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Atmospheric Chemistry and Physics Discussions

Contributions of residential coal combustion to the air quality in Beijing-Tianjin-Hebei (BTH), China: A case study

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Abstract: In the present study, the WRF-CHEM model is used to evaluate contributions of the residential coal combustion (RCC) emission to the air quality in Beijing-Tianjin-Hebei (BTH) during persistent air pollution episodes from 9 to 25 January 2014. In general, the predicted temporal variations and spatial distributions of the air pollutants mass concentrations are in good agreement with observations at monitoring sites in BTH. The WRF-CHEM model also reasonably well reproduces the temporal variations of aerosol species compared with the AMS measurements in Beijing. The RCC emission plays an important role in the haze formation in BTH, contributing about 23.1% of $PM_{2.5}$ (fine particulate matter) and 42.6% of SO_2 during the simulation period on average. Organic aerosols dominate $PM_{2.5}$ from the RCC emission, with a contribution of 42.8%, followed by sulfate (17.1%). The air quality in Beijing is remarkably improved when the RCC emission in BTH and its surrounding areas is excluded in simulations, with a 30% decrease of $PM_{2.5}$ concentrations. However, when only the RCC emission in Beijing is excluded, the Beijing's $PM_{2.5}$ level is decreased by 18.0% on average. Our results suggest that implementation of the residential coal replacement by clean energy sources in Beijing is beneficial to the Beijing's air quality, but is not expected to bring back the blue sky to Beijing. Should the residential coal replacement be carried out in BTH and its surrounding areas, the air quality in Beijing would be improved remarkably. Further studies need be conducted considering the uncertainties in the emission inventory and meteorological fields.

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1 Introduction

Over the several past decades, China has experienced rapid economic growth, accompanied by accelerating industrialization and urbanization, which have seriously deteriorated air quality (e.g., R. Zhang et al., 2009; X. Zhang et al., 2012; L. Zhang et al., 2015). Recently, the haze pollution has become the primary concern about the air quality in most key regions and cities in China, especially in Beijing-Tianjin-Hebei (BTH) and Yangtze River Delta (YRD) (e.g., Wang et al., 2005; An et al., 2007; Wang et al., 2014; Chen et al., 2016; Gao et al., 2016). The severe and persistent haze pollution with high concentrations of fine particulate matter ($PM_{2.5}$) and the consequent low visibility, is mainly caused by heavy anthropogenic emissions and unfavorable synoptic situations (e.g., Seinfeld and Pandis, 2006; Lei et al., 2011; Lv et al., 2016; Wang et al., 2016; Zíková et al., 2016). According to the China's Ministry of Environment Protection (MEP), the annual mean $PM_{2.5}$ concentration was 102 μg m^{-3} in 2013 and 93 μg m^{-3} in 2014 in BTH, far beyond the World Health Organization (WHO) interim target-1 of 35 μg m^{-3} for the annual mean $PM_{2.5}$ concentration and also the secondary class standard in the China's new National Ambient Air Quality Standard (NAAQS, GB3095-2012). Therefore, the Chinese State Council has issued the "Atmospheric Pollution Prevention and Control Action Plan (APPCAP)" in September 2013 to reduce $PM_{2.5}$ by up to 25% by 2017 relative to 2012 levels. Since implementation of the APPCAP, stringent control strategies have been carried out to reduce the pollutants emissions from power plants, industries and the transportation (Sheehan et al., 2014; Liu et al., 2015; Yang et al., 2016). Control strategies have also been implemented to reduce residential emissions to the air quality, but the mitigation effect is still lack of evaluation constrained by observations.

The air pollution in China is a typical coal-smoke pollution, which is considered to be associated closely with China's special energy consumption structure (e.g., Quan et al., 2014; Archernicholls et al., 2016; Liu et al., 2016; Xue et al., 2016). Coal plays a key role in China's

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energy structure, and as the most abundant and a relatively cheap energy resource, coal is regarded as a dominant energy supply in China in the foreseeable future. According to the BP statistical review of world energy, from 1980s to the present day, the proportion of coal in China' primary energy production and consumption has been around 70%, which is much higher than that of around 20% in OECD (Organization for Economic Co-operation and Development) countries. Entering the 21 centuries, the coal consumption in China has increased sharply, and by 2013, China's coal consumption has accounted for 50.3% of the global coal consumption, which was 4.2 and 6.7 times higher than the United States and the European Union, respectively. In 2013, coal is responsible for 79%, 54%, 40%, 35%, 40%, and 17% of the SO_2, NOx, PM_{10}, $PM_{2.5}$, BC, and OC emissions in China, respectively (Ma et al., 2016).

Residential coal combustion (RCC) is recognized as a significant source of air pollution, affecting both local and regional air quality and posing serious threat to human health and environment by releasing hazardous air pollutants (HAPs), including PM, black carbon (BC), organic carbon (OC), SO_2, NO_x, CO, CO_2, and polycyclic aromatic hydrocarbons (PAHs) (e.g., Wornat et al., 2001; Ge et al., 2004; Zhi et al., 2008; Shen et al., 2010; Cheng et al., 2016; Li et al., 2016). Chemical transport models have been used to investigate the contribution of the RCC emission to the ambient air pollution in China. Using the CMAQ model, Y. Xue et al. (2016) have shown that the contributions of the RCC emission to the mass concentrations of PM_{10}, SO_2, NO_x, and CO are 11.6%, 27.5%, 2.8%, and 7.3%, respectively, during the winter heating season of 2012 in Beijing. Simulations using the GEOS-CHEM model by Q. Ma et al. (2016) have demonstrated that the coal combustion contributes 40% of the total $PM_{2.5}$ concentration on national average in 2013. Among major coal-burning sectors, industrial coal burning contributes 17% of the $PM_{2.5}$ concentrations, followed by power plants (9.8%) and domestic sector (4.0%). J. Liu et al. (2016) have used the WRF-CHEM model to simulate the

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air pollution in BTH in January and February 2010, indicating that annual elimination of residential sources in BTH reduces emissions of primary $PM_{2.5}$ by 32%, compared with 5%, 6%, and 58% of transportation, power plants, and industrial sectors, respectively.

In the present study, we use the Weather Research and Forecasting model coupled with chemistry (WRF-CHEM) to evaluate the contribution of the RCC emission to the air quality in BTH during persistent air pollution episodes from 9 to 25 January 2014. The WRF-CHEM model configurations and methodology are described in Section 2. Model results and discussions are represented in Section 3, and conclusions are given in Section 4.

2 Model and Methodology

2.1 WRF-CHEM model and configurations

The WRF-CHEM model used in the study is developed by Li et al. (2010, 2011a, b, 2012) at the Molina Center of Energy and the Environment, based on the previous studies (Grell et al., 2005; Fast et al., 2006). The wet deposition follows the method used in the CMAQ module and the dry deposition of chemical species is parameterized following Wesely (1989). The photolysis rates are calculated using the FTUV (fast radiation transfer model), including the aerosol and cloud effects on photolysis (Tie et al., 2003; Li et al., 2005, 2011a). The inorganic aerosols are calculated using ISORROPIA Version 1.7 (Nenes, 1998). The secondary organic aerosol (SOA) is predicted using the volatility basis-set (VBS) modeling method, with contributions from glyoxal and methylglyoxal. Detailed information can be found in Li et al. (2010, 2011b).

Persistent air pollution episodes from 9 to 25 in January 2014 in BTH are simulated using the WRF-CHEM model. During the study period, the averaged $PM_{2.5}$ concentration in BTH is 161.9 μg m^{-3}, with a maximum of 323.5 μg m^{-3}. The average temperature and relative humidity in Beijing during the period is −1.7℃ and 32.3%, respectively, and the average wind speed is

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about 2.8 m s^{-1}. The model simulation domain is shown in Figure 1, and detailed model configurations can be found in Table 1.

2.2 Statistical methods for comparisons

In the present study, we use the mean bias (MB), root mean square error (RMSE) and index of agreement (IOA) to validate the WRF-CHEM model performance in simulating air pollutants and aerosol species against observations and measurements. IOA describes the relative difference between the model prediction and observation, ranging from 0 to 1, with 1 indicating perfect agreement of the prediction with the observation.

$$MB = \frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)$$

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2\right]^{\frac{1}{2}}$$

$$IOA = 1 - \frac{\sum_{i=1}^{N}(P_i - O_i)^2}{\sum_{i=1}^{N}(|P_i - \bar{O}| + |O_i - \bar{O}|)^2}$$

Where P_i and O_i are the predicted and observed pollutant concentrations, respectively. N is the total number of the predictions used for comparisons, and $\bar{P}$ and $\bar{O}$ represent the average of predictions and observations, respectively.

2.3 Pollutants measurements

The hourly near-surface CO, SO_2, NO_2, O_3, and $PM_{2.5}$ mass concentrations released by the China's Ministry of Environmental Protection can be downloaded from the website http://www.aqistudy.cn/. The sulfate, nitrate, ammonium, and organic aerosols (OA) have been measured by the Aerodyne High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) with a novel $PM_{2.5}$ lens from 9 to 26 January 2014 at the Institute of Remote Sensing and Digital Earth (IRSDE), Chinese Academy of Sciences (40.00°N, 116.38°E) in Beijing (Figure 1) (Williams et al., 2013). The Positive Matrix Factorization (PMF) technique is used with constraints implemented in SoFi (Canonaco et al., 2013) to analyze the sources of OA and five components are separated by their mass spectra and time series. The components

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include hydrocarbon-like OA (HOA), cooking OA (COA), biomass burning OA (BBOA), coal combustion OA (CCOA), and oxygenated OA (OOA). HOA, COA, BBOA, and CCOA are interpreted for surrogates of primary OA (POA), and OOA is a surrogate for SOA. Detailed information about the HR-ToF-AMS measurements and data analysis can be found in Elser et al. (2016).

3 Results and discussions

3.1 Model performance

For the discussion convenience, we have defined the reference simulation in which the emissions from various sources are considered (hereafter referred to as REF), and results from the reference simulation are compared to observations in BTH.

3.1.1 Air pollutants simulations in BTH

Figure 2 presents the distributions of predicted and observed near-surface mass concentrations of $PM_{2.5}$, O_3, NO_2, and SO_2 along with the simulated wind fields averaged from 9 to 25 January 2014 in BTH. Generally, the predicted $PM_{2.5}$ spatial pattern is well consistent with the observations at the ambient monitoring sites in BTH. The WRF-CHEM model reasonably well reproduces the high $PM_{2.5}$ concentrations exceeding 150 μg m^{-3} in the plain region of BTH. Apparently, during the simulation period, the weak or calm winds in the plain region of BTH facilitate the accumulation of $PM_{2.5}$, causing severe air pollution. The average simulated $PM_{2.5}$ concentrations exceed 250 μg m^{-3} in the south of Hebei, which is generally in good agreement with observations. The observed and simulated O_3 concentrations are rather low in the plain region of BTH with the high $PM_{2.5}$ level, varying from 10 to 30 μg m^{-3}. There are several reasons for the low O_3 concentrations in the plain region of BTH. Firstly, during wintertime, the insolation is weak in the north of China, unfavorable for the O_3 photochemical production. Additionally, high $PM_{2.5}$ concentrations and frequent occurrence of clouds during

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haze days further attenuate the incoming solar radiation in the planetary boundary layer (PBL), decreasing the O_3 level. Secondly, calm or weak winds indicate stagnant situations, lacking the O_3 transport from outside of BTH. Thirdly, high nitrogen oxides (NO_x=NO+NO_2) emissions cause titration of O_3, which is shown by the high NO_2 concentrations in the plain region of BTH. The elevated NO_2 and SO_2 concentrations are observed and simulated in the plain region of BTH, particularly in cities and their surrounding areas, ranging from 50 to 100 μg m^{-3} and 50 to 150 μg m^{-3}, respectively. It is worth noting that the simulated NO_2 is generally distributed evenly in the plain region of BTH, indicating the dominant contribution of area sources.

Figure 3 presents the diurnal profiles of observed and simulated near-surface $PM_{2.5}$, O_3, NO_2, SO_2, and CO mass concentrations averaged over all monitoring sites in BTH from 9 to 25 January 2014. The WRF-CHEM model well reproduces the diurnal variations of the $PM_{2.5}$ mass concentrations compared with the observations in BTH during the simulation period. The *MB* and *RMSE* is -2.7 and 40.9 μg m^{-3}, respectively, and the *IOA* is 0.94. During the three haze episodes occurred in BTH, the model generally replicate well the haze developing stage, but tends to underestimate the $PM_{2.5}$ concentration compared to observations during the haze dissipation stage. One of the most possible reasons is the uncertainty of the simulated meteorological fields, which determine the formation, transformation, diffusion, transport, and removal of the air pollutants in the atmosphere (Bei et al., 2012, 2013). Should the predicted winds be intensified earlier than observations in BTH during the haze dissipation stage, the simulated $PM_{2.5}$ concentrations would fall off earlier, causing the model underestimation. The predicted NO_2 diurnal variations are generally well consistent with the observations, with a *MB* of 4.2 μg m^{-3} and an *IOA* of 0.93. The model also reasonable well yields the SO_2 and CO temporal variations with the *IOA* exceeding 0.85. However, the *RMSE* for SO_2 is rather large, showing considerable dispersions of the SO_2 simulations. A great deal of SO_2 are emitted from power plants or agglomerated industrial zones, which can be regarded as point sources, so the

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188 transport of SO_2 is more sensitive to uncertainties of the simulated wind fields.

189 **3.1.2 Aerosol species simulations in Beijing**

190 Figure 4 presents the temporal profiles of measured and simulated OA, CCOA, sulfate,
191 nitrate, and ammonium concentrations at IRSDE site in Beijing from 9 to 25 January 2014.
192 The model generally performs reasonably well in simulating the diurnal variations of the
193 aerosol species compared to the HR-ToF-AMS measurements, with *IOAs* exceeding 0.80. As
194 a primary species, OA is primarily determined by direct emissions from various sources,
195 including vehicles, cooking, biomass burning, coal combustion, and transport from outside of
196 Beijing, so uncertainties in emissions and the simulated meteorological fields markedly
197 influence the OA simulations (Bei et al., 2017). Although the *IOA* for OA is 0.84, the model
198 slightly overestimates the OA concentrations with a *MB* of 5.1 μg m^{-3}, and the dispersion of
199 OA simulations is also large, with a *RMSE* of 42.3 μg m^{-3}. In addition, the model fails to
200 reproduce the measured OA peaks during the nighttime on 11 and 17 January 2014, which is
201 perhaps caused by the emission uncertainty. The model also generally tracks the measured
202 diurnal variations of CCOA concentrations, with an *IOA* of 0.81. The model frequently
203 underestimates or overestimates the CCOA concentrations and is also subject to missing the
204 observed CCOA peaks. The CCOA is mainly emitted from industries and the residential coal
205 combustion. In general, the CCOA emissions from industries have clear diurnal variations, and
206 vice versa for those from residential coal combustion, causing large model biases for the CCOA
207 simulation. The simulated diurnal variations of sulfate, nitrate, and ammonium are also in good
208 agreement with the observations, with *IOAs* of 0.83, 0.87, and 0.90, respectively. The model
209 considerably overestimates the inorganic aerosol concentrations from 16 to 18 January. One of
210 the possible reasons is the decreased emissions, particularly from industries before the Chinese
211 New Year, which are not reflected in the emission inventory used in the study.

212 Figure 5 presents the contributions of aerosol species to the simulated $PM_{2.5}$ concentration

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in BTH and Beijing averaged from 9 to 25 January 2014. The simulated $PM_{2.5}$ mass concentration averaged during the simulation period is 111.6 and 97.7 μg m^{-3} in BTH and Beijing, respectively. OA is the dominant constituent of the simulated $PM_{2.5}$, consisting of around 43% of the $PM_{2.5}$ mass in BTH. Although the simulated O_3 concentration is low, the secondary aerosols, including SOA, sulfate, nitrate, and ammonium still make up about 50% of the $PM_{2.5}$ mass, in which the contribution of sulfate, nitrate, and ammonium is 11.3%, 12.4%, 9.6%, and 7.5%, respectively. Elemental carbon and the unspecified aerosol species account for 7.5% and 16.2% of the $PM_{2.5}$ mass, respectively. In Beijing, OA, sulfate, nitrate, and ammonium constitutes 44.1%, 10.6%, 14.0%, and 9.1% of the $PM_{2.5}$ mass, respectively, during the simulation period, which is close to the source apportionment results obtained by Huang et al. (2014) during the wintertime of 2013. It is worth noting that the simulated sulfate contribution to the $PM_{2.5}$ mass in Beijing is lower than the source apportionment result in Huang et al. (2014), and vice versa for the nitrate aerosol. Implementation of the APPCAP since 2013 September has considerably decreased SO_2 emissions in BTH, lowering the sulfate formation. Additionally, the decrease of the sulfate aerosol reduces its competition for ammonia in the atmosphere, facilitating the nitrate formation.

The good agreements of the simulated mass concentrations of air pollutants with observations at ambient monitoring sites in BTH and aerosol species with HR-ToF-AMS measurements in Beijing show that the simulated wind fields and emission inventory used in present study are generally reasonable, providing a reliable base for the further evaluation.

3.2 Contributions of the RCC emission to the air quality in BTH

The contribution of the residential coal combustion (RCC) emission to the air quality in BTH is investigated by the sensitivity study without RCC emissions in BTH and its surrounding areas compared to the reference simulation (hereafter we define the sensitivity simulation as SEN-BTH). Figure 6 shows the spatial distribution of the average contribution

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of the RCC emission in BTH and its surrounding areas to the $PM_{2.5}$ concentrations during the simulation period (defined as (REF − SEN-BTH)). The RCC emission plays an important role in the $PM_{2.5}$ level in the plain area of BTH, with a contribution varying from 30 to 70 μg m^{-3}. Over the mountain areas of BTH, the contribution of the RCC emission to the $PM_{2.5}$ concentration is generally less than 10 μg m^{-3}.

Table 2 presents the average change of air pollutants mass concentrations during the simulation period in BTH and Beijing. The average $PM_{2.5}$ concentration is 111.6 μg m^{-3} in BTH in the REF case and decreased to be 85.8 μg m^{-3} in the SEN-BTH case when the RCC emission is excluded. The RCC emission contributes about 23.1% of $PM_{2.5}$ mass in BTH on average. In addition, the RCC emission is an important SO_2 and CO source, contributing about 35.8% of SO_2 and 22.5% of CO. The RCC emission does not substantially influence the NO_2 level in BTH, with a contribution of 4.2%. When the RCC emission is not considered in the SEN-BTH case, the O_3 concentration is slightly increased due to decrease of the NO_2 concentration. The $PM_{2.5}$ concentration is decreased by around 30% in Beijing on average when the RCC emission in BTH is excluded, showing that the air quality in Beijing would be remarkably improved if the residential coal in BTH and its surrounding areas could be replaced by other clean energy sources, such as natural gas or electricity. Furthermore, the RCC emission in BTH and its surrounding areas contributes about 42.6% of SO_2 and 26.5% of CO in Beijing.

Figure 7 shows the average chemical composition of $PM_{2.5}$ contributed by the RCC emission in BTH and Beijing during the simulation period. The RCC emission contributes about 25.8 μg m^{-3} $PM_{2.5}$ in BTH on average, of which about 42.8% is from OA. The sulfate aerosol constitutes 17.1% of $PM_{2.5}$ contributed by the RCC emission, exceeding the contribution from unidentified aerosol species (15.8%), black carbon (11.5%), ammonium (9.5%) and nitrate (3.3%) aerosol. The results indicate that the priority to mitigate effects of the RCC emission on the air quality in BTH is to decrease the emissions of OA and SO_2 from

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263 RCC. In Beijing, OA is still the major contributor to $PM_{2.5}$ from the RCC emission, accounting
264 for about 48.5% which is more than that averaged in BTH. The sulfate and ammonium
265 contribution to $PM_{2.5}$ from the RCC emission is 13.3% and 7.2%, respectively, less than those
266 averaged in BTH. The chemical composition of $PM_{2.5}$ from the RCC emission in Beijing shows
267 more contribution of OA and less contribution of SO_2 from Beijing local RCC emission.

268 **3.4 Contributions of RCC emissions to the air quality in Beijing**

269 As the capital of China, the air quality in Beijing often becomes the focus of attention in
270 China or globally. Beijing is situated at the northern tip of the North China Plain (NCP), one
271 of the most polluted areas in China, caused by rapid industrialization and urbanization (Zhang
272 et al., 2013). In addition, Beijing is surrounded from the southwest to the northeast by the
273 Taihang Mountains and the Yanshan Mountains which block the dispersion of air pollutants
274 when the south or east winds are prevalent in NCP (Long et al., 2016). Therefore, except the
275 contribution of local emissions, the air quality in Beijing is also substantially influenced by the
276 transport of air pollutants from outside (Wu et al., 2017).

277 Since implementation of the APPCAP issued in September 2013, Beijing has carried out
278 aggressive emission control strategies to improve the air quality. Great efforts have been made
279 to replace the coal used in residential living by natural gas or electricity, which is highly
280 anticipated to clean the air in Beijing. However, frequent occurrence of heavy haze with
281 extremely high levels of $PM_{2.5}$ during the wintertime of 2015 and 2016 has caused controversial
282 issue about the effect of the coal replacement plan in Beijing. Therefore, a further sensitivity
283 study has been performed, in which only the RCC emission in Beijing is excluded (hereafter
284 we define the sensitivity simulation as SEN-PEK) to explore the contribution of the local RCC
285 emission in Beijing to the haze formation. Comparisons of the REF case with the SEN-PEK
286 case show that when the RCC emission is not considered or the residential coal is replaced by
287 other clean energy sources in Beijing, the Beijing's $PM_{2.5}$ level decreases from 97.7 to 80.1 μg

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m^{-3} or by 18.0% on average during the simulation period. The average decrease of SO_2 and CO concentrations is 24.2% and 19.9%, respectively. Therefore, the coal replacement plan in Beijing can improve the air quality in Beijing considerably, but is not as expected to bring back the blue sky to Beijing.

It is still disputatious on whether local emissions or transport from outside dominates the air quality in Beijing (Guo et al., 2010, 2014; Li et al., 2015; Zhang et al., 2015; Wu et al., 2017). Sensitivity studies show that when only the RCC emission in Beijing is excluded in simulations, the $PM_{2.5}$ level is decreased by 18%, much less than about 30% decrease caused by the exclusion of the RCC emission in BTH and its surrounding areas, showing the important contribution of trans-boundary transport to the air quality in Beijing. Analyses are further made to examine the contribution of the RCC emission in Beijing to the $PM_{2.5}$ concentrations under different pollution levels. The simulated hourly near-surface $PM_{2.5}$ concentrations in REF case during the whole episode in Beijing are first subdivided into 6 bins according to the air quality standard in China for $PM_{2.5}$ (Feng et al., 2016), i.e., 0~35 (excellent), 35~75 (good), 75~115 (lightly polluted), 115~150 (moderately polluted), 150~250 (heavily polluted), and greater than 250 (severely polluted) μg m^{-3}. $PM_{2.5}$ concentrations in REF case and SEN-PEK case as the bin $PM_{2.5}$ concentrations in REF case following the grid cells are assembled respectively, and an average of $PM_{2.5}$ concentrations in each bin is calculated. Figures 8 presents the contribution of the RCC emission in Beijing to the local $PM_{2.5}$ concentrations. Apparently, the mitigation effect is the best under the good and lightly polluted conditions, and the $PM_{2.5}$ concentration decreases by around 25% when the RCC emission in Beijing is not considered, indicating that the good and light $PM_{2.5}$ pollution is mainly caused by local emissions. However, with deterioration of the haze pollution from moderately to severely polluted conditions, the $PM_{2.5}$ contribution of the local RCC emission decreases from 20% to 15%, showing the regional transport of $PM_{2.5}$.

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313

314 **4 Summary and Conclusions**

315 In the present study, persistent air pollution episodes in BTH from 9 to 25 January 2014
316 are simulated using the WRF-CHEM model to evaluate the contributions of the RCC emission
317 to the air quality in BTH. In general, the WRF-CHEM model performs well in simulating the
318 temporal variations and spatial distributions of air pollutants compared to observations over
319 monitoring sites in BTH. The simulated diurnal variations of aerosol species are also in good
320 agreement with the HR-ToF-AMS measurements in Beijing.

321 Sensitivity studies show that, on average, the RCC emission contributes about 23.1% of
322 $PM_{2.5}$ mass concentrations in BTH during the simulation period and is also an important SO_2
323 and CO source, accounting for about 35.8% of SO_2 and 22.5% of CO. OA is the major
324 contributor to $PM_{2.5}$ from the RCC emission, with a contribution of 42.8%, followed by sulfate
325 (17.1%), unidentified aerosol species (15.8%), black carbon (11.5%), ammonium (9.5%) and
326 nitrate (3.3%) aerosol. Exclusion of the RCC emission in BTH and its surrounding areas
327 decreases the $PM_{2.5}$ concentration by around 30% in Beijing, indicating that the air quality in
328 Beijing will be remarkably improved if the residential coal in BTH and its surrounding areas
329 can be replaced by other clean energy sources.

330 When only the RCC emission in Beijing is excluded in simulations, the Beijing's $PM_{2.5}$
331 level decreases by 18.0% on average during the simulation period. Hence, the coal replacement
332 plan in Beijing is beneficial to the Beijing's air quality, but is not as anticipated to bring back
333 the blue sky to Beijing. The mitigation effect of the coal replacement plan on $PM_{2.5}$ in Beijing
334 is the best under the good and lightly polluted conditions, decreasing the $PM_{2.5}$ concentration
335 by around 25%. However, under the heavy or severe haze pollution, the local RCC emission
336 contributes about 15% of $PM_{2.5}$ in Beijing, showing contributions of the regional transport of
337 $PM_{2.5}$.

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This study mainly aims to quantitively evaluate the contributions of the RCC emission to the air quality in BTH. Our results suggest that if the residential coal replacement is only implemented in Beijing, the Beijing's air quality will not be improved substantially, considering the impact of trans-boundary transport. Implementation of the residential coal replacement in BTH and its surrounding areas can remarkably improve the Beijing's air quality. Although the WRF-CHEM model reasonably well captures the temporal and spatial variations of air pollutants in BTH and diurnal variations of aerosol species in Beijing, the model biases still exit. Future studies need to be conducted to improve the WRF-CHEM model simulations, considering the rapid changes in anthropogenic emissions since the implementation of the APPCAP. Further sensitivity simulations of various emission mitigation measures also need to be performed to provide efficient emission control strategies to improve the air quality in BTH.

5 Data availability

The real-time O_3 and $PM_{2.5}$ concentrations are accessible to the public on the website http://106.37.208.233:20035/ (China MEP, 2013a). One can also access the historic profiles of the observed ambient air pollutants by visiting http://www.aqistudy.cn/ (China MEP, 2013b).

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621 Table 1 WRF-CHEM model configurations.

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Region	**Beijing-Tianjin-Hebei (BTH)**
Simulation period	January 9 to 26, 2014
Domain size	150 × 150
Domain center	39°N, 117°E
Horizontal resolution	6km × 6km
Vertical resolution	35 vertical levels with a stretched vertical grid with spacing ranging from 30 m near the surface, to 500 m at 2.5 km and 1 km above 14 km
Microphysics scheme	WSM 6-class graupel scheme (Hong and Lim, 2006)
Boundary layer scheme	MYJ TKE scheme (Janjić, 2002)
Surface layer scheme	MYJ surface scheme (Janjić, 2002)
Land-surface scheme	Unified Noah land-surface model (Chen and Dudhia, 2001)
Longwave radiation scheme	Goddard longwave scheme (Chou and Suarez, 2001)
Shortwave radiation scheme	Goddard shortwave scheme (Chou and Suarez, 1999)
Meteorological boundary and initial conditions	NCEP 1°×1° reanalysis data
Chemical initial and boundary conditions	MOZART 6-hour output (Horowitz et al., 2003)
Anthropogenic emission inventory	Developed by Zhang et al. (2009) and Li et al. (2017)
Biogenic emission inventory	MEGAN model developed by Guenther et al. (2006)

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628 Table 2 Average mass concentrations of air pollutants in REF case and SEN-BTH case from 9
629 to 25 January 2014 in BTH and Beijing. (Unit: μg m^{-3} for $PM_{2.5}$, O_3, NO_2, SO_2 and mg m^{-3}
630 for CO)
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Air pollutants	BTH				Beijing			
	REF	SEN-BTH	Mass change	Percentage change	REF	SEN-BTH	Mass change	Percentage change
$PM_{2.5}$	111.6	85.8	25.8	23.1%	97.7	68.9	28.8	29.5%
O_3	39.1	39.4	-0.3	-0.8%	39.3	39.8	-0.5	-1.3%
NO_2	45.7	43.7	2.0	4.3%	51.5	49.4	2.1	4.1%
SO_2	45.0	28.9	16.1	35.8%	42.2	24.2	18.0	42.6%
CO	1.7	1.3	0.4	22.5%	1.5	1.1	0.4	26.5%

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Atmospheric Chemistry and Physics Discussions

638 **Figure Captions**

639

640 Figure 1 (a) Map showing the location of Beijing-Tianjin-Hebei and (b) WRF-CHEM model
641 simulation domain with topography. In (b), the filled red circles represent centers of
642 cities with ambient monitoring site and the size of the circle denotes the number of
643 ambient monitoring sites of cities. The filled black rectangle denotes the deployment
644 location of the HR-ToF-AMS in Beijing.

645 Figure 2 Pattern comparisons of simulated (color counters) vs. observed (colored circles)
646 near-surface mass concentrations of (a) $PM_{2.5}$, (b) O_3, (c) NO_2, and (d) SO_2
647 averaged from 9 to 25 January 2014. The black arrows indicate simulated surface
648 winds.

649 Figure 3 Comparisons of observed (black dots) and simulated (solid red lines) diurnal profiles
650 of near-surface hourly mass concentrations of (a) $PM_{2.5}$, (b) O_3, (c) NO_2, (d) SO_2,
651 and (d) CO averaged at monitoring sites in BTH from 9 to 25 January 2014.

652 Figure 4 Comparisons of measured (black dots) and simulated (solid red lines) diurnal
653 profiles of (a) organic aerosol (OA), (b) coal combustion organic aerosol (CCOA),
654 (c) sulfate, (d) nitrate, and (e) ammonium in Beijing from 9 to 25 January 2014.

655 Figure 5 Chemical composition of $PM_{2.5}$ averaged from 9 to 25 January 2014 in (a) BTH and
656 (b) Beijing.

657 Figure 6 Spatial distribution of the average contribution of the RCC emission in BTH and its
658 surrounding areas to $PM_{2.5}$ concentrations from 9 to 25 January 2014.

659 Figure 7 Chemical composition of $PM_{2.5}$ from the RCC emission averaged from 9 to 25
660 January 2014 in (a) BTH and (b) Beijing.

661 Figure 8 Average contributions of the RCC emission in Beijing to the Beijing's $PM_{2.5}$
662 concentration under different haze pollution levels from 9 to 25 January 2014.

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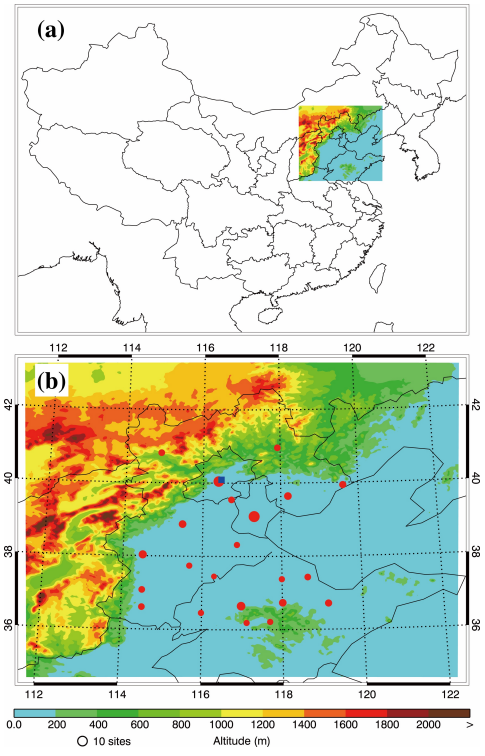

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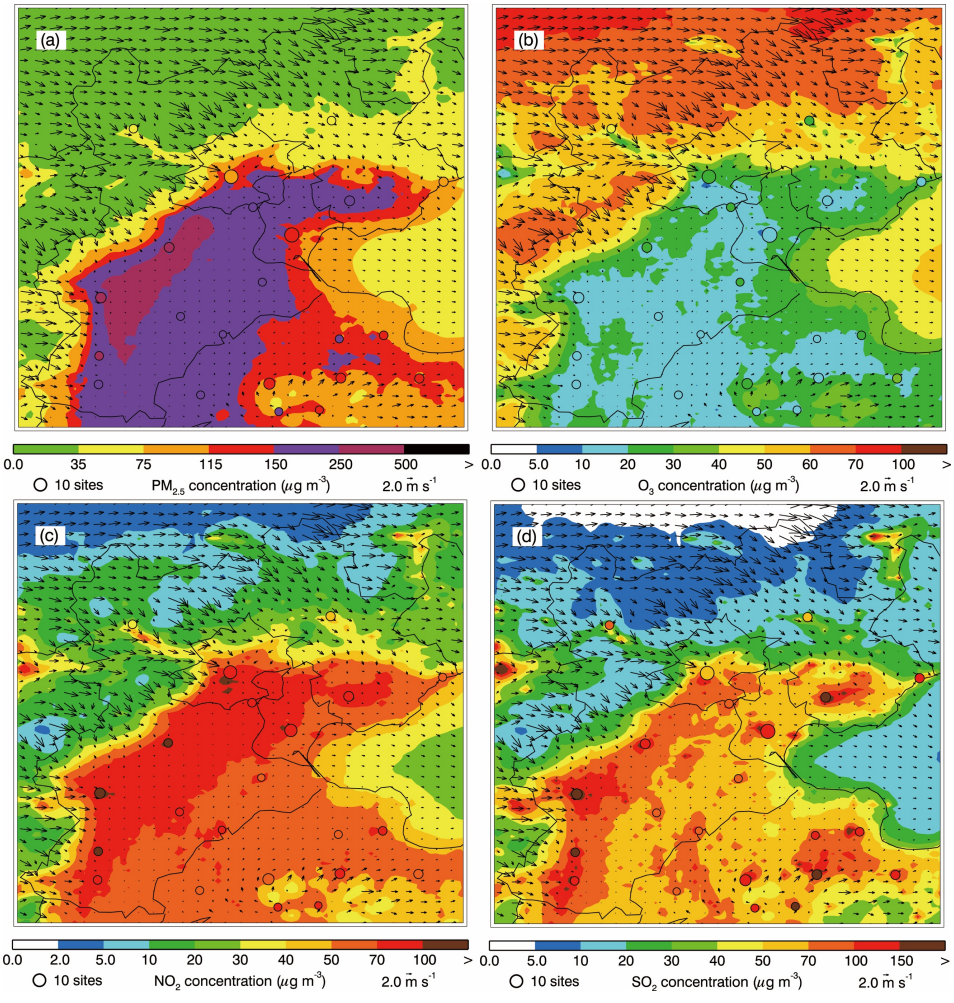

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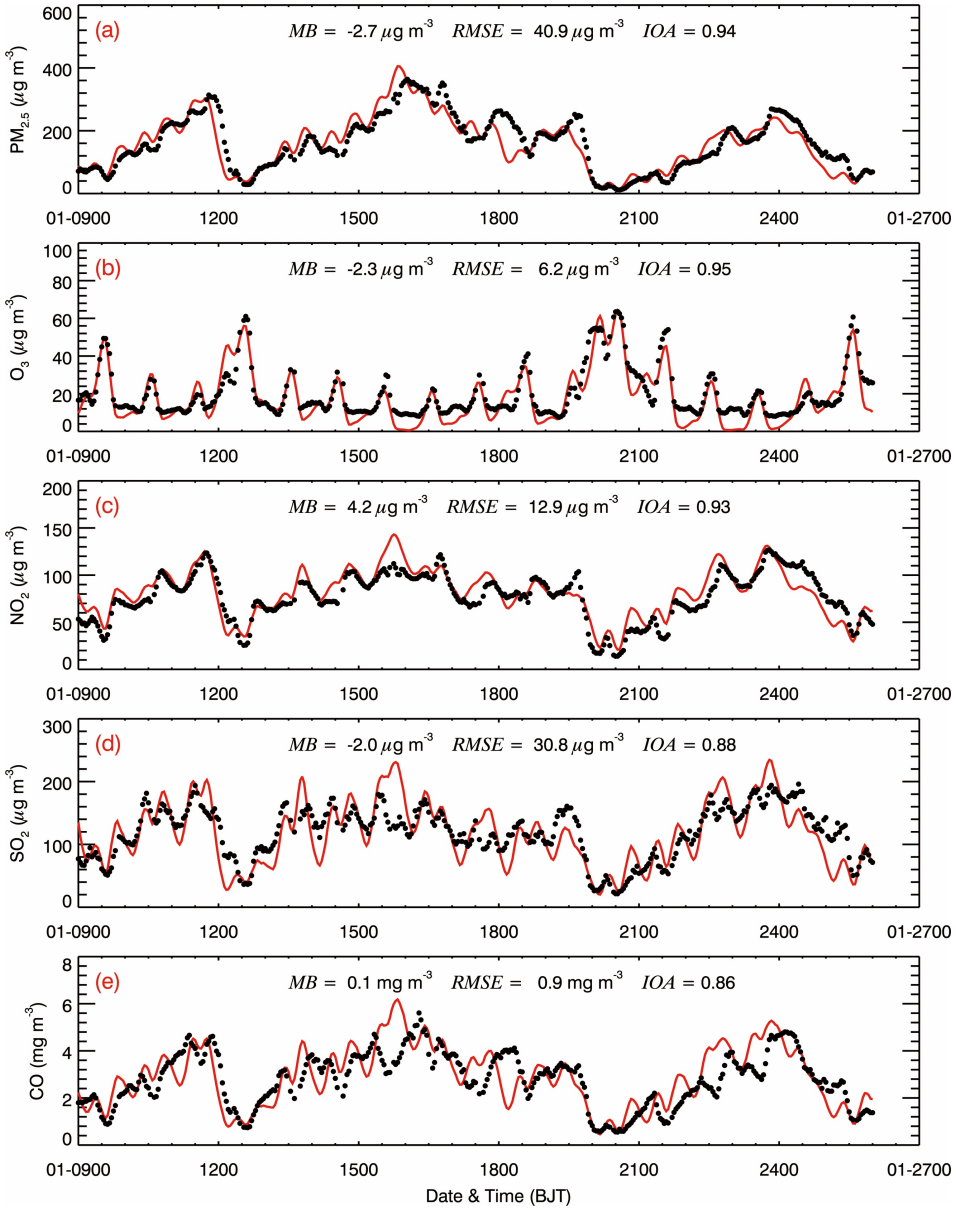

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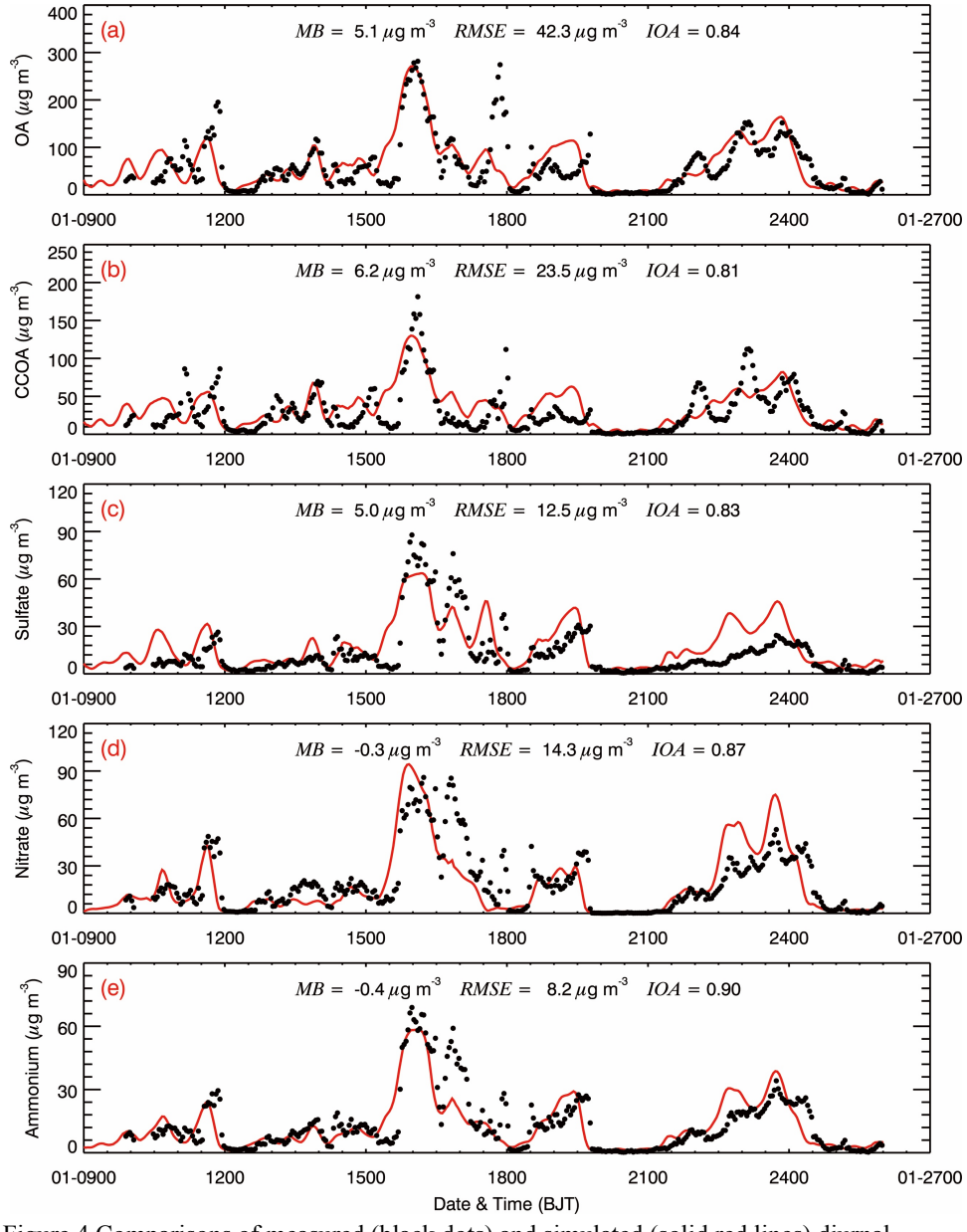

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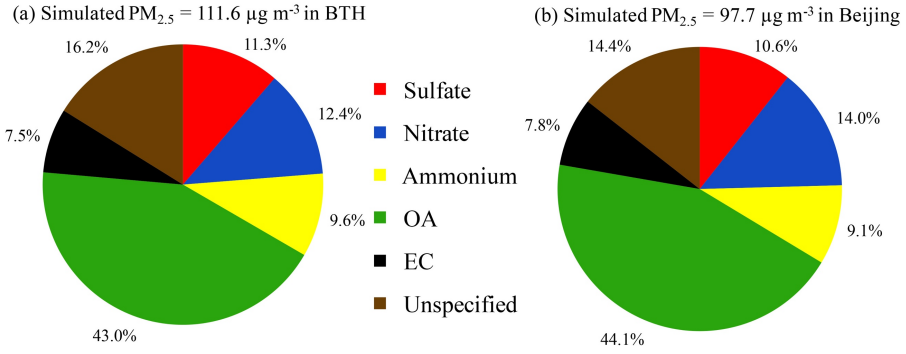

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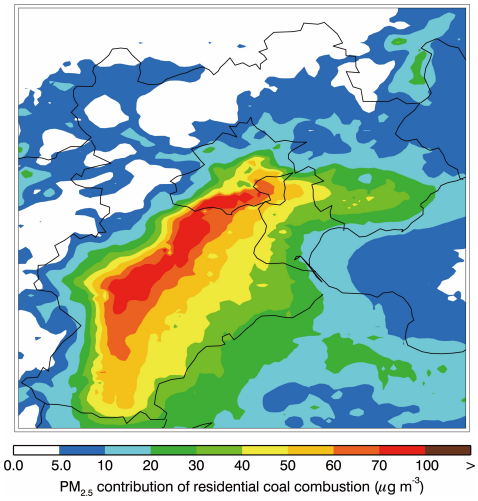

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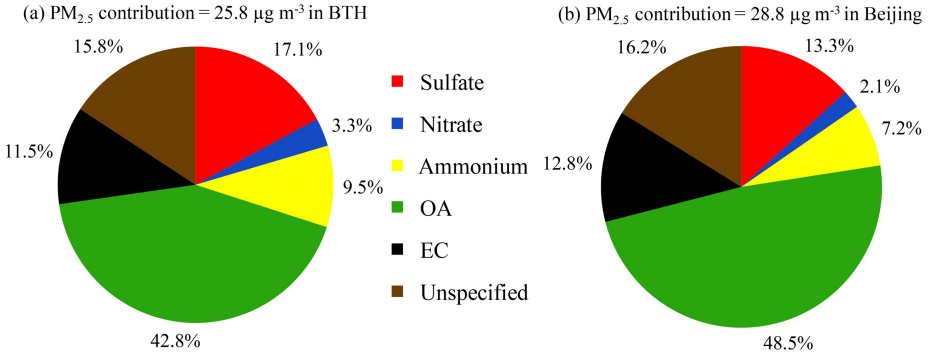

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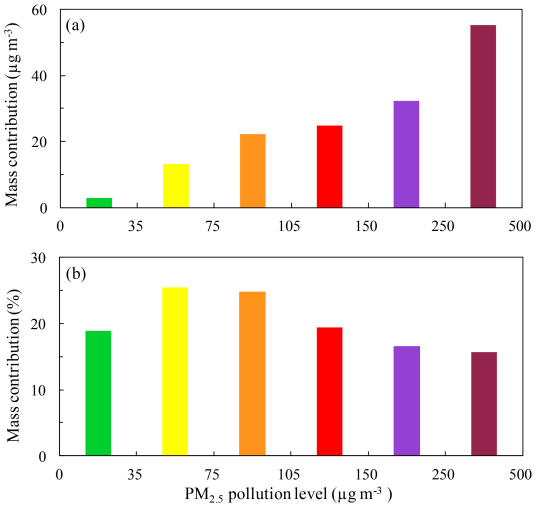

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