



Air quality and climate responses to anthropogenic black carbon emission changes from East Asia, North America and Europe



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HIGHLIGHTS

- We model the short-term climatic effects of regional black carbon (BC) emissions.
- Emissions would linearly impact the BC burden over source and nearby downwind areas.
- Some climatic factors preserve a robust linear relationship only in source areas.
- The perturbation of atmospheric circulation complicates much of BC climatic forcing.

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ABSTRACT

East Asia, North America and Europe are the world largest emitters of anthropogenic black carbon (BC). In this study, the role of each region's anthropogenic BC emissions on domestic air quality and climate is investigated. A ten-year six-member parallel simulation (i.e., with anthropogenic emissions in each region reduced by 0%, 50% or 100%, or increased by 200%, 500% or 1000%) is conducted based on the state-of-the-art Community Earth System Model (CESM). Linearity of the emission-response relationship is examined for a variety of air quality and climate indicators. Generally, a change in BC emissions tend to linearly influence BC concentrations over both source and nearby downwind regions even taking into account the effect of BC-induced climate perturbations. Aerosol optical depth (AOD) and the net radiative flux perturbation at top of atmosphere (TOA) tend to preserve a similar linear relationship to local BC emission changes, with a robust signal confined only to the source areas. However, the response of temperature in most places is inconsistent to BC emission changes. Though the presence of BC in the atmosphere absorbs solar and terrestrial radiation which has a tendency to warm the atmosphere, the perturbed atmospheric circulation induces substantial meridional exchanges of warm and cold air masses, which overpasses the warming tendency of BC exerted on the atmosphere. This indicates that reducing/increasing regional BC emissions immediately ameliorate/deteriorate local air quality proportionally, but the associated effects on climate perturbation may lack a clear trend within the initial 10-year time span.

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1. Introduction

Black carbon (BC) aerosol, emitted from a variety of combustion processes, affects the Earth's climate system directly through absorbing and scattering the solar radiation, and indirectly through altering the cloud processes as well as the melting of snow and ice

due to its unique physical properties (Bond et al., 2013). Though the freshly emitted BC (hydrophobic BC) is insoluble in water or common organic solvents, its aging processes in which water-soluble substances (e.g., sulfuric acid) accumulate on its surface would enhance its cloud condensation nuclei (CCN) activity. Aging (Onischuk et al., 2003; Oshima et al., 2009) complicates much of the issues of the lifetime and removal rate of BC.

The direct radiative forcing (DRF) is the most commonly studied climate forcing terms for BC (Chung and Seinfeld, 2005; Hansen et al., 2005; Sokolov, 2006), and the BC DRF from all present-day sources was estimated to be $+0.88 \text{ W m}^{-2}$ with 90% uncertainty

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bounds of +0.17 to +1.48 W m⁻². In addition, it was found that BC contributed to the warming of Arctic regions (Sand et al., 2013; Shindell and Faluvegi, 2009). The semi-direct effect refers to the alteration of cloud distribution by DRF of BC. The RF of BC induces adjustments of different time scales within the climate system. Chung et al. (2002) found that radiation absorbing aerosol (e.g. BC) could warm the atmosphere while cool the land surface below, stabilize the boundary layer, reduce the evaporation and sensible heat flux from the land, and furthermore impact the monsoon dynamics in South Asia. Menon et al. (2002) and Ramanathan et al. (2005) found that regional climate trends of temperature fields and hydrological cycle could be attributed to lifting BC atmospheric burdens in Asia. The large uncertainty in modeled BC RF could be attributed to the diversity in estimating BC's atmospheric burden (e.g., emission, lifetime) and optical properties (e.g. mass absorption cross section and forcing efficiency) (Bond et al., 2013). Besides, the radiative forcing of BC depends highly on the altitude location (Ban-Weiss et al., 2012), vertical profile (Samset et al., 2013), and mixing state with co-emitted species (Cappa et al., 2012; Chung and Seinfeld, 2002; Haywood and Shine, 1995).

The cloud and ice/snow effects of BC even remain more uncertain (Denman et al., 2007; Flanner et al., 2007; Heintzenberg and Charlson, 2009). Nonetheless, the total climate forcing integrating all forcing terms was estimated to be +1.1 W m⁻² with 90% uncertainty bounds of +0.17 to +2.1 W m⁻². BC is potentially the second most important climate warming agent only inferior to carbon dioxide (Bond et al., 2013), whose RF was estimated to be 1.66 W m⁻² (Forster et al., 2007). The predominant sources for BC include fossil fuels (38%), solid fuels (20%) and open burning (42%), and are largely contributed by anthropogenic combustion (Bond et al., 2004). This motivates the establishment of sound emission metrics for BC in mitigation of its adverse climate impacts. However, to our knowledge, little literature has investigated how BC emission in a certain region changes along the gradient (i.e., BC emissions are changed by -100%, -50%, 0%, 200% and 1000%, respectively) would impact the global climate.

In this study, we utilized a fully-coupled earth system model CESM to evaluate the global short-term climate response due to anthropogenic BC emitted from East Asia, North America and Europe, which are considered to be the biggest contributors in global anthropogenic BC emissions. Rather than identifying the equilibrium climate response from a BC emission perturbation (which may take more than century's period of simulation), this study focuses on the "closest to real" short-term effect (Jacobson, 2010) of BC on our environment. Specifically, we'd like to understand whether the disturbance of air quality and regional climate in different areas response linearly or nonlinearly to the fluctuation of BC emissions in a particular region. Only direct and semi-direct effects of BC on radiation are involved in this configuration. Therefore, the result reflects a short-term climate perturbation in a particular configuration of CESM. From there, we evaluate the extent that a robust emission-response relationship (i.e., air quality and climate perturbations) exists for BC from different regions. This may acknowledge policymakers the potential outcomes if BC emissions over the region are well controlled or uncontrolled.

The configuration of CESM is adapted in this study to allow chemistry processes coupled into general circulation model. That atmospheric component of CESM (i.e., CAM-Chem) utilized in this experiment has a comprehensive treatment of aerosol processes and its interaction with climate. We investigate various indicators related to climate perturbation, including black carbon burden, aerosol optical depth, radiation budget, cloud cover, the surface air temperature (SAT), surface pressure and wind fields, etc. We describe our experiment design and model configuration in Section 2, and show the air quality relevant results in Section 3 and climate

relevant findings in Section 4. Finally, conclusions are drawn in Section 5.

2. Methods

2.1. Model description

The Community Earth System Model (CESM) version 1.1.2 (released in July 2013) was used in this study. CESM is a fully coupled climate model with components of an atmospheric model of Community Atmospheric Model Version 4 (CAM4), a land model of Community Land Model Version 4 (CLM4), an ocean model Parallel Ocean Program Version 2 (POP2), models of sea ice, land ice and river, and a high-performance coupler. The 1.1z release of CESM has been scientifically validated, and the outputs of multi-decadal model runs have been evaluated (<http://www2.cesm.ucar.edu/models/scientifically-supported>). CAM-Chem (CAM4 with implementation of chemistry) has its chemistry process expanded but mostly equivalent to MOZART-4 (Lamarque et al., 2012). Though, interactions between climate and chemistry processes are only through radiation, CAM4 does not include cloud–aerosol interactions (i.e., aerosols indirect effect). CAM4 was released as part of CESM and proved scientifically valid through simulations and comparisons with observation data (Collins et al., 2004; Neale et al., 2010). The finite volume dynamic core is the default in CAM4 due to its superior tracer transport properties. Deep convection is parameterized using the Zhang and McFarlane (1995) scheme. Other parameterization methods on clouds and precipitation processes can be found in Boville et al. (2006). Processes in the planetary boundary layer are parameterized following Holtlag and Boville (1993).

Dry deposition in CAM-Chem is calculated following the resistance approach (Wesely, 1989). The computation of canopy resistance in CAM-Chem takes advantage of its coupling to Community Land Model. The BULK scheme in CAM-Chem uses fixed dry deposition velocities of black and organic aerosols as 0.1 cm s⁻¹ over all surfaces (Lamarque et al., 2012). Wet removal of soluble gas-phase species is the combination of two processes: in-cloud, or nucleation scavenging (rain-out) and below-cloud, or impaction scavenging (washout). Removal can be seen as a simple first-order loss process.

There are three aerosol treatment methods in this model, BAM (Bulk Aerosol Model), MAM3 (3 Mode Aerosol Model) and MAM7 (7 Mode Aerosol Model). In this study, the aerosol treatment scheme we utilized is based on the Bulk Aerosol Model (BAM) and assumes the lognormal distribution of black carbon aerosols follows a mean radius of 11.8 nm, geometric standard deviation of 2.00 and density of 1.0 g cm⁻³. We didn't use MAM (both MAM3 and MAM7 include various indirect effects of aerosols) since this paper mainly focuses on the direct and semi-direct effects of black carbon on the climate, which relies less on aerosol size distributions and thus is associated less to the uncertainties in modeling the indirect effects. Black carbon in the BULK aerosol scheme is simulated as two separate tracers: one is hydrophobic (CB1) which represents black carbon particles have insufficient coating of soluble materials and thus cannot be treated as cloud condensation nuclei (CCN), and the other is hydrophilic (CB2) which represents internally mixed BC which can activate cloud droplets. The conversion of carbonaceous aerosols from hydrophobic to hydrophilic is assumed to occur as a fixed aging timescale of 1.6 days (Tie et al., 2001).

2.2. Model configuration

The model is set up with a grid resolution of 1.9 × 2.5°

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