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# **Geophysical Research Letters**

## **RESEARCH LETTER**

10.1029/2018GL080941

## **Key Points:**

- BC weakens East Asian winter monsoon winds by changing cloud structure and land-sea thermal contrast
- The impact on monsoon winds per unit change in Arctic BC is similar to that of local BC emissions but has a larger uncertainty
- Reducing BC emissions could have significant indirect benefits for air quality control in the North China Plain

## **Supporting Information:**

Supporting Information S1

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### **Citation**:

Lou, S., Yang, Y., Wang, H., Smith, S. J., Qian, Y., & Rasch, P. J. (2019). Black carbon amplifies haze over the North China Plain by weakening the East Asian winter monsoon. Geophysical Research Letters, 46, 452-460, https:// doi.org/10.1029/2018GL080941

Received 15 OCT 2018 Accepted 8 DEC 2018 Accepted article online 17 DEC 2018 Published online 10 JAN 2019

## Black Carbon Amplifies Haze Over the North China Plain by Weakening the East Asian Winter Monsoon

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**Abstract** Black carbon (BC) has previously been found to intensify haze in China by stabilizing the planetary boundary layer. With ocean, sea ice, and cloud feedbacks included in a global aerosol-climate model, we show that BC emitted from the North China Plain can be transported to the oceans, which in turn changes cloud structure and land-sea thermal contrast. As a result, East Asian winter monsoon wind speeds decrease over the North China Plain. This decrease causes air stagnation that can further intensify haze. Our results suggest that in addition to the local BC-induced interactions between aerosol and the planetary boundary layer, BC can also amplify haze in the North China Plain by weakening East Asian winter monsoon through ocean, sea ice, and cloud feedbacks. It implies that reducing BC emissions could have significant indirect benefits for air quality in the North China Plain.

Plain Language Summary China has been experiencing severe winter air pollution in recent years. The pollution particles harm human health, affect atmospheric visibility, and cause adverse effects on economic and societal activities. The poor air quality was linked to high emissions from human activities and air stagnation associated with meteorological conditions and climate change. Recent studies reported that black carbon (BC), one of the by-products when wood or coal is burned, could intensify pollution in China by stabilizing the lower atmosphere (i.e., weakening vertical air mixing and dispersion of pollution plumes). In this study, we show that in addition to the impact on atmospheric mixing, BC can also amplify particle pollution in the North China Plain by weakening the East Asian winter monsoon through ocean, sea ice, and cloud feedbacks. This implies that reducing BC emissions could help mitigate air pollution in the North China Plain.

## 1. Introduction

China has been suffering severe and persistent winter haze in recent years (Ding & Liu, 2014; Huang et al., 2014; Zhang et al., 2015), characterized by high concentrations of fine particles with diameter less than  $2.5 \,\mu m$  (particulate matter  $2.5 \,[PM_{2.5}]$ ). In January 2013 and December 2015, China was hit by extreme haze events, with maximum daily PM<sub>2.5</sub> concentrations reaching 500  $\mu$ g/m<sup>3</sup>, much higher than the Grade II Air Quality National Standard of 75 µg/m<sup>3</sup> in China (Gao et al., 2017; Wang et al., 2014; Zhang, Gong, et al., 2017). High concentrations of  $PM_{25}$  particles harm human health by damaging respiratory and cardiovascular systems, causing morbidity and mortality (Fajersztajn et al., 2013; West et al., 2016; Zhang, Jiang, et al., 2017). They also affect atmospheric visibility and transportation, hence, leading to adverse effects on economic and societal activities (Zhang et al., 2014). In addition, these pollutants can reach distant regions through long-range transport, causing global air quality impacts (Guo et al., 2017; Uno et al., 2008; Yu et al., 2012; Yang, Wang, et al., 2017; Yang, Wang, Smith, Zhang, Lou, Yu, et al., 2018; Yang, Wang, Smith, Zhang, Lou, Qian, et al., 2018).

Haze occurs more in the winter season due to increased emission from coal consumption for heating, together with frequent air stagnation and weaker rainfall. In addition to high anthropogenic emissions (Smith et al., 2011) and fast particle formation (Cheng et al., 2016; Song et al., 2018; Wang et al., 2016), studies have reported that winter haze in China is strongly influenced by changes in meteorological conditions associated with synoptic systems and climate change (Cai et al., 2017). For instance, Yang et al. (2016) found that changes in meteorology dominated the interannual variability of wintertime  $PM_{2,5}$  in eastern China and changes in wind fields explained 37% of this variability. Wang et al. (2015) reported that 45-67% of the interannual to interdecadal variability of winter haze days in eastern China can be explained

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by Arctic sea ice variability, a finding confirmed by Zou et al. (2017). Q. Li, Zhang, et al. (2016) reported a statistically significant correlation coefficient of -0.41 between East Asian winter monsoon index and wintertime fog-haze days. Zhao et al. (2016) also found that the Pacific Decadal Oscillation can influence the number of winter haze days in central-eastern China, with a statistically significant correlation coefficient of 0.66 between the Pacific Decadal Oscillation index and wintertime haze days.

Recently, many studies started to consider aerosol-meteorology interactions as an important mechanism of haze occurrence (Z. Li, Guo, et al., 2017). One specific interaction is through influencing the dynamics of the planetary boundary layer (PBL) through changing radiative balance in the atmosphere, which in turn influences air pollutant dispersion and leads to extreme haze episodes (aerosol-PBL interaction).

Black carbon (BC) aerosol can influence meteorology and climate through directly absorbing solar radiation, affecting cloud formation and lifetime, and reducing surface albedo through deposition on snow and ice (Bond et al., 2013; Liao et al., 2015). Based on a regional atmospheric model (Weather Research and Forecasting model coupled to Chemistry [WRF-Chem]) simulation, Ding et al. (2016) found that through solar absorption in the upper PBL, BC reduces the amount of shortwave radiation reaching the surface and therefore decreases the sensible heat fluxes that drive the development of the PBL. Moreover, BC increases atmospheric stability through producing a temperature inversion due to solar absorption by BC (Z. Li, Guo, et al., 2017). These in turn suppress air pollutant dispersion and contribute to extreme haze episodes.

However, regional climate models do not consider responses of sea surface temperature and sea ice to BC perturbation and the associated cloud changes, which are potentially important to meteorological changes and pollutant accumulation in China (Yang, Russell, et al., 2017; Zou et al., 2017), which we examine in this work.

Meteorology in China is dominated by the East Asian monsoon (Chang et al., 2006). Some studies have examined relationships between BC and the East Asian winter monsoon (Jiang et al., 2017; Z. Li, Lau, et al., 2016; Mao et al., 2017). For instance, using a global chemical transport model, Mao et al. (2017) reported a negative correlation coefficient of -0.7 between the strength of the East Asian winter monsoon and surface BC concentrations in eastern China without considering aerosol feedback on meteorology. Jiang et al. (2017) found that BC can influence the East Asian winter monsoon through BC-induced Tibetan Plateau warming. Some studies investigated impacts of aerosols on South Asian monsoon (Bollasina et al., 2011; Lau & Kim, 2006). Bollasina et al. (2011) showed a -0.95 mm/day decrease in South Asian summer monsoon precipitation attributed to aerosol-induced energy imbalance. But few of these studies directly address air pollution in China.

In this study, using a fully coupled global aerosol-climate model that includes interactive ocean and sea ice components, we examine the response of meteorological variables associated with the East Asian winter monsoon to BC emitted from the Arctic and midlatitudes, respectively, and their potential impacts on winter haze in the North China Plain. This study helps to better understand the interactions between meteorology and aerosols in China, which can eventually be used to improve air quality prediction and assessment of potential changes in air quality and climate in response to air pollutant emission reductions.

## 2. Methods

To examine the impacts of BC on China haze via aerosol-meteorology interactions, simulations are performed using the coupled global aerosol-climate model CESM (Community Earth System Model; Hurrell et al., 2013), which is widely used to quantify aerosol-meteorology interactions (Yang et al., 2017). In CAM5 (Community Atmosphere Model version 5), the atmospheric component of CESM, BC is first emitted into the primary-carbon mode and then aged into the accumulation mode using the four-mode Modal Aerosol Module (MAM4). MAM4 is chosen because it has better representation of the BC aerosol life cycle than the three-mode MAM3, but it is more computationally efficient compared to the seven-mode MAM7, the other two aerosol module options in CAM5 (Liu et al., 2012). Aerosol modifications including aerosol aging, improvements in convective transport, and wet scavenging are included in the model (Wang et al., 2013). With the treatment of aging process for carbonaceous aerosols, MAM4 increases global BC burden and facilitates BC transport to remote regions (e.g., the Arctic), resulting from reduced wet scavenging of BC, compared to the MAM3 aerosol module without aerosol aging (Liu et al., 2016; Wang et al., 2013).





**Figure 1.** (a) Spatial distribution of December-January-February (DJF) mean near-surface black carbon (BC) concentrations (contour,  $\mu g/m^3$ ) and 850 hPa wind fields (vector, m/s) from the present-day simulation. The boxed region and black dot in panel a represent the North China Plain and Beijing, respectively. (b) Time series of DJF mean modeled near-surface BC concentrations (blue line,  $\mu g/m^3$ ) and 850 hPa wind speed (red line, m/s) in Beijing for the last 80 years of the present-day simulation. (c) Time series of DJF mean observed near-surface particulate matter 2.5 (PM<sub>2.5</sub>) concentrations (blue line,  $\mu g/m^3$ ) collected from the Mission China air quality monitoring program of the U.S. Department of State and 850 hPa wind speed (red line, m/s) from ERA-Interim reanalysis in Beijing over 2010–2017. Correlation coefficient (*R*) between wind speed and BC/PM<sub>2.5</sub> concentration is shown on the top right of panels b and c.

Anthropogenic emissions of carbonaceous aerosols BC and OC used here are updated versions of the emissions described in Bond et al. (2007) and Lamarque et al. (2010), which are reported in units of carbon mass, with this BC mass then tracked within the model. BC optical properties and chemical composition are assumed to be close to those for elemental carbon, which is also known as soot.

Aerosol-radiation and aerosol-cloud interactions, as well as BC albedo effects on snow and sea ice, are physically treated in the model. A comprehensive aerosol model characterization can be found in Liu et al. (2016). The model performance in simulating BC has been evaluated in previous studies (Liu et al., 2016; Yang, Wang, et al., 2017; Yang, Wang, Smith, Zhang, Lou, Yu, et al., 2018). Compared to CAWNET (China Meteorological Administration Atmosphere Watch Network; Zhang et al., 2012), the simulated December-January-February (DJF) near-surface BC concentration has a low bias of 5% in the North China Plain (Yang, Wang, et al., 2017), far smaller than emissions uncertainty (Bond et al., 2007). In a companion study using same CESM simulations (Yang, Smith, et al., 2018), we compared climate responses to anthropogenic BC emissions with those from previous studies (Baker et al., 2015; Sand et al., 2013, 2015; Stjern et al., 2017). Overall, CESM reproduced well both the spatial pattern and magnitude of global and regional climate responses.

Following Sand et al. (2013), three 100-year parallel simulations are conducted, including (1) a present-day (average of 2008-2012) BC emission simulation (PD); (2) the same simulation as PD except that anthropogenic BC emissions over the Arctic (60-90°N) are scaled by 150 times (ARC150X); (3) the same simulation as PD except that anthropogenic BC emissions over the midlatitudes (28-60°N) are scaled by 7 times (MID7X), which includes most of China; and (4) a preindustrial (1850) BC emission simulation (PI). The large scaling factors (150 times in the Arctic and 7 times in midlatitudes) are used so that climate response signals are stronger than internal variability in the climate model. These specific scaling factors follow Sand et al. (2013) and are chosen to give radiative forcing of BC within the Arctic of similar magnitude between ARC150X and MID7X. Using the same scaling factors also allows a direct comparison of climate responses in our results to Sand et al. (2013), as discussed in Yang, Smith, et al. (2018). Examining emission perturbations in multiple regions allows us to determine if the impacts of BC on China haze via aerosol-meteorology interactions may depend on the location of BC emissions.

Other emissions and conditions are set at their year 1850 level. The average of model results for the last 80 years is used for analysis. Anthropogenic aerosol emissions are from CMIP6 (Coupled Model

Intercomparison Project Phase 6) data set (version 2017-05-18; Hoesly et al., 2018). The spatial distribution of DJF anthropogenic BC emissions as used in this work is shown in Figure S1 in the supporting information. Eastern China, especially the North China Plain, has a high BC emission rate.

## 3. Results

The North China Plain features northwesterly winds in DJF associated with the East Asian winter monsoon (Figure 1a). Due to high emission rates, the North China Plain also experiences high surface BC concentrations. In Beijing, the major city in the North China Plain, modeled DJF mean near-surface BC concentration, and 850 hPa wind speed show a statistically significant negative correlation (-0.77) with all responses and feedbacks included from the 80-year PD simulation (Figure 1b). Observed DJF mean PM<sub>2.5</sub> concentrations





**Figure 2.** Changes in (top) high and (bottom) low cloud fraction (%) in December-January-February for (left) MID7X and (right) ARC150X relative to present-day. The dotted areas indicate statistical significance with 95% confidence from a two-tailed Student's *t* test.

in Beijing have a similar negative correlation (-0.61) with wind speed from reanalysis data between 2010 and 2017 (Figure 1c). The negative correlation is mainly because in weak wind conditions air stagnation leads to the accumulation of aerosols in the boundary layer and a high aerosol concentration near the surface. However, these correlations can also result from BC-induced changes in aerosol-meteorology interactions, such as a BC-induced increase in PBL stability and/or possible changes in the East Asian winter monsoon circulation.

Under the midlatitude BC emission perturbation in the MID7X experiment, more BC is transported to the western Pacific Ocean between 30 and 45°N (Figure S2). Through absorbing solar radiation in cloud layers, BC-induced heating evaporates cloud and affects cloud dynamical processes (Koch & Del Genio, 2010), which drives significant temperature change and variability in the atmosphere (Yang, Smith, et al., 2018). Figure 2 shows changes in DJF mean cloud fraction due to BC emission perturbations. BC in MID7X reduces the amount of low cloud over Bohai Sea, Yellow Sea, and East China Sea between 30 and 45°N and increases high cloud amount over these regions, as compared to PD.

The BC-induced increase in high cloud amount in ARC150X occurs at a higher latitude than in MID7X because BC emission is perturbed over the Arctic, and the decrease in low cloud is away from China mainland. The decrease in low cloud amount and increase in high cloud amount lead to an increase in sea surface temperature over oceans east of China mainland (Figure S3), which further warms atmosphere through sensible heat.

Figure 3 presents BC-induced changes in zonal and meridional air temperature and wind profiles in DJF for the two regional BC perturbations. The transported BC, mainly from the North China Plain (Yang, Wang, et al., 2017), causes strong warming between 30 and 45°N over 850–700 hPa east of the China mainland (Figure 3a). The air temperature increase over eastern China land areas is not as strong as that over the ocean. There are even decreases in near-surface temperature over the North China Plain, although these are not statistically significant, due to BC atmospheric solar absorption, which can further stabilize PBL and decrease PBL height (Ding et al., 2016). The increase in temperature over oceans and decrease over land result in an anomalous land-sea thermal contrast, which in turn drives an anomalous easterly wind component in the lower atmosphere that can weaken the westerly wind component of the East Asian winter monsoon. Since that Arctic BC emissions under ARC150X warm the atmosphere in high latitudes, they have a very small impact on zonal winds over the North China Plain (Figure 3b).



**Figure 3.** Changes in (a and b) zonal (averaged over  $30-45^{\circ}$ N) and (c and d) meridional (averaged over  $110-122.5^{\circ}$ E, bottom) temperature (K) and wind fields (m/s) in December-January-February for (left) MID7X and (right) ARC150X relative to present-day. Pressure velocity (Pa/s) is scaled by a factor of -100. The dotted areas for temperature indicate statistical significance with 95% confidence from a two-tailed Student's *t* test.

The Arctic Front, which features temperature inversions in the lower atmosphere of the Arctic, blocks the transport of aerosols between the Arctic and midlatitudes. In winter, the Arctic Front shifts southward, favoring exchanges of air mass between these two regions. The prescribed increase in Arctic BC emissions in ARC150X results in increased surface concentrations and a strong warming of the atmosphere below 850 hPa along 40–50°N averaged over 110–122.5°E (Figure 3d). This could be due to a combination of transport to the North China Plain at low altitudes within the Arctic Front (Figure S4) together with BC-induced local stagnation. Warming from midlatitude BC emissions under MID7X occurs from the surface to 500 hPa, at a higher altitude compared to ARC150X (Figure 3c). In a companion study (Yang, Smith, et al., 2018), both midlatitude and Arctic BC emissions are found to produce anomalous southerly winds in the lower atmosphere through changing large-scale circulations due to the BC-induced air stagnation in the midlatitude and Arctic sea ice melt. Following the adiabatic flow, the lower altitude warming in ARC150X is in favor of anomalous southerly wind component in the lower atmosphere, reducing northerly wind component of the East Asian winter monsoon in the North China Plain in winter, while the anomalous southerly wind component is blocked in the lower atmosphere by the extended warming up to 500 hPa in MID7X.

Figure 4 shows changes in 850 hPa winds due to BC emission perturbations. As discussed above, BC emissions from the midlatitudes and the Arctic result in anomalous North China Plain easterly and southerly winds, respectively. This decreases the zonal and meridional components of prevailing northwesterly



**Figure 4.** Changes in (top) 850 hPa wind fields (m/s) and (bottom) wind speed (m/s, bottom) in December-January-February for (left) MID7X and (right) ARC150X relative to present-day. The boxed region represents the North China Plain, and the region with black dot is Beijing. The dotted areas indicate statistical significance with 95% confidence from a two-tailed Student's *t* test.

winds of the East Asian winter monsoon, respectively, and in turn cause the accumulation of pollutants in the boundary layer. BC emissions from the midlatitudes lead to a decrease in wind speed over most of the North China Plain, while Arctic BC emissions mainly reduce wind speed over northeast part of the North China Plain.

To compare the efficiency of BC emissions in affecting wind speed, we scale BC-induced changes in wind speed by the mass of the annual emission change. Wind speeds decrease by 0.024 ( $\pm$ 0.007) m/s in Beijing per unit (1 Tg C) of midlatitude BC emissions, similar in magnitude to 0.025 ( $\pm$ 0.013) m/s decrease per unit Arctic BC emissions, indicating that increase in both Arctic and midlatitude BC emissions can weaken East Asian winter monsoon and therefore lead to air stagnation in Beijing. However, midlatitude BC emissions have increased more than the Arctic since the preindustrial era. Scaling BC emissions to present-day levels, these results imply that the midlatitude BC has caused a decrease in wind speed in Beijing of -0.072 ( $\pm$ 0.021) m/s, whereas the influence of present-day Arctic BC emission is negligible. The impact of midlatitude BC is very likely due to North China Plain local emissions, since these account for more than 70% and 50% of the winter BC column burden over the North China Plain and oceans east of China mainland, respectively (Yang, Wang, et al., 2017; Yang, Wang, Smith, Zhang, Lou, Qian, et al., 2018). The magnitude of this BC-induced decrease in wind speed is similar to the -0.06 ( $\pm$ 0.05) m/s decrease in wind speed due to dustwind interactions reported by Yang, Russell, et al. (2017), which was found to account for 13% of increasing PM<sub>2.5</sub> concentrations over eastern China in polluted years (dust-wind interactions), revealing the importance of BC-induced weakening of East Asian winter monsoon in China haze formation.

We also tested linearity of the wind response to emissions perturbation size by using additional simulations in which anthropogenic BC emissions are scaled by 3.5 (MID3.5X) and 14 (MID14X) times, respectively, over the midlatitudes and 75 (ARC75X) times over the Arctic (Figure S5). Anomalous easterly winds are stronger when midlatitude BC emissions are scaled by a larger factor. The per-unit impacts on wind speed in Beijing of BC emissions are similar between the three midlatitude perturbations (-0.031 m/s for MID3.5X, -0.024 m/s for MID7X, and -0.029 m/s for MID14X per unit BC emission). The impacts on wind speed in Beijing per unit BC emissions in ARC75X is -0.015 ( $\pm 0.023$ ) m/s, which has much a higher relative

uncertainty than ARC150X ( $-0.025 [\pm 0.013]$  m/s), indicating that there is a larger uncertainty in the simulated impact of Arctic BC emissions on pollution conditions over the North China Plain.

## 4. Conclusions

This study examines responses of meteorological variables to strong BC emission perturbations and their potential impacts on winter haze in the North China Plain based on 100-year simulations using a fully coupled global aerosol-climate model with ocean, sea ice, and associated cloud feedbacks. Midlatitude BC emissions reduce low clouds and increase high cloud amount over oceans east of China, leading to increases in sea surface temperature and BC-induced warming of the atmosphere. This produces an anomalous land-sea thermal contrast and a decrease in zonal component of prevailing northwesterly winds of East Asian winter monsoon. When Arctic BC emission is scaled up by a large factor, considering BC-induced anomalous southerly winds due to Arctic sea ice melt, the meridional wind component of East Asian winter monsoon is reduced along with BC-induced lower-atmosphere warming, although there is a larger uncertainty in the impact of Arctic BC emissions.

The decrease in wind speed from BC-induced weakening of the East Asian winter monsoon can further intensify winter haze in the North China Plain, which partly explains the negative correlation between BC concentration and wind speed. We find that present-day BC emissions in the PD simulation also lead to anomalous easterly winds (at 850 hPa) over the North China Plain (Figure S6), which are similar to the pattern in the larger MID7X perturbation (Figure 4), supporting the role of present-day midlatitude BC emissions as compared to preindustrial conditions. Note that changes in winds outside China do not show the same pattern in PD compared to MID7X case, in part because emissions were only perturbed over the midlatitudes in MID7X.

This study extends the finding of BC-induced China haze via aerosol-PBL interactions by Ding et al., 2016). In addition to aerosol-PBL interactions, we find that BC can also influence China haze through weakening the East Asian winter monsoon. Solely considering aerosol-PBL interactions may underestimate the impact of BC on China haze. East Asian winter monsoon wind speeds have weakened between 1998 and 2012 (Yang, Wang, Smith, Zhang, Lou, Qian, et al., 2018), which intensified haze in the North China Plain during that time, but the underlying mechanisms of this weakening are still unclear. The present study suggests that the increase in BC emissions in China during that time could be one possible contributor.

Our results also show that both midlatitude and Arctic BC emissions impact Beijing winter wind speed by a similar per-unit amount. Although the magnitude of Arctic BC emissions is relatively small compared to those from midlatitudes, increasing human activities within the Arctic can potentially exacerbate both global climate change and regional air quality.

Due to air quality regulations,  $SO_2$  emissions, the precursor of sulfate aerosol, have been reduced significantly in China (C. Li, McLinden, et al., 2017). BC emissions have also decreased but not as much as  $SO_2$  (Zheng et al., 2018). Additional BC reductions in China and some foreign regions would directly and indirectly improve air quality. BC also exerts a warming influence on climate, particularly in Arctic regions. Our study indicates that BC emission reductions would help mitigate both regional air pollution and climate change.

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## Acknowledgments

This research is based on work that was supported by the U.S. Department of Energy (DOE) Office of Science, Biological and Environmental Research as part of the Regional and Global Climate Modeling program and by the U.S. Environmental Protection Agency. The Pacific Northwest National Laboratory is operated for DOE by Battelle Memorial Institute under contract DE-AC05-76RLO1830. Observed PM<sub>2.5</sub> concentrations in Beijing are collected from the Mission China air quality monitoring program of the U.S. Department of State (www. stateair.net/web/post/1/1.html). ERA-Interim reanalysis can be obtained from http://apps.ecmwf.int/datasets/data/ interim-full-daily/levtype=sfc/. The National Energy Research Scientific Computing Center (NERSC) provided computational support. Model results are available through NERSC at http:// portal.nersc.gov/project/m1199/yangyang/Arctic\_BC/.

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