Southwest China). The authors then compared the resulting reductions in national average sulfate, population-weighted sulfate, and export of sulfur (SO₂ + sulfate) to the West Pacific for different scenarios, and concluded that controlling SO₂ emissions from North China will have the greatest benefit in terms of reducing national average sulfate and export of sulfur species, while controlling SO₂ emissions from South China will have greater benefit in reducing population-weighted sulfate concentration. Sensitivity tests were also conducted to investigate the effects of meteorology and the amount by which SO₂ emissions are reduced on the conclusion. Overall, this is a well-designed study with interesting results that may have some implications for pollution control strategies for China. The writing is understandable (although can still use some improvement) and the figures are mostly clear. I feel that the paper would be suitable for publication in Atmos. Chem. Phys. after the following comments have been addressed.

Specific Comments:

1. The authors compared the model simulated AOT, sulfate, and sulfate deposition with measurements, but only briefly mentioned the regional comparison results for AOT, which is not a direct measurement of sulfate. I wonder if the authors can comment on the regional biases in modeled sulfate and how the biases can affect the conclusions of this study.

Response: The reviewer's point is well taken. In the original paper we compared simulated sulfate with measurements at one surface site located in North China. The question of concern here is whether the model bias differs by region and if so, what is the implication for our conclusions. To address this question, we've added model-measurement comparisons at another two sites: Jinsha located in South China and Chengdu located in Southwest China. The model has an annual mean bias of 5% at the Jinsha site and -8% at the Chengdu site (new Figure 5b and c), although seasonal biases are higher partly due to the fact that the simulation and measurements are for different years. By comparison, the annual-mean bias at the Miyun site is 4%. Since the annual mean model biases are consistently within $\pm 10\%$ for the three regions of interest, we argue that the regional biases in modeled sulfate will not affect the conclusion of our paper. Please refer to the new Figure 5 and the revised Section 2 in the revised manuscript.

2. Again, AOT over China can be affected by a number of factors such as dust and humidity. Have the authors looked into other satellite datasets such as SO₂ for model evaluation?

Response: We have conducted additional model evaluation using total SO₂ columns from the OMI satellite instrument. Compared to AOT, satellite-derived SO₂ columns provide a more direct evaluation of sulfur simulation in the model since SO₂ is the direct precursor of sulfate. The original horizontal resolution of the Level 3 OMI data is 0.25°×0.25°, and the GEOS-Chem simulation has a resolution of 0.5°×0.667°. We regridded both OMI and modeled SO₂ columns to 1°×1° resolution for comparison. The spatial distribution of SO₂ column densities from GEOS-Chem correlates well with those from OMI, with the correlation coefficient being 0.79, 0.73 and 0.64, for NC, SC and SWC, respectively. Compared with the OMI SO₂ retrievals, GEOS-Chem simulated SO₂ columns are 6% higher in NC, 2% higher in SC and 8% lower in SWC. The discrepancy between the modeled and OMI SO₂ is within ±10% for all the three regions, indicating an overall good simulation of SO₂ by the GEOS-Chem model. Please refer to the revised manuscript Section 2 and new Figure 3.

3. I understand that this is merely a sensitivity study, but can the authors comment on the actual SO₂ emission change during and/or before the study period (given that the emission inventory seems to be available for multiple years)? How do the actual national/regional trends compare with the different scenarios tested in the study?

Response: We have included relevant discussion in Section 5 of the revised manuscript. In this study, we recommend that SO₂ emission control over NC should be stressed to maximize the air quality benefits for China and downwind regions. However, from 2006 to 2010 (the 11th Five-Year-Plan period), SO₂ emissions from NC have decreased at a much slower rate than the national total emissions. Based on the MEIC inventory, total SO₂ emissions from China were 9.4% lower in 2010 than 2006, and emissions from NC, SC and SWC have decreased by 4.7%, 16.1% and 23.1%, respectively, during the same period. The relative reduction of SO₂ emissions in NC is thus one third or less of that for the other two regions and is only half of the reduction at the national mean level. This indicates that China has not prioritized SO₂ emission control in NC in the past. Our study suggests this should be corrected in the future in order to maximize the benefits of SO₂ control. Please refer to the revised manuscript in Section 5.

4. I assume that the population weighted sulfate concentration can be calculated on a grid-box basis instead of for each province – by weighing the sulfate concentration with a ratio between population density in each grid box and the national average population density? Some provinces seem to be in more than one study region and that may lead to uncertainty in the population-weighted sulfate.

Response: Actually, even if one or two provinces are in more than one study region, this will not lead to uncertainty in our calculation of the population weighted

sulfate concentration (PWC) for each region. This is because our calculation is based on individual model grid boxes rather than provincial boundaries. We first multiply sulfate concentration by population in each grid box within a region, then sum them up, and finally divide the sum by the total population within the region. As the reviewer suggested, if we calculate the PWC by weighing the sulfate concentration with a ratio between population density in each grid box and the national average population density, the PWC will not be at the same order of magnitude with surface sulfate concentration because the population density at some grid boxes is tens of times greater than the national mean population density. We clarified this in the revised manuscript.

5. Can the authors comment on the seemingly larger bias in the modeled sulfate for the second half of the period covered by Figure 3a?

Response: The larger bias of the model during the late spring and summer can be explained by the model's weakness in simulating large precipitation and high wind speeds at the local scale (Zhang L. et al., 2012, Wang, Y. et al. 2013a). We have added a discussion on this issue (Section 2 in the revised manuscript).

6. It will be useful to mark the scenarios for Figures 4b and 4c.

Response: We have marked the scenario names in the figures. Please refer to the new Figure 6b and 6c.

7. Can the authors point out where the "other regions" in Figure 6b are? Northeast China?

Response: Not only Northeast China, but all the rest Chinese regions as well as global influence. We clarified this in the new Figure 8b.

8. Can the authors provide an explanation for the more dominant role of gas-phase photochemistry for North China than South and Southwest China? Less humidity? Stronger NO_x emissions? Also how does the presumably stronger washout (and shorter lifetime) in the southern part of China affect the conclusions of the study?

Response: The lower atmospheric humidity over NC will inhibit the aqueous phase oxidation, while the stronger NO_x emissions from NC will result in higher OH concentrations and thus enhance the gas phase oxidation. Both factors are responsible for higher percentage of gas phase SO₂ oxidation over North China. The shorter lifetime of sulfate over SC and SWC makes it harder to transport over long distances to downwind regions, so it is another reason why SO₂ emission control from NC has the largest efficiency factor in reducing national mean surface sulfate concentration. We have added these two points in Section 4, please refer to the revised manuscript.

Reviewer #2

1. SO₂ control will accompany with the change of NO_x and hydrocarbons emissions which can induce the O₃, OH and H₂O₂ variation. This would lead to gas phase and aqueous oxidation process of SO₂. The uncertainty analysis should be done to consider these processes.

Response: We agree with the reviewer that SO₂ emission control may accompany with changing emissions of NO_x and VOCs, because these pollutants have common sources as SO₂. However, while coal combustion is the dominant source of SO₂, it is not the most important source for NO_x or VOCs (transportation being another equally important source for those species). In addition, the technologies used to control SO₂ emissions from coal power plants cannot remove NO_x or VOCs to the same extent as SO₂, and they may not affect NO_x and VOCs equally. To address the impact of changing emissions of co-emitted species on SO₂ chemistry, we conducted a new set of sensitivity tests considering the extreme circumstance in which NO_x and VOCs emissions are also decreased by 8%, equal to the magnitude of SO₂ emission change in the S1 scenario. The results from this sensitivity test are within $\pm 2\%$ of those from S1 for all the metrics we considered. For example, the decrease of national mean SO₂ and sulfate concentrations is 7.80% and 5.76%, respectively; the corresponding value is 7.90% and 5.70% from the S1 simulation. This indicates that the change in NO_x and VOCs emissions as a result of SO₂ emission control processes has a negligible impact on SO₂ oxidation and as such it will not affect the conclusion of this study. Please refer to the revised manuscript in Section 4.3.

2. In Page 14, the authors analyzed SO₂ conversion and emphasized that the aqueous oxidation is very important process. However in page 15, the results showed that all three regions, the relative decrease of gas phase oxidation is greater than that of aqueous oxidation, more explanations should be given to this point.

Response: Aqueous phase oxidation (mainly by H₂O₂) of SO₂ contributes 45%, 64% and 73% to sulfate over NC, SC and SWC, respectively. The lower humidity over NC inhibits the aqueous phase oxidation, and the stronger NO_x emissions from NC raise the OH concentration and then enhance the gas phase oxidation, so both of them are the reasons for the higher percentage of gas phase SO₂ oxidation over North China. SO₂ emission changes affect both gas- and aqueous-phase oxidation processes, but the magnitude of the influence depends on whether the process is SO₂ limited or not. In most-polluted areas with high NO_x emissions (like China), the aqueous oxidation tends to be oxidants limited rather than SO₂-limited, because of the impact of high-NO_x concentrations on OH, H₂O₂, and O₃. Previous sensitivity studies (Berglen et al., 2004) have found that the gas-phase oxidation is more limited by SO₂. Therefore, SO₂ emission change will have a stronger impact on gas phase oxidation than aqueous phase oxidation and this explains why when SO₂ emissions decrease, the relative decrease of gas phase oxidation is larger than that of aqueous phase oxidation. We have clarified this point in the text; please refer to the revised manuscript in section 4.1.

3. More detailed information should be added to describe how to calculate the sulfur outflow flux. Some references should be cited to show the Winter and Spring are the significant seasons for pollutants export.

Response: Previous studies have found that pollution outflow from East Asia to the Pacific peaks in spring (Chin et al., 2007; Clarisse et al., 2011), while winter is also a significant contributor (Feng et al., 2007), so we calculate the sulfur flux to Western Pacific in each scenario for the winter and spring seasons. We have added those references in the manuscript. The transport flux is evaluated at the boundary of 123 °E and 22 °N - 42 °N within the troposphere, and the sulfur flux includes both SO₂ and sulfate. We define eastward flux as positive, the sulfur fluxes for both seasons are positive, indicating the net export of sulfur compounds from China to the Western Pacific. Please refer to the revised section 3.4 in the manuscript.

4. Why SO₂ concentration is not one of the impact metrics?

Response: The reviewer's point is well taken. We have added SO₂ as one of the impact metrics. Comparing SO₂ metrics from the four emission scenarios, the national mean SO₂ concentration is also most sensitive to SO₂ emission changes from NC (S2 scenario), resulting in the emission control efficiency factor of 1.0. SO₂ emissions from NC are 36% and 129% higher than those from SC and SWC, respectively. Because of the short lifetime of SO₂, reducing SO₂ emissions from one region has a small effect on SO₂ concentrations over the other regions, and thus the national-mean SO₂ concentration is most sensitive to SO₂ emissions from NC. This result is also robust without a strong dependence on meteorology or magnitude of SO₂ emission reduction. Please refer to Section 3.2, 4.3 and the new Figure 6a in the revised manuscript.

1 **Regional differences in Chinese SO₂ emission control efficiency**
2 **and policy implications**

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17 **Abstract**

18 SO₂ emission control has been one of the most important air pollution policies in
19 China since 2000. In this study, we assess regional differences in SO₂ emission
20 control efficiencies in China through the modeling analysis of four scenarios of SO₂
21 emissions, all of which aim at reducing the national total SO₂ emissions by 8% or 2.3
22 Tg below the 2010 emissions level, the target set by the current 12th **Five-Year Plan**
23 (FYP) (2011-2015), but differ in the spatial implementation. The GEOS-Chem
24 chemical transport model is used to evaluate the efficiency of each scenario on the
25 basis of **four** impact metrics: surface **SO₂** **and** sulfate concentrations,
26 population-weighted sulfate concentration (PWC), and sulfur export flux from China
27 to the Western Pacific. The efficiency of SO₂ control (β) is defined as the relative
28 change of each impact metric to a 1% reduction of SO₂ emissions from the 2010

29 baseline. The S1 scenario, which adopts a spatially uniform reduction of SO₂
30 emissions in China, gives a β of 0.99, 0.71, 0.83, and 0.67 for SO₂ and sulfate
31 concentrations, PWC, and export flux, respectively. By comparison, the S2 scenario,
32 which implements all the SO₂ emissions reduction over North China (NC), is found
33 most effective in reducing national-mean surface SO₂ and sulfate concentrations and
34 sulfur export fluxes, with β being 1.0, 0.76 and 0.95 respectively. The S3 scenario of
35 implementing all the SO₂ emission reduction over South China (SC) has the highest β
36 in reducing PWC ($\beta = 0.98$) because SC has the highest correlation between
37 population density and sulfate concentration. Reducing SO₂ emissions over Southwest
38 China (SWC) is found to be least efficient on the national scale, albeit big
39 within-region benefits. The difference in β by scenario is attributable to the regional
40 difference in SO₂ oxidation pathways and source-receptor relationship. Among the
41 three regions examined here, NC shows the largest proportion of sulfate formation
42 through gas phase oxidation, which is more sensitive to SO₂ emission change than
43 aqueous oxidation. In addition, NC makes the largest contribution to inter-regional
44 transport of sulfur within China and to the transport fluxes to Western Pacific. The
45 policy implication is that China needs to carefully design a regionally specific
46 implementation plan of realizing its SO₂ emissions reduction target in order to
47 maximize the resulting air quality benefits not only for China but for the downwind
48 regions, with emphasis on reducing emissions from NC where SO₂ emissions have
49 decreased at a slower rate than national total emissions in the previous FYP period.

50 **1. Introduction**

51 SO₂ is the precursor of ambient sulfate, which is a major component of particulate
52 matter with dynamic diameter less than 2.5 μm (PM_{2.5}) and makes up about 20-35%
53 of total PM_{2.5} mass (Pathak et al., 2009). SO₂ emissions from China contribute about
54 25% of global SO₂ emissions and 50% of Asian emissions (Streets et al., 2003; Lu et
55 al., 2011). Since 2000, the Chinese government has made great efforts in controlling
56 SO₂ emissions in order to reduce atmospheric PM concentrations and acid deposition.
57 A 10% SO₂ emission reduction target was set in both the tenth **Five-Year** Plan (10th
58 FYP, 2000-2005) and the 11th FYP (2006-2010). While SO₂ emissions increased about
59 28% during the 10th FYP (Schreifels et al., 2012), by the end of the 11th FYP China
60 has achieved the goal of 10% SO₂ emission reduction in 2010 relative to the 2005
61 level (Lu et al., 2011). Itahashi et al. (2012a) reported that aerosol optical depths
62 (AOD) retrieved from the Moderate Resolution Imaging Spectroradiometer (MODIS)
63 over East Asia showed an increase from 2001 to 2005 and then a decrease until 2010,
64 consistent with the reported trend of SO₂ emissions from China.

65 In 2012, PM_{2.5} was introduced into China's ambient air quality standard by the
66 Ministry of Environmental Protection (MEP). In response to **the increasingly** severe
67 haze pollution, the Action Plan on Prevention and Control of Air Pollution was
68 delivered by the Central Government of China in September 2013. Aiming to improve
69 air quality across China in the next five to ten years, the Action Plan calls for more
70 efforts to reduce emissions from the heavily polluted regions in East China (22 °-42 °N,
71 110 °-122 °E). The Action Plan requires that by 2017, PM_{2.5} concentrations should

72 decrease by 25% over the Beijing-Tianjin-Hebei region, 20% over the Yangtze River
73 Delta, and 15% over the Pearl River Delta. While the Action Plan presents guidelines
74 and advice for energy consumption and cleaner production, like other laws and
75 regulations in China there is no specific emission control target for primary PM or its
76 gaseous precursors for the whole country or by specific regions.

77 The purpose of this study is to assess the regional differences in SO₂ emission
78 control efficiency in China and discuss the implications for future emission control
79 strategies. We choose four impact metrics to evaluate the efficiency of different
80 scenarios of SO₂ emissions reduction in China. The first two metrics are surface
81 concentrations of SO₂ and sulfate. The GEOS-Chem chemical transport model is used
82 to quantify the response of SO₂ and sulfate concentrations to changes in SO₂
83 emissions. Since public health is an important issue of concern for PM_{2.5}, the third
84 metric is population-weighted sulfate concentration at the surface. SO₂ emissions
85 from China also have significant impacts on air quality and public health of foreign
86 regions due to long-range transport. Park et al. (2004) suggested that trans-Pacific
87 transport of sulfate accounts for 30% of sulfate background in the U.S. Itahashi et al.
88 (2012b) reported that central eastern China is the dominant source region for sulfate
89 aerosols over Oki Island during two air pollution events in July 2005. Previous studies
90 have recognized the Western Pacific as the predominant transport pathway of air
91 pollution exported from China (Heald et al., 2006; Fairlie et al., 2010; Li et al., 2010a;
92 He et al., 2012). Given the global impact of changing Chinese emissions, it is
93 important to understand the response of pollution outflow to different emission

94 control strategies in China. Therefore, the **fourth** metric is the outflow flux of sulfur
95 from China to the Western Pacific.

96 Within the context of currently available emission control plans in China, we
97 design four different spatial realizations of **reducing China's total SO₂ emissions** by 8%
98 or 2.3 Tg below the 2010 emissions level, which is the target set by the current 12th
99 FYP (2011-2015). In the first scenario SO₂ emissions are reduced uniformly by 8%
100 over China, while in the other three scenarios the emissions reduction is implemented
101 over three different regions which make the largest contributions to the national SO₂
102 emissions and have highest sulfate concentrations: **North China (NC, 33 °-42 °N,**
103 **110 °-122 °E), South China (SC, 22 °-33 °N, 110 °-122 °E), and Southwest China (SWC,**
104 **23 °-33 °N, 102 °-110 °E) (Figure 1)**. Since sulfate aerosols exhibit regionally specific
105 formation and transport characteristics (Wang Y et al., 2013a), the response of a given
106 impact metric to the same amount of SO₂ emission reduction is expected to differ by
107 region.

108 It is clear that the target of 8% reduction of Chinese SO₂ emissions is far from
109 sufficient to meet the goal of reducing PM_{2.5} concentrations set by the Action Plan
110 (Wang Y et al., 2013a). However, there is no other specific target of SO₂ emissions
111 available in current Chinese policies to serve as an alternative reference point. Since
112 our study focuses on the regional difference in emission control efficiency, we choose
113 an 8% perturbation of total SO₂ emissions that is large enough to capture the regional
114 difference.

115 The paper is organized as follows. Section 2 describes the model and the

116 evaluation of model results by observations. In Section 3 we present the different
117 reduction scenarios of SO₂ emissions in China and analyze the regionally different
118 responses of the selected impact metrics to those scenarios. Section 4 analyzes sulfate
119 formation and sulfur transport by region to understand the mechanisms behind the
120 regional difference of SO₂ emission control efficiency, followed by sensitivity tests of
121 our results. The concluding remarks are presented in Section 5.

122 **2. Model description and evaluation**

123 **2.1 Model description**

124 We use the GEOS-Chem chemical transport model version 9-01-01 to simulate
125 the coupled aerosol-oxidant chemistry on the global and regional scale. The model is
126 driven by the assimilated meteorological data from the Goddard Earth Observation
127 System (GEOS) with 6-hour averaged winds, temperature, cloud and convective mass
128 flux, and 3-hour averaged surface quantities and mixing depths. Here we use the
129 nested-grid capability of GEOS-Chem with a 0.5°×0.667° horizontal resolution over
130 East Asia, which was originally described by Wang Y (2004) and Chen et al. (2009).
131 The global simulation with a horizontal resolution of 4°×5° is used to provide
132 boundary conditions for the nested-grid domain.

133 The sulfate-nitrate-ammonium (SNA) aerosol simulation coupled to the HO_x-
134 NO_x-VOC-ozone gas chemistry was originally developed by Park et al. (2004).
135 Emitted SO₂ in the model is oxidized to sulfate by hydroxyl radicals (OH) in the gas
136 phase and by hydrogen peroxide (H₂O₂) and ozone (O₃) in the aqueous phase. The

137 gas-particles equilibrium of aerosols is calculated using the ISOROPIA II (Fontoukis
138 and Nenes, 2007) aerosol equilibrium model. The aerosols are removed through dry
139 and wet deposition.

140 The Global Emission Inventory Activity (GEIA) inventory (Benkovitz et al.,
141 1996) is used in the global simulation, which is overwritten by the NEI05 inventory
142 over the US, the EMEP inventory over Europe, and the INTEX-B inventory over East
143 Asia (Wang Y et al., 2013a). For the nested-grid simulation, the Multi-resolution
144 Emission Inventory for China (MEIC) for the year 2010 is adopted over China (He,
145 2012) and emissions over the rest of East Asia are taken from the INTEX-B emission
146 inventory. The MEIC inventory uses an improved technology-based methodology to
147 estimate anthropogenic emissions from China, including emissions of SO₂, NO_x, NH₃,
148 BC, OC, NMVOCs, CO, CO₂, and fine and coarse mode PM. The MEIC inventory
149 has an open access dataset for the period from 1990 to 2010
150 (<http://www.meicmodel.org>), providing monthly emission data in the horizontal
151 resolution of 0.25°×0.25°, 0.5°×0.5° and 1°×1°. The MEIC emission inventory **has**
152 **been shown to provide** good estimation of total emissions with some uncertainties in
153 the spatial allocations for the fine grid resolutions within cities (Wang L. et al., 2014).
154 According to the MEIC inventory, SO₂ emissions from China are 28.4 Tg in 2010
155 (Figure 1).

156 **2.2 Model evaluation**

157 The GEOS-Chem simulation of sulfate and PM_{2.5} over China has been evaluated
158 by Wang Y et al. (2013a; 2014) and Lou et al. (2014). Wang Y et al. (2013a) indicated

159 that the GEOS-Chem model had a good performance in simulating sulfate
160 distributions ($R^2 = 0.64 \sim 0.79$) and concentrations (mean bias of -10%) in East Asia.
161 Lou et al. (2014) reported a higher correlation between simulated and observed sulfate
162 ($R^2 = 0.86$), but a larger model bias (-41%) which may be partly due to the fact that
163 they used a uniform factor of 0.6 to infer sulfate concentrations in $PM_{2.5}$ from those in
164 observed PM_{10} concentrations. Wang Y et al. (2014) further evaluated the model
165 performance in reproducing the concentrations and the spatiotemporal patterns of
166 $PM_{2.5}$ over China during a severely polluted month of January 2013. The model
167 shows a good correlation of 0.6 with $PM_{2.5}$ spatial distributions but underestimates the
168 concentrations of $PM_{2.5}$ and sulfate over North China during a severe haze period,
169 which is largely explained by underestimated emissions from this heavily polluted
170 region. The sulfate underestimation is further attributed to the heterogeneous reaction
171 of SO_2 on pre-existing aerosols that are deliquescent under the condition of higher
172 relative humidity during the severe haze period (Wang Y et al., 2014). In this study we
173 extend the previous model evaluation by using **four** additional datasets over China: (1)
174 AOD retrieved from MODIS for January, July, and annual mean of 2010; (2) SO_2
175 total columns retrieved by the Ozone Monitoring Instrument (OMI) satellite
176 instrument; (3) sulfate concentrations observed at three surface sites in China: the
177 Miyun site ($40^{\circ}29'N$, $116^{\circ}47'N$) in NC, the Jinsha site ($29^{\circ}38'N$, $114^{\circ}12'N$) in SC
178 (Zhang et al., 2014), and the Chengdu site ($30^{\circ}39'N$, $104^{\circ}2'N$) in SWC (Tao et al.,
179 2014); (4) monthly wet deposition fluxes at 5 sites from January 2009 to December
180 2010, which are from the Acid Deposition Monitoring Network in East Asia (EANET,

181 <http://www.eanet.asia/>). Locations of all the surface sites used in this study are
182 displayed in Figure 2a.

183 The spatial distributions of AOD over China for annual mean, January and July
184 of 2010 from MODIS (top) and the model (bottom) are displayed in Figure 3. The
185 model shows a strong spatial correlation ($R > 0.6$) with MODIS AOD. Both the model
186 and MODIS indicate higher annual mean AOD in North and South China and a shift
187 of the highest AOD values from central China in January to NC in July. The model
188 has a positive bias of more than 10% for the annual, January, and July mean AOD
189 levels over China as a whole. There is an obvious positive bias of the model over
190 North China, with an overestimation of 26% and 20%, respectively, for the annual and
191 January mean. The model shows a higher correlation (0.73) and smaller bias (12%)
192 over North China in July. The model bias over South China and Southwest China is
193 comparatively smaller and negative at -3% for SC and -1% for SWC. The positive
194 bias of AOD in North China is due to the overestimates of nitrate concentrations
195 (Wang Y et al., 2013a) and overestimation of dust emissions from the
196 Taklimakan-Gobi area which are transported to this region (Wang et al., 2012).

197 Figure 4 compares annual-mean total SO_2 column densities from OMI and
198 GEOS-Chem for 2010. Compared to AOD, satellite-derived SO_2 columns provide a
199 more direct evaluation of sulfur simulation in the model since SO_2 is the direct
200 precursor of sulfate. The original horizontal resolution of the Level 3 OMI data is
201 $0.25^\circ \times 0.25^\circ$, and the GEOS-Chem simulation has a resolution of $0.5^\circ \times 0.667^\circ$. We
202 regridded both OMI and modeled SO_2 columns to $1^\circ \times 1^\circ$ resolution for comparison.

203 The spatial distribution of SO₂ column densities from GEOS-Chem correlates well
204 with those from OMI, with the correlation coefficient being 0.79, 0.73 and 0.64 for
205 NC, SC and SWC, respectively. Compared with the OMI SO₂ retrievals, GEOS-Chem
206 simulated SO₂ columns are 6% higher in NC, 2% higher in SC and 8% lower in SWC.
207 The discrepancy between the modeled and OMI SO₂ is within $\pm 10\%$ for all the three
208 regions, indicating an overall good simulation of SO₂ by the GEOS-Chem model.
209 Wang Y et al. (2014) reported a 50% underestimate of SO₂ emissions from North
210 China by the GEOS-Chem model for an extremely polluted month of January 2013.
211 Since we used a different bottom-up inventory with higher SO₂ emission from NC and
212 simulated a different year, we did not find evidence that SO₂ emissions were
213 underestimated in winter 2010.

214 The simulated and observed surface sulfate concentrations at the three surface
215 sites are compared in Figure 5. Weekly-mean sulfate concentrations from January to
216 June 2010 are shown for the Miyun site (Figure 5a). The model agrees well with the
217 observed variability with a correlation of 0.75, and it shows a small positive bias of
218 4% for sulfate concentration. Wang Y et al. (2013a) reported a correlation of 0.7 and
219 overestimation of 15% of the model in comparison with the Miyun observations in
220 2007, using an old version of model and a different anthropogenic emission inventory
221 for China. Similar to Wang Y et al. (2013a), we find here that the model cannot
222 capture the sawtooth-like variation of sulfate during late spring and summer at the
223 Miyun site, which is caused by the model's weakness in simulating large precipitation
224 and high wind speeds at the local scale (Zhang L. et al., 2012). Observations at the

225 Jinsha and Chengdu site are collected from published literature. Jinsha is a regional
226 background site located in SC (Zhang et al., 2014) and the sampling period was from
227 March 2012 to March 2013. Chengdu is an urban site (Tao et al., 2014) located in
228 SWC and sulfate concentrations were collected during January, April, July and
229 October in 2011. The model has an annual mean bias of 5% at the Jinsha site (Figure
230 5b) and -8% at the Chengdu site (Figure 5c), with higher seasonal biases partly due to
231 the fact that the simulation and measurements are for different years.

232 Figure 2b displays the scatter plot of simulated versus observed monthly-mean
233 sulfate wet deposition fluxes from January 2009 to December 2010 at 5 EANET sites
234 in China (Figure 2a). The model reproduces the annual-mean sulfate wet deposition
235 fluxes with a high correlation of 0.83 and small negative bias of -9%. Seasonally, the
236 model tends to overestimate sulfate wet deposition fluxes in the winter (bias = 30%; R
237 = 0.63), but underestimate them in other seasons (R = 0.8 ~ 0.88, biases = -10% ~
238 -19%).

239 In summary, through comparison of the model results with satellite-derived
240 AOD and SO₂ columns, surface sulfate observations at three surface sites located in
241 each region, and sulfate wet deposition fluxes for 5 sites in China, we conclude that
242 the model has some capability to reproduce the spatial and temporal distributions of
243 sulfate over China with a small to moderate bias. While the model performance of
244 sulfate and SO₂ simulation differs by site and season, the annual-mean model biases
245 are all within ±10% for the three regions of interest and thus not expected to affect the
246 comparison of the emission scenarios.

247 3. Efficiency of SO₂ emission control strategies

248 3.1 Simulation scenarios

249 In this study, one standard simulation using the 2010 MEIC inventory of Chinese
250 emissions and four sensitivity simulations with different SO₂ emission reduction
251 scenarios (S1 – S4) are conducted, which are described in Table 1. The standard
252 simulation is carried out for the period from January 2009 to December 2010, with the
253 first year for spin up and the next year for analysis. SO₂ emissions from China in 2010
254 are 28.4 Tg in the standard simulation. All the emission reduction scenarios have an 8%
255 (2.3 Tg) reduction in national total SO₂ emissions below their 2010 level, following
256 the goal of the 12th FYP, but they differ in the spatial distributions of those reductions.
257 In the S1 scenario, the 8% emission reduction is implemented uniformly over the
258 whole country; in the S2, S3, and S4 scenario, the 2.3 Tg reduction of SO₂ emissions
259 is implemented by decreasing regional emissions from three sub-regions of China
260 which make the largest contributions to the national total emissions and have highest
261 sulfate concentrations (Zhang X. Y. et al., 2012; Wang Y et al., 2013a). The three
262 regions are North China (NC, S2 scenario), South China (SC, S3) and Southwest
263 China (SWC, S4). **The region definitions are shown in Figure 1.** Since the baseline
264 emissions from the three regions are different, the percentage of the emission
265 reduction differs by region, being 21.3%, 29.5% and 48.9% of the baseline emissions
266 from NC, SC and SWC, respectively. ~~For the emission reduction scenarios, only a~~
267 3-month initialization is conducted **for each of the emission reduction scenarios.**

268 Following Lamsal et al. (2011) and Zhang et al. (2014), we define a relationship

269 between the change of an impact metric (X) and the change of SO₂ emissions (E):
270 $\frac{\Delta X}{X} = \beta \times \frac{\Delta E}{E}$, where ΔX is the change in the metric, with X being surface SO₂ or
271 sulfate concentrations, population-weighted sulfate concentration, or sulfur outflow
272 fluxes from China to Western Pacific; $\frac{\Delta X}{X}$ is the relative change of the metric; ΔE is
273 the change of SO₂ emissions; $\frac{\Delta E}{E}$ is the relative emission change, which is 0.08 for all
274 the emission reduction scenarios on the national scale; and β is a unitless term
275 describing the relative changes of the metrics of concern in response to a 1% change
276 in SO₂ emissions. We call β the efficiency factor, and it is used to compare the
277 sensitivity of each metric to SO₂ emission changes between different emission
278 reduction scenarios. The larger β , the larger impact SO₂ emission change will have on
279 the related metrics. β tends to be ≤ 1 because the relative rate of change in sulfate will
280 not exceed that of SO₂ emissions. Considering that the emission reduction is
281 regional-specific in S2-S4 scenarios, a regional-specific efficiency factor $\beta_{r,A-B}$ is also
282 defined: $(\frac{\Delta X}{X})_B = \beta_{r,A-B} \times (\frac{\Delta E}{E})_A$, where A denotes the region where emissions are
283 reduced, and B denotes the region where the impact is evaluated. For example, $\beta_{r,NC-SC}$
284 of sulfate denotes the sensitivity of sulfate concentration change over SC to SO₂
285 emission reduction over NC. Here the regional $\frac{\Delta E}{E}$ is 0.08 for all the regions in S1,
286 0.213 for NC in S2, 0.295 for SC in S3, and 0.489 for SWC in S4 scenario. Since the
287 S1 scenario does not have a regional-specific reduction in emissions, the regional
288 sensitivity factor is simply $\beta_{r,B}$. All the β and $\beta_{r,A-B}$ are displayed in Figure 6.

289 **3.2 Response of surface SO₂ and sulfate concentrations**

290 In the S1 scenario, a uniform 8% of reduction in SO₂ emissions (totally 2.3Tg)

291 over China results in a 7.9% and 5.7% decrease of SO₂ and sulfate concentration,
292 respectively, and the corresponding national mean efficiency factor β is 0.99 for SO₂
293 and 0.71 for sulfate (Figure 6a). Sulfate efficiency factor is smaller than that of SO₂,
294 indicating the nonlinear response of sulfate to SO₂ emission change due to chemistry
295 and transport. The reduction of regional-mean sulfate concentration ranges from 6.2%
296 in SC ($\beta_{r,SC} = 0.78$) to 7.2% in NC ($\beta_{r,NC} = 0.9$). The regional efficiency factors of
297 sulfate over NC, SC and SWC are all greater than the national-mean value, indicating
298 higher emission control efficiency for sulfate by reducing SO₂ emissions from regions
299 with higher emissions. Seasonally, sulfate concentration decrease is smaller in winter
300 than summer at all three regions, indicating that SO₂ emission changes have larger
301 influence on sulfate in summer.

302 In the S2 scenario, SO₂ emissions from NC are reduced by 21.3%, equivalent to
303 a reduction of 2.3 Tg or 8% of the national total, while emissions from the rest of the
304 country remain unchanged. SO₂ concentration decrease is 19.6% for NC ($\beta_{r,NC-NC}=0.92$),
305 2.5% for SC ($\beta_{r,NC-SC}=0.08$), and 0.9% for SWC ($\beta_{r,NC-SWC}=0.04$). The
306 annual-mean efficiency factor is 1.0 for national mean surface SO₂. The S2 scenario
307 results in a 14.4%, 4.9% and 3.0% decrease of annual-mean sulfate concentrations
308 over NC, SC, and SWC, respectively. β for national mean sulfate concentration is 0.76,
309 larger than that in S1 (0.71). The regional sulfate efficiency factors (β_r) to SO₂
310 emission change over NC are: $\beta_{r,NC-NC} = 0.68$, $\beta_{r,NC-SC} = 0.23$, and $\beta_{r,NC-SWC} = 0.14$
311 (Figure 6b and c). The $\beta_{r,NC-NC}$ of both SO₂ and sulfate in the S2 scenario is smaller
312 than $\beta_{r,NC}$ in S1, because the S1 scenario includes reduced transport of SO₂ and sulfate

313 resulting from decreased emissions outside NC. The fact that $\beta_{r, NC-SC}$ and $\beta_{r, NC-SWC}$
314 are both significantly larger than zero indicates the important impact of NC emissions
315 on **SO₂ and sulfate** concentrations over other regions by way of inter-regional
316 transport. The fact that $\beta_{r, NC-SC}$ is 64% larger than $\beta_{r, NC-SWC}$ provides a clear indication
317 that SO₂ emissions from NC has a larger influence on **SO₂ and sulfate** concentrations
318 over SC than those over SWC (Figure 6c). **The inter-regional efficiency factors ($\beta_{r, NC-SC}$
319 $\beta_{r, NC-SWC}$) for sulfate is much larger than those for SO₂ reflecting the longer
320 atmospheric lifetime of sulfate.**

321 In the S3 scenario in which SO₂ emissions from SC are reduced by 2.3 Tg or
322 29.5%, the efficiency factor is **0.94 and 0.69 for national mean SO₂ and sulfate**
323 concentration, respectively, both smaller than the corresponding values in S1 and S2.
324 **For SO₂, there is a 25.1% decrease of SO₂ concentrations over SC and the**
325 **corresponding $\beta_{r, SC-SC}$ is 0.85. Because of the short lifetime of SO₂ in the atmosphere,**
326 **the SO₂ inter-regional efficiency factors are much smaller ($\beta_{r, SC-NC} = 0.08$, and $\beta_{r, SC-SWC} = 0.04$).** Sulfate concentrations decrease by 14.8% over SC, and the
327 **corresponding efficiency factor $\beta_{r, SC-SC}$ is 0.50. Compared with $\beta_{r, NC-NC}$ of 0.68 in S2,**
328 **the lower $\beta_{r, SC-SC}$ in S3 indicates that sulfate over SC is less sensitive to within-region**
329 **SO₂ emission change than that over NC. The regional efficiency factor of sulfate over**
330 **NC to changing SC emissions ($\beta_{r, SC-NC}$) is 0.14 for the annual mean, lower than $\beta_{r, NC-SC}$ of 0.23 derived from S2. Seasonally, SO₂ emissions from SC have a larger**
331 **influence on sulfate over NC in summer ($\beta_{r, SC-NC} = 0.16$) than winter ($\beta_{r, SC-NC} = 0.11$),**
332 **because of different prevailing wind directions in the two seasons. Sulfate**

335 concentrations over SWC have a small sensitivity to changing SO₂ emissions from SC,
336 with a $\beta_{r, SC-SWC}$ of only 0.05.

337 The S4 scenario, which reduces SO₂ emissions from SWC by 48.9%, has the
338 least effect on national mean sulfur (both SO₂ and sulfate) concentration ($\beta = 0.91$ for
339 SO₂ and 0.68 for sulfate) despite of a 48.9% reduction of SO₂ emissions from SWC
340 where the emission reduction is implemented. Sulfate concentrations over SWC have
341 a relatively small efficiency factor to within-region SO₂ emission changes with the $\beta_{r,}$
342 SWC-SWC of 0.50. $\beta_{r, SWC-NC}$ and $\beta_{r, SWC-SC}$ are both less than 0.05 for sulfate, indicating
343 the limited impact of SO₂ emissions on other regions due to the terrain of Sichuan
344 Basin.

345 To summarize the above analysis of the four emission scenarios, we find that
346 among the three regions selected, national mean surface concentrations of both SO₂
347 and sulfate are most sensitive to SO₂ emission changes from NC. SO₂ emissions from
348 NC are 36% and 129% higher than those from SC and SWC, respectively. Because of
349 the short lifetime of SO₂, reducing SO₂ emissions from one region has a small effect
350 on SO₂ concentrations over the other regions, and thus the national-mean SO₂
351 concentration is most sensitive to SO₂ emissions from NC. Sulfate over NC shows the
352 largest sensitivity to within-region emission changes ($\beta_{r, NC-NC} = 0.68$, compared with
353 $\beta_{r, SC-SC}$ of 0.50 and $\beta_{r, SWC-SWC}$ of 0.50). Sulfate has a longer atmospheric lifetime than
354 SO₂ and can be transported over long distance. SO₂ emission reductions over NC thus
355 have the largest influence on sulfate over adjacent regions with $\beta_{r, NC-SC}$ and $\beta_{r, NC-SWC}$
356 the largest among the regional efficiency factors of inter-regional transport. As a result,

357 the national-mean β of sulfate is the highest in the S2 scenario (0.76), followed by S1
358 (0.71), and the mechanism to explain this regional difference will be further discussed
359 in Section 4. The above analysis indicates that a nationwide uniform reduction of SO₂
360 emissions may not be the most effective way to reduce surface SO₂ and sulfate
361 concentrations, and SO₂ emission reduction over NC should receive a higher priority
362 in the national policies.

363 3.3 Response of population-weighted sulfate concentration

364 Compared with the area-mean sulfate concentration, the population-weighted
365 sulfate concentration (PWC) is a better metric to reflect the public exposure to
366 atmospheric sulfate aerosols because it considers the spatial heterogeneity of
367 population distribution. The PWC is calculated for each province or region by two
368 steps (Stedman et al., 2002): first multiplying the surface sulfate concentration by the
369 population data for individual model grids, then summing up the values of all grids
370 within a province or region and dividing the sum by the total population to get the
371 PWC for each province or region. The population data over China were adopted from
372 SEDAC (SocioEconomic Data and Applications Center,
373 <http://sedac.ciesin.columbia.edu/data/collection/gpw-v3>) for the year 2010. The
374 original population data from the SEDAC database is at the resolution of about 5 km x
375 5 km ((1/24)° × (1/24)°). We regrid them to the resolution of 0.5° × 0.667° to match
376 with that of the sulfate concentrations from the model.

377 The annual mean sulfate concentrations and provincial PWC are displayed in
378 Figure 7. Higher sulfate concentrations and PWCs occur over the east and southwest

379 part of China, in accordance with the spatial distribution of anthropogenic SO₂
380 emissions (c.f. Figure 1). The annual mean sulfate concentration is highest over SWC
381 (9.9 μg m⁻³), followed by NC (9.7 μg m⁻³) and SC (8.1 μg m⁻³). The annual mean
382 PWC over NC, SC and SWC is 11.2 μg m⁻³, 9.8 μg m⁻³ and 12.7 μg m⁻³, respectively.
383 The highest provincial PWC is the Sichuan province (including Chongqing) in SWC
384 and the Hubei province in SC. The correlation between sulfate concentration and
385 population density is stronger over SC and SWC than that over NC. As a result, SWC
386 and SC exert higher weightings in the PWC metric than in the **area-mean**
387 concentration metric.

388 The effects of the four emission scenarios on PWC in China are summarized in
389 Table 2. The S3 scenario shows the largest decrease of PWC, with an 8.3% reduction
390 in mean PWC of the three regions and 7.8% for the whole country. The S2 scenario
391 has the second-largest impact, with the corresponding change of 7.6% and 7.5%
392 respectively. The efficiency factor of the national mean PWC is highest in S3 (0.98)
393 and lowest in S1 (0.81). This regional difference indicates that SO₂ emission
394 reductions in SC (i.e., S3) is the most effective way to reduce human exposure to
395 ambient sulfate aerosols, while the national-mean scenario (S1) is the least effective.

396 **3.4 Response of sulfur outflow to the Pacific**

397 **Previous studies have found that pollution outflow from East Asia to the Pacific**
398 **peaks in spring (Chin et al., 2007; Clarisse et al., 2011), while winter is also a**
399 **significant contributor to the annual outflow flux (Feng et al., 2007). We calculate**
400 **the sulfur flux to Western Pacific in each scenario for the winter and spring seasons-**

401 ~~when the export of pollutants from China is most significant.~~ The transport flux is
402 evaluated at the boundary of 123 E and 22 N - 42 N within the troposphere and the
403 sulfur flux includes both SO₂ and sulfate. We define eastward flux as positive. The
404 sulfur fluxes for both seasons are positive, indicating net export of sulfur compounds
405 from China to the Western Pacific. Figure 8a displays the seasonal fluxes of each
406 scenario. The standard simulation gives a flux of 490 kt S month⁻¹ in winter and 450
407 kt S month⁻¹ in spring. Compared with the standard simulation, the S1 scenario shows
408 a 5.4% decrease of sulfur flux in winter and 5.3% in spring, with the mean value of β
409 being 0.67 for average fluxes of the two seasons. The S2 scenario shows the largest
410 sulfur flux decrease of 7.2% in winter and 8.0% in spring and mean β of 0.95,
411 indicating that SO₂ emission control over NC has the strongest effects on reducing
412 sulfur fluxes to the Western Pacific. The S3 and S4 scenarios have a much smaller
413 impact compared to S1 and S2, and the S3 scenario results in the least response ($\beta =$
414 0.50). This can be explained by the contribution of each region to the transport fluxes,
415 which will be discussed in Section 4.2.

416 In summary, the comparison between the different spatial-realizations of the
417 same amount of SO₂ emission reduction for China reveals different impacts not only
418 by region but also by the impact metrics of choice. Reducing SO₂ emissions over NC
419 results in the highest efficiency in reducing surface sulfur concentration in China as a
420 whole with an efficiency factor of 1.0 for SO₂ and 0.76 for sulfate as well as in
421 reducing transport of sulfur to the Western Pacific with the mean β of 0.95. On the
422 other hand, the sensitivity of population-weighted sulfate concentration is highest in

423 the S3 scenario ($\beta = 0.98$), so SO₂ emission control in SC is most effective to reduce
424 human exposure to sulfate aerosols over China.

425 **4. Regional differences in sulfur chemistry and transport**

426 To better understand mechanistically the regional differences in the efficiency factors
427 presented in Section 3, we investigate in this section the regional differences in the
428 conversion of SO₂ to sulfate and inter-regional transport of the major sulfur
429 compounds (SO₂ and sulfate) on the basis of GEOS-Chem model outputs. Sensitivity
430 tests of our findings to meteorology and emissions are also presented.

431 **4.1 SO₂ conversion to sulfate**

432 We have presented in Section 3.2 that the sensitivity of sulfate concentrations to
433 local SO₂ emission changes differs from region to region, and the regional efficiency
434 factor of SO₂ emission control is larger in NC ($\beta_{r, NC-NC}$) than other regions (i.e., $\beta_{r, SC-SC}$
435 and $\beta_{r, SWC-SWC}$). Here we attribute this difference to regional characteristics of
436 sulfate chemistry using the GEOS-Chem model. As described in Section 2.1, in the
437 model SO₂ is oxidized by OH to form sulfate in the gas phase, or by H₂O₂ and O₃ in
438 the aqueous phase. The two pathways are the main source of atmospheric sulfate. The
439 direct oxidation of SO₂ by O₂ catalyzed by transition metals (Alexander et al., 2009)
440 and the heterogeneous reaction of SO₂ on pre-existing aerosols (Wang Y. et al., 2014)
441 are not included in the current simulation. Globally aqueous phase oxidation plays a
442 larger role than gas phase oxidation in sulfate formation (Unger et al., 2006), while
443 their relative contributions vary regionally and seasonally. In the standard simulation,

444 aqueous phase oxidation of SO₂ contributes 45%, 64% and 73% to sulfate over NC,
445 SC and SWC, respectively. The lower atmospheric humidity and stronger NO_x
446 emissions over NC are two important factors responsible for the higher percentage of
447 gas phase SO₂ oxidation in this region. H₂O₂ oxidation makes up more than 90% of
448 aqueous phase oxidation for all the three regions. Barth and Church (1999) reported a
449 more than 80% contribution of aqueous phase oxidation to sulfate over Southeast
450 China. Increasing emissions of NO_x and hydrocarbons from China since the 1990s are
451 expected to increase the relative importance of gas phase oxidation for sulfate
452 (Berglen et al., 2004; Unger et al., 2006), which explains the lower fraction of
453 aqueous oxidation in our study.

454 Table 3 presents the relative changes of SO₂ emissions, SO₂ oxidation rate (gas
455 and aqueous phase and their total), and sulfate concentrations in the regional-specific
456 scenarios (S2-S4) compared with the standard simulation. Reducing SO₂ emissions
457 has different influences on gas and aqueous oxidation. Over all three regions, the
458 relative decrease of gas phase oxidation is greater than that of aqueous oxidation, so
459 the region with a higher contribution from gas phase oxidation will show a larger
460 sensitivity of sulfate to SO₂ emissions reduction; in our study, NC is the region with
461 the largest fraction of gas phase oxidation. Adopting the relationship between the
462 impact metric and SO₂ emission changes defined in Section 3, we derive the regional
463 sensitivity of SO₂ oxidation to be 0.76 for NC (S2), larger than that of 0.67 for SC (S3)
464 and 0.64 for SWC (S4), explaining the larger response of sulfate over NC to
465 within-region SO₂ emissions than the other two regions.

466 SO_2 emission changes affect both gas- and aqueous-phase oxidation processes,
467 but the magnitude of the influence depends on whether the process is SO_2 -limited or
468 not. In most-polluted areas with high NO_x emissions (like China), the aqueous
469 oxidation tends to be oxidants limited rather than SO_2 -limited because of the impact
470 of high- NO_x concentrations on OH, H_2O_2 , and O_3 . Previous sensitivity studies
471 (Berglen et al., 2004) have found that the gas-phase oxidation is more limited by SO_2 .
472 Therefore, SO_2 emission change will have a stronger impact on gas phase oxidation
473 than aqueous phase oxidation and this explains why when SO_2 emissions decrease, the
474 relative decrease of gas phase oxidation is larger than that of aqueous phase
475 oxidation. ~~Roelofs et al. (1998) and Berglen et al. (2004) compared the simulated~~
476 ~~sulfate concentrations from a model simulation using prescribed concentrations of~~
477 ~~oxidants with those using coupled oxidants. They showed that over regions with high~~
478 ~~SO_2 emissions, simulated sulfate concentration with fixed oxidants are higher than~~
479 ~~that from the simulation using coupled oxidants. They also found that in the run with~~
480 ~~prescribed oxidants, H_2O_2 oxidation became more important, while gas phase OH~~
481 ~~oxidation was little affected. Their results indicate that regions with high SO_2~~
482 ~~emissions (like China) are oxidation limited, especially for the aqueous phase H_2O_2~~
483 ~~oxidation pathway. Under the oxidation limited conditions, the oxidation rate of SO_2~~
484 ~~is less sensitive to SO_2 emission changes, so changing SO_2 emissions will have a~~
485 ~~larger influence on gas phase oxidation than aqueous phase oxidation. Since the~~
486 ~~proportion of gas phase oxidation in NC (55%) is much larger than that in SC (36%)~~
487 ~~and SWC (27%), the total SO_2 oxidation rates in NC is more sensitive to SO_2~~

488 emission changes than those over SC and SWC.

489 **4.2 Sulfur transport**

490 The decrease of sulfate concentrations for each region is less than the extent that
491 can be attributed to within-region SO₂ oxidation and the difference is due to the
492 transport of sulfur between regions. To further separate the impact of within-region vs.
493 inter-regional transport on sulfate concentrations by region, we conducted additional
494 model experiments in which SO₂ emissions from NC, SC and SWC are zeroed off
495 separately. The difference of sulfate concentrations between the standard simulation
496 and each of the zeroing-off model experiments represents the contribution of SO₂
497 emissions from the corresponding region with zero emissions. The resulting
498 decomposition of monthly mean sulfate concentrations by SO₂ source region is
499 displayed in Figure 9 for NC, SC, and SWC separately.

500 For the annual average, within-region SO₂ emissions contribute 68% of sulfate
501 concentrations over NC (Figure 9a), followed by 15% from SC emissions. Seasonally,
502 contributions from SC emissions on NC sulfate range from 10% during winter to 17%
503 during summer. SO₂ emissions from SWC have a small influence (4%) on sulfate over
504 NC, and the remaining 13% of sulfate over NC is from other source regions. For SC
505 (Figure 9b), only 59% of sulfate comes from SO₂ emitted within SC. NC is an
506 important source region for sulfate over SC, contributing 23% annually and ranging
507 from 11% in winter to 30% in summer. Transport from SWC has a very small
508 contribution of only 4%. For SWC, within-region emissions provide 61% of sulfate
509 (Figure 9c), while transport from NC and SC contributes 10% and 8%, respectively,

510 with the remaining 21% from other source regions. The combined contribution of
511 inter-regional transport among the three regions is 19%, 27%, and 18% to sulfate over
512 NC, SC, and SWC, respectively. The shorter lifetime of sulfate over SC and SWC
513 makes it harder to transport over long distances to downwind regions, so among all
514 the inter-regional transport of sulfate examined here, SO₂ emissions from NC exert
515 the largest impacts to sulfate concentrations in other regions, contributing 23% and 10%
516 to sulfate over SC and SWC, respectively. This explains why $\beta_{r, NC-SC}$ and $\beta_{r, NC-SWC}$
517 derived from the S2 scenario are larger than the regional sensitivity factors of
518 inter-regional transport in other emission scenarios. As a result, for a given amount of
519 SO₂ emission reduction in China, implementing it over NC is found most effective in
520 getting the largest benefit of reducing surface sulfate concentrations over China as a
521 whole.

522 We further quantify the contribution of each region to the transport fluxes of total
523 sulfur (SO₂ + sulfate) to the Western Pacific for winter, spring and the mean of the
524 two seasons (Figure 8b). NC is the largest contributor and contributes ~40% of total
525 fluxes for each season. This explains the largest sensitivity factor of the export fluxes
526 to NC emissions in the S2 scenario ($\beta = 0.95$). The contribution from SWC is the
527 second largest, being 17% in spring and 24.1% in winter and a mean of 20.7% for the
528 two seasons. Most of the export fluxes from SWC are found above the boundary layer
529 and as such they have a small effect on surface sulfate concentrations over NC or SC.
530 SC contributes the least (20%) to the export fluxes, resulting in the smallest sensitivity
531 factor of sulfur export flux to the Western Pacific ($\beta = 0.50$) in the S3 scenario.

532 4.3 Robustness test

533 The chemistry of SO₂ conversion to sulfate and the transport of sulfur
534 compounds are dependent on both meteorology and emissions. We used a single
535 year's meteorology and emissions (2010) to derive the efficiency factors. To assess
536 the uncertainty of our analysis to the choice of meteorology and the magnitude of
537 emission reductions, sensitivity tests were carried out by changing the year of
538 meteorology to 2009 and by doubling the amount of SO₂ emissions reductions. The
539 national mean sensitivity factor of surface sulfate concentration, population-weighted
540 sulfate concentration and eastward transport fluxes are calculated and compared with
541 that from the S1-S4 emission reduction scenarios.

542 First, to examine the sensitivity of the model results to the meteorological fields,
543 we conducted a series of one-year test simulations with the 2009 meteorology. In the
544 tests, we used the same emissions as in the standard simulation and the S1-S4
545 scenarios. With the 2009 meteorology, the national mean SO₂ and sulfate
546 concentration and eastward transport fluxes are also most sensitive to SO₂ emission
547 reduction from NC (Figure 6a, green short line). The discrepancy in the value of β
548 between simulation with 2009 and 2010 meteorology is within 5%. The efficiency
549 factors for national mean SO₂ and sulfate concentration and sulfur flux are 1.0, 0.78
550 and 0.94, respectively, compared with the 1.0, 0.76 and 0.95 from the 2010
551 meteorology. SO₂ emission reduction in SC is most effective in reducing PWC with
552 the national mean β of 0.94 with the 2009 meteorology, compared to the
553 corresponding value of 0.98 from the 2010 meteorology.

554 Second, the magnitude of SO₂ emissions reduction is doubled in each of the
555 S1-S4 scenarios to check the sensitivity of model results to emissions. These tests are
556 run for one year and with the 2010 meteorology. The efficiency factor for national
557 mean SO₂ and sulfate concentration, PWC and sulfur transport fluxes to the Western
558 Pacific from this test are displayed in Figure 6a with yellow short line. When SO₂
559 emission reduction is doubled, the national mean SO₂ and sulfate concentration, and
560 the export sulfur fluxes are also most sensitive to SO₂ emission reduction from NC,
561 and β for national mean PWC is the largest when SO₂ emission reduced from SC.
562 However, there is a relatively significant decrease in the value of β , especially for
563 national mean sulfate concentration and PWC (more than 20%). This indicates that
564 SO₂ oxidation rate becomes less sensitive when SO₂ emission reduction is greater.

565 Furthermore, SO₂ emission control may accompany with changing emissions of
566 NO_x and VOCs because these pollutants have common sources as SO₂. NO_x and
567 VOCs are precursors of tropospheric O₃, and their emissions change can influence the
568 concentrations of H₂O₂, O₃ and OH. While coal combustion is the dominant source of
569 SO₂, it is not the most important source for NO_x or VOCs (transportation being
570 another equally important source for those species). In addition, the technologies used
571 to control SO₂ emissions from coal power plants cannot remove NO_x or VOCs to the
572 same extent as SO₂. To address the impact of changing emissions of co-emitted
573 species on SO₂ chemistry, we conducted a third set of sensitivity tests considering the
574 extreme circumstance in which NO_x and VOCs emissions are also decreased by 8%,
575 equal to the magnitude of SO₂ emission change in the S1 scenario. The results from

576 this sensitivity test are within $\pm 2\%$ of those from S1 for all the metrics we considered.
577 For example, the decrease of national mean SO₂ and sulfate concentrations is 7.80%
578 and 5.76%, respectively; the corresponding value is 7.90% and 5.70% from the S1
579 simulation. This indicates that the change in NO_x and VOCs emissions as a result of
580 SO₂ emission control processes has a negligible impact on SO₂ oxidation and as such
581 it will not affect the conclusion of this study.

582 In summary, we find SO₂ emissions reduction has a larger influence on gas phase
583 oxidation than aqueous phase oxidation. Because sulfate in NC has the largest relative
584 contribution from gas phase oxidation, NC shows the largest sensitivity to
585 within-region SO₂ emission changes. Besides, inter-regional transport contributes 18%
586 ~ 27% of sulfate over the three regions. SO₂ emissions from NC contribute 23% and
587 10% to sulfate over SC and SWC, respectively, which are the largest among all the
588 inter-regional transport of sulfate. This explains why $\beta_{r, NC-SC}$ and $\beta_{r, NC-SWC}$ are the
589 largest among all the efficiency factors to external emission changes (Section 3.2 and
590 Figure 6c). SO₂ emissions from NC contribute the most (~40%) to the sulfur transport
591 fluxes from China to the Western Pacific, resulting in the largest sensitivity factor
592 (0.95) of the transport flux in the S2 scenario. Contribution from SC is the least (20%)
593 and thus SO₂ emission reduction from SC has the least influence on the transport flux.
594 The robustness tests demonstrate that the ranking of different scenarios are robust
595 with respect to different meteorology year, different magnitude of SO₂ emission
596 reduction, and changing emissions of the co-emitted species (NO_x and VOCs) as SO₂.

597 5. Conclusion and discussion

598 We have designed and compared model sensitivities in which the same amount
599 of SO₂ emission reduction (2.3 Tg, 8% of total SO₂ emission from China in 2010,
600 following the 12th FYP) is implemented uniformly in China as a whole (S1) and in
601 three sub-regions only (NC, SC and SWC) to investigate the emission control
602 efficiencies in different regions. The GEOS-Chem chemical transport model is used in
603 this study to quantify the response of different concentration and flux metrics to SO₂
604 emissions change.

605 National mean and inter-regional efficiency factors (β and β_r) are defined as the
606 percentage change of the concerned metrics caused by a 1% decrease of SO₂ emission
607 changes. The metrics include surface SO₂ and sulfate concentrations, the
608 population-weighted sulfate concentration and sulfur transport from China to the
609 Western Pacific. SO₂ emission reduction from NC (S2 scenario) has the largest
610 influence on national mean SO₂ concentration with the efficiency factor of 1.0. The
611 S2 scenario is also most effective in reducing the mean sulfate concentration over
612 China as a whole with the highest national-mean β of 0.76, which can be explained in
613 two aspects. On one hand, SO₂ oxidation in gas phase is found to be more sensitive to
614 the change of SO₂ emissions than aqueous phase oxidation, and NC is the region with
615 the largest fraction of gas phase SO₂ oxidation. This makes sulfate over NC most
616 sensitive to within-region emission changes with the largest efficiency factor ($\beta_{r, NC-NC}$
617 = 0.68, $\beta_{r, SC-SC} = 0.5$, $\beta_{r, SWC-SWC} = 0.50$). On the other hand, comparison of
618 inter-regional sulfate transport reveals that SO₂ emissions from NC exert the largest
619 impacts to sulfate concentrations in other regions (23% for SC and 10% for SWC).

620 This leads to $\beta_{r, NC-SC}$ and $\beta_{r, NC-SWC}$ being the largest among the regional efficiency
621 factors of inter-regional transport.

622 Among the three regions, NC contributes most (~40%) to the transport fluxes of
623 sulfur from China to the Western Pacific, so the Western Pacific region will benefit
624 most from SO₂ reduction in NC with the mean β of 0.95. Contribution from SC is the
625 least among the three regions studied here, resulting in the smallest efficiency factor
626 of sulfur export flux to the Western Pacific ($\beta = 0.50$) in the S3 scenario. We also find
627 that SO₂ emission control in SC is most effective to reduce human exposure to sulfate
628 aerosols over China as a whole, as indicated by the highest sensitivity of
629 population-weighted sulfate concentration in the S3 scenario ($\beta = 0.98$). The
630 efficiency factors and their spatial differences are found to be robust and not
631 dependent on the year of meteorology, the magnitude of SO₂ emissions change **or the**
632 **change in emissions of co-emitted NO_x and VOCs.**

633 **Based on the analysis above,** we recommend that a nationwide uniform reduction
634 of SO₂ emissions may not result in the largest emission control efficiency.
635 **Considering that NC makes the largest contribution to inter-regional transport of**
636 **sulfur within China and to the transport fluxes to the Western Pacific, SO₂ emission**
637 **reduction over NC should receive a higher priority in the national policies in order to**
638 **maximize the air quality benefits for China and downwind regions. However, from**
639 **2006 to 2010 (the 11th Five-Year Plan period), SO₂ emissions from NC have**
640 **decreased at a much slower rate than the national total emissions. Based on the MEIC**
641 **inventory, total SO₂ emissions from China were 9.4% lower in 2010 than 2006, and**

642 emissions from NC, SC and SWC have decreased by 4.7%, 16.1% and 23.1%,
643 respectively, during the same period. The relative reduction of SO₂ emissions in NC is
644 thus one third or less of that for the other two regions and is only half of the reduction
645 at the national mean level. This indicates that China has not prioritized SO₂ emission
646 control in NC in the past. Our study suggests this should be corrected in the future in
647 order to maximize the benefits of SO₂ control. ~~SO₂ emission reduction over NC~~
648 ~~should receive higher priority in the national policies to reduce surface SO₂ and~~
649 ~~sulfate concentration and sulfur transport fluxes to the Western Pacific, while SO₂~~
650 ~~emission reduction over SC is most favorable to reduce people's exposure to sulfate~~
651 ~~aerosols.~~

652

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656 thank the free use of surface measurements from EANET, AOD data from MODIS
657 and SO₂ columns data from OMI.

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789 **Tables**

790

791 **Table 1: simulation scenarios and SO₂ emission in each study**

Simulation scenario	description	SO ₂ emission (Tg)			
		China	NC	SC	SWC
standard	Standard, 2010 inventory	28.4	10.8	7.8	4.7
S1	SO ₂ emission is reduced uniformly across China by 8% (2.3 Tg)	26.1	9.9	7.2	4.3
S2	Implement 2.3 Tg SO ₂ reduction on NC, emission from other regions unchanged	26.1	8.5	7.8	4.7
S3	Implement 2.3 TgSO ₂ reduction on SC, emission from other regions unchanged	26.1	10.8	5.5	4.7
S4	Implement 2.3 TgSO ₂ reduction on SWC, emission from other regions unchanged	26.1	10.8	7.8	2.4

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796 **Table 2: change in population-weighted sulfate concentrations, μg m⁻³**

	standard simulation	Difference with the standard simulation			
		S1	S2	S3	S4
NC+SC+SWC	10.9	-6.7%	-7.6%	-8.3%	-7.1%
China	9.7	-6.6%	-7.5%	-7.8%	-6.9%

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801 **Table 3: percentage changes of SO₂ conversion to sulfate and sulfate concentration over NC,**

802 **SC and SWC in response to within-region SO₂ emission changes:**

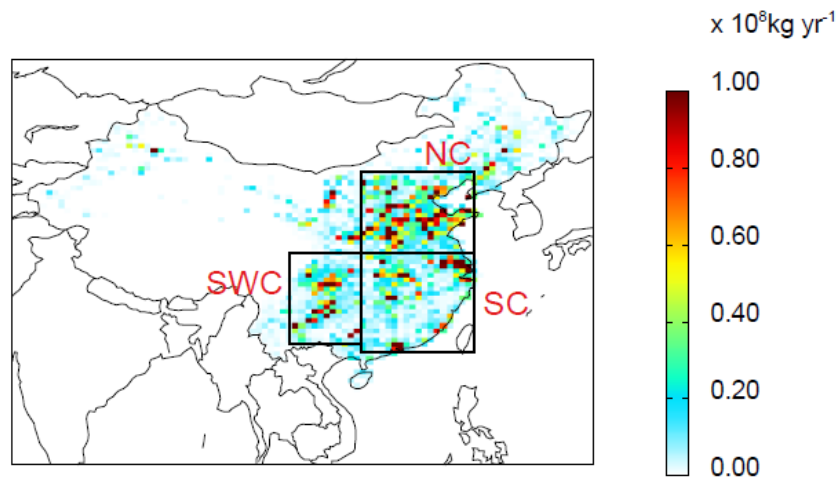
	SO ₂ emission	SO ₂ conversion			Sulfate concentration
		Gas phase	Aqueous phase	total	
NC (S2)	-21.3%	-20.6%	-10.4%	-16.1%	-14.4%
SC (S3)	-29.5%	-22.6%	-17.1%	-19.9%	-14.8%
SWC (S4)	-48.9%	-41.8%	-30.1%	-31.4%	-24.5%

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805 **Figures**

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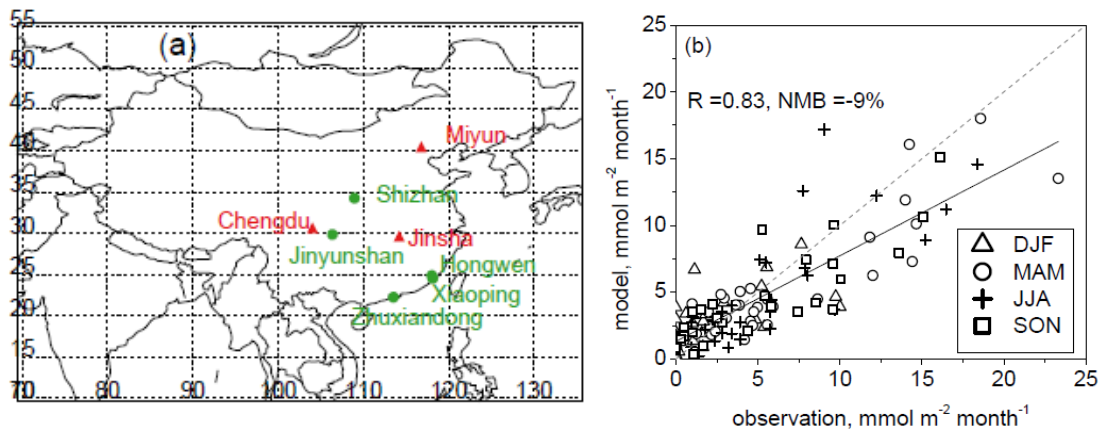
808 **Figure 1. SO₂ emissions from China in the year of 2010.**

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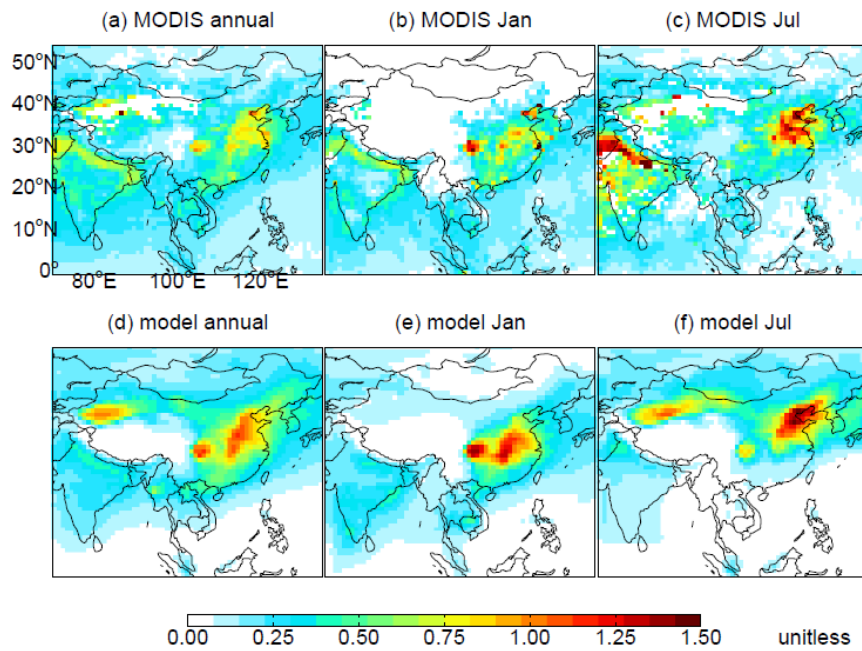
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814 **Figure 2. (a) Locations of observation sites. The red triangles represent sites with surface**
815 **sulfate concentration, and the green dots represent sites with sulfate wet deposition fluxes. (b)**
816 **Scatter plot of simulated versus observed sulfate wet deposition fluxes in 5 sites over China,**
817 **from January 2009 to December 2010.**

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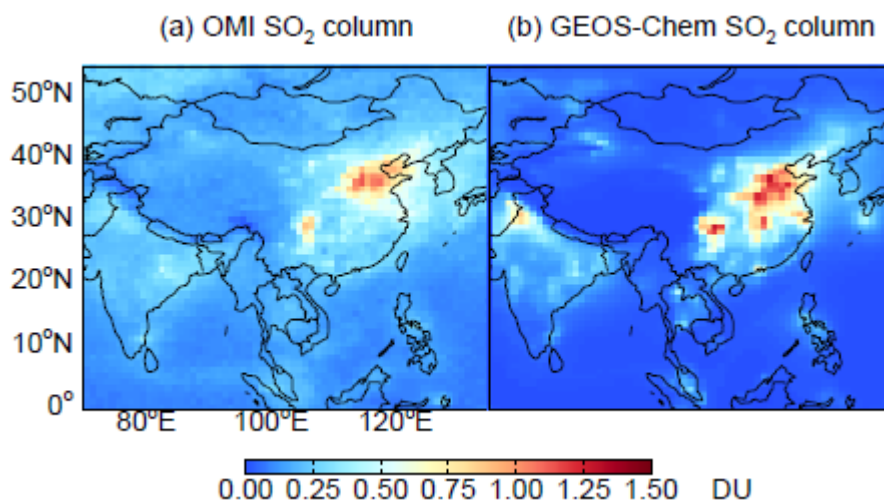


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820 **Figure 3.** Aerosol optical depth (AOD) over East Asia from MODIS for (a) 2010 annual mean,
 821 (b) January, and (c) July, and from the GEOS-Chem model: (d) annual mean, (e) January,
 822 and (f) July.

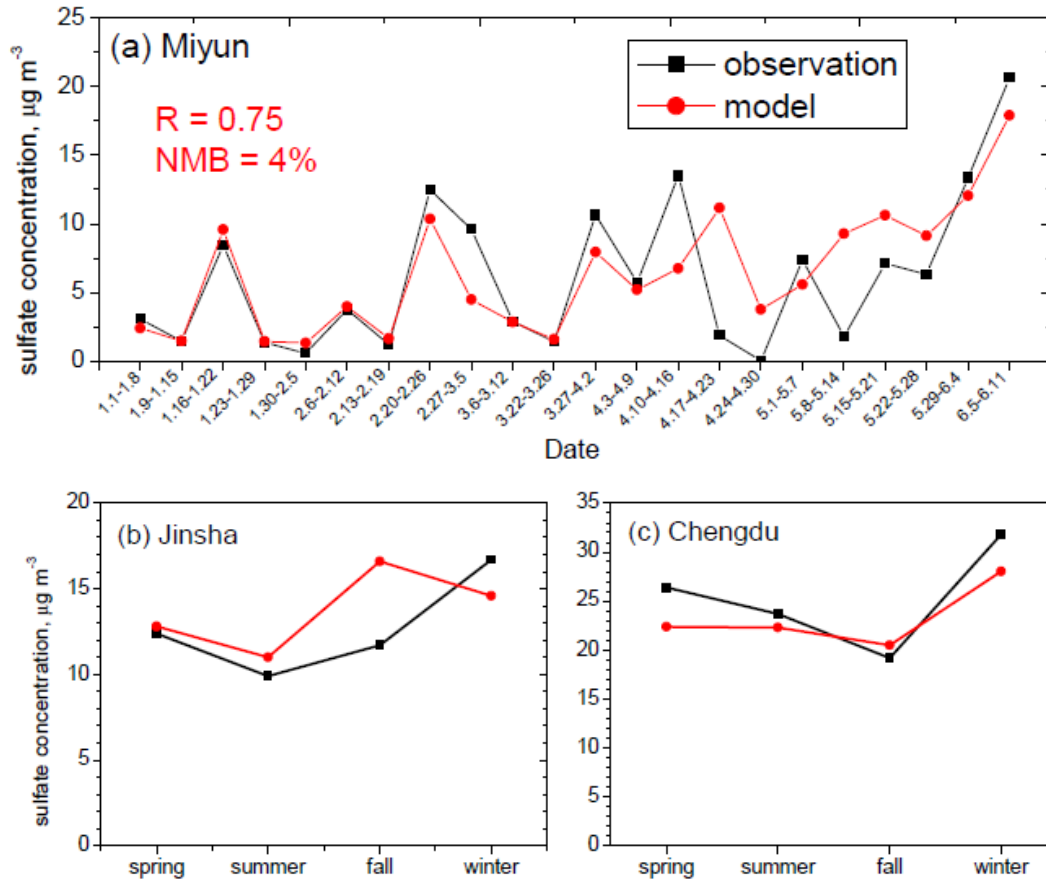
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826 **Figure 4.** Annual mean total SO₂ columns from (a) OMI satellite instrument and (b)
 827 GEOS-Chem simulation for the year 2010.



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829 **Figure 5. Comparison of observed (black line) and simulated (red line) surface sulfate**
 830 **concentrations at (a) Miyun site with weekly sulfate concentration from January to June in**
 831 **2010; (b) Jinsha site with seasonal mean sulfate concentration from Zhang et al. (2014), and**
 832 **the observation year is 2012; (c) Chengdu site with monthly mean sulfate concentration from**
 833 **Tao et al. (2014), the observation time is January, April, July, and October in 2011.**

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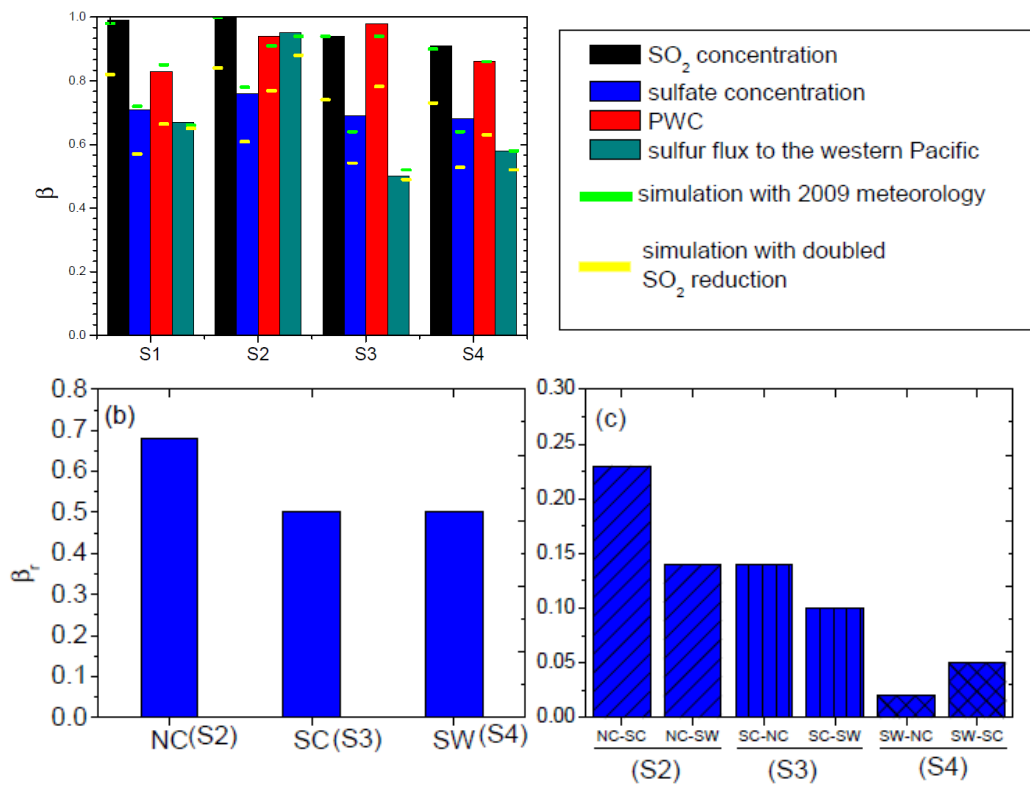
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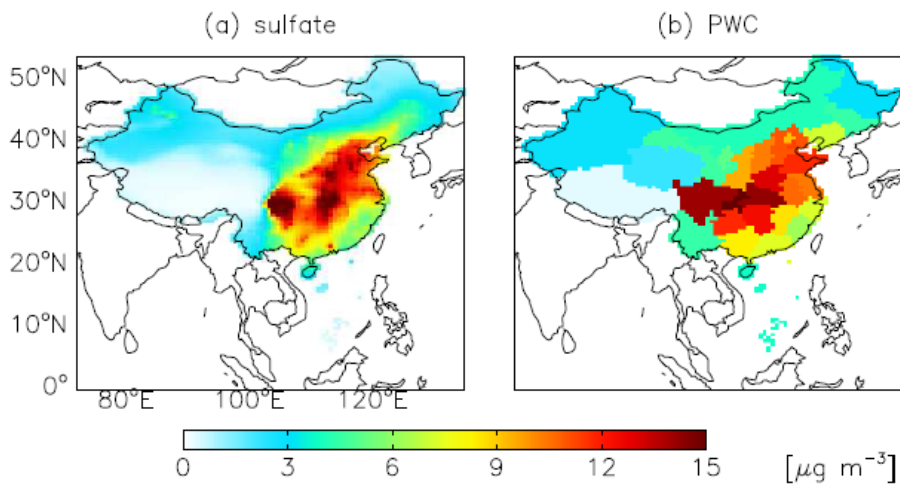
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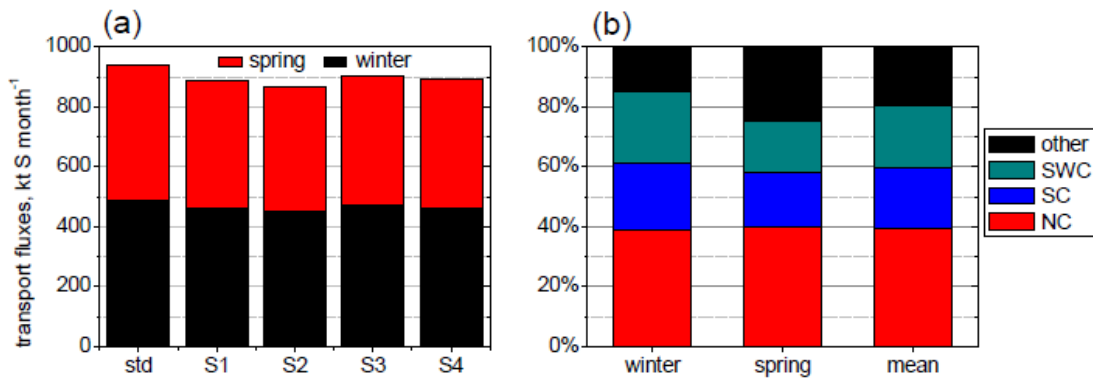
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842 **Figure 6. (a) Emission control efficiency factors (β) of national mean SO₂ and sulfate**
 843 **concentrations, population-weighted sulfate concentrations (PWC), and sulfur fluxes from**
 844 **China to the west Pacific in S1-S4 simulation scenarios. (b) Regional efficiency factors (β_r) of**
 845 **sulfate concentrations over NC, SC and SWC to within-region SO₂ emission changes. (c)**
 846 **Inter-regional efficiency factor by scenario. The efficiency factor of national mean sulfate**
 847 **concentrations, PWC, and eastward sulfur transport fluxes in the robustness sensitivity tests**
 848 **are presented in (a): the green short line represents results from simulation with meteorology**
 849 **for 2009, the yellow short line represents results from doubled magnitude of SO₂ emission**
 850 **reduction simulation.**



851 **Figure 7. Annual mean sulfate concentration and population-weighted sulfate concentration**
 852 **over China (sulfate and population data of Taiwan Province are not available).**

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855 **Figure 8. (a) Sulfur ($\text{SO}_2 + \text{sulfate}$) flux at the 123 E, 22°-42 N tropospheric plane from**
 856 **China to the Western Pacific, and (b) percentage contribution of NC, SC, SWC and other**
 857 **regions (the rest of Chinese regions as well as global influence) to sulfur ($\text{SO}_2 + \text{sulfate}$)**
 858 **transport fluxes from China to the Western Pacific.**

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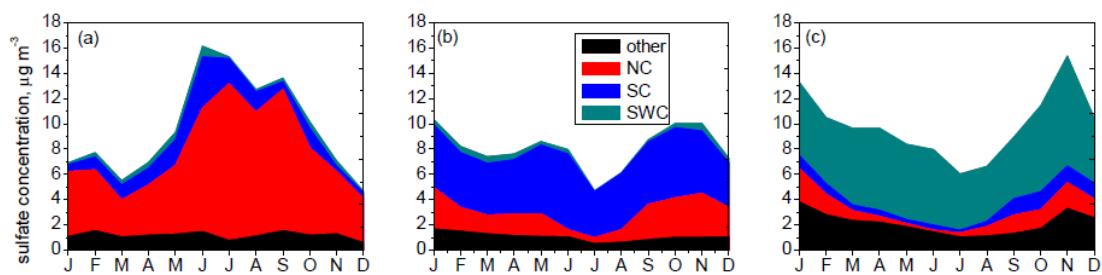
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867 **Figure 9. Monthly and regional mean sulfate concentrations over (a) NC, (b) SC and (c)**

868 **SWC, with contributions from within-region and inter-regional transport. Here “other”**

869 **includes the rest of Chinese regions as well as foreign influence.**

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