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Interprovincial Reliance for Improving Air Quality in China: A Case Study on Black Carbon Aerosol

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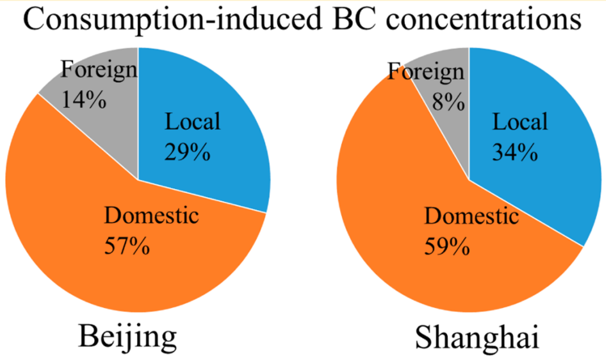
Supporting Information

ABSTRACT: Black carbon (BC) is of global concern because of its adverse effects on climate and human health. It can travel long distances via atmospheric movement and can be geographically relocated through trade. Here, we explored the integrated patterns of BC transport within 30 provinces in China from the perspective of meteorology and interprovincial trade using the Weather Research and Forecasting with Chemistry (WRF/Chem) model and multiregion input-output analysis. In general, cross-border BC transport, which accounts for more than 30% of the surface concentration, occurs mainly between neighboring provinces. Specifically, Hebei contributes 1.2 $\mu g{\cdot}m^{-3}$ BC concentration in Tianjin. By contrast, trade typically drives virtual BC flows from developed provinces to heavily industrial provinces, with the largest net flow from Beijing to Hebei (4.2 Gg). Shanghai is most vulnerable to domestic consumption with an average interprovincial consumption influence efficiency of 1.5×10^{-4} ($\mu g{\cdot}m^{-3}$)/(billion $Yuan{\cdot}yr^{-1}$). High efficiencies ($\sim 8 \times 10^{-5}$ ($\mu g{\cdot}m^{-3}$)/(billion $Yuan{\cdot}yr^{-1}$)) are also found from regions including Beijing, Jiangsu, and Shanghai to regions including Hebei, Shandong, and Henan. The above source−receptor relationship indicates two control zones: Huabei and Huadong. Both mitigating end-of-pipe emissions and rationalizing the demand for pollution-intense products are important within the two control zones to reduce BC and other pollutants.



INTRODUCTION

Black carbon (BC), which is generated by the incomplete combustion of carbonaceous fuels,[1,2] is an important combustion component of fine particulate matter ($PM_{2.5}$).[3,4] Moreover, the scientific community has been increasingly concerned about its adverse impact on climate change, air quality, and human health.[5−7] BC aerosols influence climate both regionally and globally by absorbing solar radiation, which reduces the atmospheric lapse rate and burns off cloud droplets.[3,7] BC level varies consistently with carbon monoxide (CO), nitric oxide (NO), and other traffic-related gaseous pollutants and occupies roughly a fixed proportion of particulate matter (PM) concentration in summer and autumn.[8,9] Additionally, pollution containing BC has been proven to have a robust epidemiological association with many types of mortality, particularly cardiovascular.[10,11] Thus, it is acknowledged that BC may serve as an effective indicator of air quality and its health effects in helping to mitigate air pollution including PM, CO and NO.[4,11,12] Once emitted into the atmosphere, BC has a lifetime of 2−10 days and can be transported long distances by atmospheric movement,[13−16] indicating its well-mixed condition in lower troposphere and regional, rather than local, character.[17]

China has been the world's largest emitter of anthropogenic BC, organic matter (OM) and other $PM_{2.5}$ precursors.[18−20] In 2014, approximately 90% of the major cities in China failed to meet the national air quality standard for $PM_{2.5}$.[21] Emissions from the industrial and transport sectors have been identified as the major sources of BC and other combustion $PM_{2.5}$,[22] which has led to the need for serious emissions control in China. Recently, the "Law of the People's Republic of China on the Prevention and Control of Atmospheric Pollution" has been revised to emphasize the national target of air quality improvement from a concentration-based perspective and the supervision of pollution sources using emissions-based strategies.[23] This law also calls for collaborative efforts across administrative boundaries for emissions control and air quality improvement. Consequently, a quantitative understanding of the interprovincial source−receptor relationship of air pollution transport and the underlying economic drivers is of great importance.[24,25]

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Previous studies have evaluated the possible sources of air pollution for a target region.[13,26] For example, Guo et al. analyzed the observational data collected at Changdao Island north of Shanghai and found that a significant amount of BC is transported from Shandong and Jiangsu provinces.[27] Xue et al. utilized particulate source apportionment technology (PSAT) in the CAMx model and found that approximately 40% of ambient $PM_{2.5}$ concentrations in Beijing, Shanghai and Jiangsu are contributed by cross-boundary transport.[28] If the source−receptor relationship were accepted by the relevant provincial governments, it might be the basis for promoting multiprovince cooperation on emissions control.[29]

Apart from observed atmospheric transport, domestic trade also affects the distribution of emissions to a large extent and thus changes air pollution levels geographically.[30,31] The production of traded products increases local emissions while reducing the emissions in consuming regions. This trade-induced virtual emissions transfer among provinces in China has been well documented for carbon dioxide (CO_2), sulfur dioxide (SO_2), primary $PM_{2.5}$ and other atmospheric pollutants, which demonstrates that some developed provinces shift emissions to less-developed provinces by importing products.[25,30] As with atmospheric transport, this virtual transfer of emissions and the degradation of air quality via trade also lead to a source−receptor relationship. When considering the location disparity between consumers and producers, the emissions generated in one province might be significantly driven by final consumption in a different province. Thus, for purposes of cross-provincial action on air pollution control, it is equally important to identify both the sources and the drivers of air pollution.

In this study, we used BC as a proxy to establish the source−receptor relationship involving the atmospheric transport and trade-induced virtual transfer of primary pollution among 30 provinces in mainland China in 2007 (excluding only Tibet, where reliable data are unavailable). BC was chosen as a representative for cross-regional air pollutant mitigation because of its impact on environment, regional character and representative for other fine aerosol species. The model simulation of BC transport was undertaken using the Weather Research and Forecasting with Chemistry (WRF/Chem) model. An explicit tagging method was implemented in the WRF/Chem model to efficiently track the pathways of BC transport.[32] We also used multiregion input-output (MRIO) analysis to examine the virtual transfer of BC emissions resulting from trading goods and services.[33] By combining both physical and virtual transfers of BC emissions, we quantified the direct and indirect interprovincial linkages in terms of pollution transport. This quantification leads to feasible suggestions on the priority of BC reduction and the possibility of cooperative responsibility for pollution mitigation in China.

■ METHODOLOGY AND DATA

Emission Inventory and Data Sources. A production-based emissions inventory was developed by multiplying the energy consumption data and BC emission factors.[34,35] Energy consumption data for 30 provinces in China (as listed in Table S1 in the Supporting Information) were derived from provincial statistical yearbooks and energy balance tables from the 2008 Chinese Statistical Yearbooks (data were based on the investigation of year 2007) for each province.[36] We aggregated the provincial BC emissions into 17 sectors (listed in Table S2 in the Supporting Information) to conform with the Chinese MRIO Table.[37] Emission factors for 8 types of energy (i.e., coal, coke, gasoline, kerosene, diesel, fuel oil, liquefied petroleum gas and natural gas) were obtained from previous studies (listed in Table S3 in the Supporting Information).[34,35] The derivation of production-based BC emissions attributable to energy consumption for province f is expressed as

$$C_p^f = \sum_{i=1}^{17} \sum_{m=1}^{8} E_{i,m}^f \times \mathrm{EF}_{i,m} \qquad (1)$$

where $E_{i,m}^f$ is the energy consumption of fuel m in sector i, province f; $\mathrm{EF}_{i,m}$ is the emission factor of fuel m in sector i.

Here, we considered only industrial BC emissions from all 17 aggregated economic sectors, because industrial emissions can "flow" in interprovincial trade. According to the Chinese MRIO Table, every particular sector has a more or less monetary output to support nonlocal industry. By contrast, residential energy consumption cannot "flow" in trade, and was thus excluded in our analysis. We used the industrial BC emissions in WRF/Chem modeling for consistency with the MRIO analysis by mapping the emissions with high spatial resolution.[34] We also conducted additional simulations using revised all-source anthropogenic BC emission inventory from Wang et al. for model evaluation.[34]

Model Description and Configuration. WRF/Chem is a meteorological model that enables the simulation of atmospheric phenomena across scales ranging from meters to thousands of kilometers.[38] WRF/Chem includes chemical processes such as emissions, gas/aqueous phase chemistry and dry/wet deposition.[39] WRF/Chem has been widely applied to simulate the transport of BC and its radiative impact.[38,40,41] Real meteorological data are used as the initial and lateral boundary condition input for the WRF/Chem model to simulate the physical transport of BC aerosols. Here, we applied a data set from the NCEP FNL Operational Model Global Tropospheric Analyses, which provide data every 6 h for the period from December 16, 2006, to December 31, 2007, for model simulation. The first 2 weeks of the simulation were used for model spin-up.

To quantify source−receptor relationships among the 30 provinces, an explicit tagging technique was used in WRF/Chem to avoid modifying BC emissions.[42] This method differs from the traditional sensitivity approach to avoid reducing BC emissions that may strongly disturb the local climate. Similar approaches have been previously applied in global models to estimate the long-range transport of BC, OC and $PM_{2.5}$ between continental regions.[13,32,43] In this tagging approach, two classes of BC tracers are used for each "tagged" region. One is for hydrophobic BC, which represents freshly emitted BC species, and the other is for hydrophilic BC, which represents aged BC and has sufficient soluble coating to behave as cloud condensation nuclei (CCN). Therefore, 30 nonoverlapping geographical regions were tagged individually with additional variables to track their transportation and transformation until deposition. Tagged BC has the same physical and chemical properties as untagged BC, and the model thus accurately predicts the pathways of BC dispersion and its influence on surface concentration.

We use the WRF/Chem model to track the interprovincial source−receptor relationships for BC in 2007 with a 0.2 × 0.2° horizontal resolution. In general, the model agreed within a factor of 2 with the observations. (As shown by Figure S1 in the Supporting Information, observational data were collected from published literature.[44−57] The spatial distribution of the data is shown in Figure S2.) The output results were archived hourly and used to calculate the average surface concentrations for a province over a given period of time for analysis.

Multiregion Input−Output (MRIO) Analysis. Originating from Leontief,[58] input−output analysis has been widely used to link global and regional environmental issues with final consumption.[31,33] In the past decade, environmental MRIO analysis has been developed to quantify emissions transfer via inter-regional trade.[33,59,60] Here, we used the Chinese MRIO Table from 2007 that was developed by Liu et al. to quantify BC emissions embodied in traded products.[37] The MRIO table consists of three parts. Part One is the intermediate input/output for 17 sectors in 30 provinces. Part Two consists of provincial final consumption (i.e., urban household consumption, rural household consumption, government consumption and investment) and international export. Part Three consists of production-based BC emissions for 17 sectors in 30 provinces.

For the entire system covering all provincial economies, we have the following balance of monetary flows:

$$X = AX + Y \tag{2}$$

where X is a vector representing total monetary output for every province, A is a matrix with its elements defined as intermediate input to produce a unit output, and Y is a vector representing the total output of final consumption and international export in each province.

Consumption-based BC emissions can be obtained by introducing emission intensity, EI:

$$C_c = \mathrm{EI}(I - A)^{-1}Y_c \tag{3}$$

where EI is a vector with its elements defined as the direct BC emissions per unit of economic output, $(I - A)^{-1}$ is the Leontief inverse matrix and Y_c is the final consumption.

This basic formula can be further used to quantify emissions from the production of traded products. For instance, BC emissions embodied in the products exported from province f to province s can be calculated as

$$C_c^{fs} = \mathrm{EI}^f(I - A)^{-1}Y_c^s \tag{4}$$

where EI^f is a vector of BC emission intensity for province f but zero for all others and Y_c^s is the final consumption of province s.

■ RESULTS

Physical Transport of BC via Atmospheric Movement. Figure 1 shows the major cross-boundary influence pattern of the area-weighted annual mean surface BC concentration caused by industrial emissions. The annual mean surface BC concentrations range from 0.025 μg·m^{-3} (Qinghai) to 5.7 μg·m^{-3} (Shanghai). Shanghai and Tianjin (4.2 μg·m^{-3}) have the highest BC concentration. Major local sources of pollution for these two coastal megalopolises are traffic and transport sectors (as suggested by Figure S3), while emissions in their contiguous provinces also exert considerable influence. Industry-dominant provinces including Shandong (2.8 μg·m^{-3}), Henan (2.9 μg·m^{-3}) and Liaoning (2.0 μg·m^{-3}) also have heavy BC concentrations. Moreover, provinces with heavier BC pollution are likely to be located along or near the coastline.

Provincial BC concentrations are profoundly influenced by trans-boundary transport. The reciprocal effect between two contiguous provinces whose emissions share resemblances is generally comparable. It is particularly noticeable between Hebei and Shandong, Shandong and Henan, Jiangsu and Anhui, and Jiangsu and Shanghai, where approximately 10% of the BC concentrations in these provinces are contributed by one another. The northern provinces tend to be net contributors to the pollution load of the more southerly provinces in eastern China. Remarkably, Hebei is responsible for 0.59 μg·m^{-3} (24%) and 1.2 μg·m^{-3} (28%) surface BC concentrations in Beijing and Tianjin, respectively. It is also responsible for 0.13 μg·m^{-3} (7%) and 0.17 μg·m^{-3} (6%) of the BC concentration in Shanxi and Henan, respectively. Whereas Shanxi contributes 0.20 μg·m^{-3} (22%) BC in Shaaxi, and Henan contributes 0.23 μg·m^{-3} (19%) of the BC concentration in Hubei and 0.17 μg·m^{-3} (8%) in Anhui.

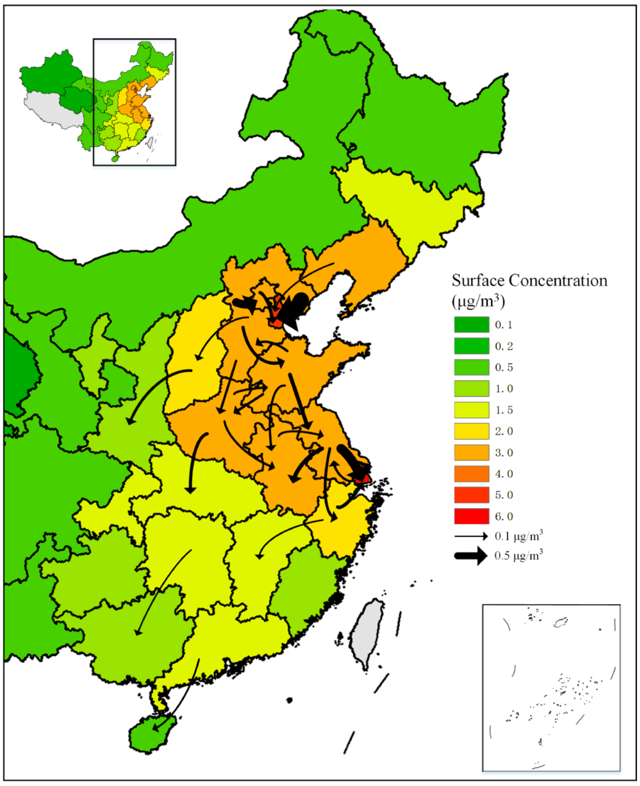

Figure 1. Largest surface BC concentration contribution via atmospheric transport within 30 provinces. The colors in the map indicate annual mean surface BC concentration. Arrows on the map reflect a typical contribution above 0.1 μg·m^{-3}. The thickness of the arrows indicates the relative magnitude of the absolute interprovincial contribution of surface concentration.

Virtual Transfer of BC via Interprovincial Trade. Figure 2(a) shows the comparison of total production-based and consumption-based BC emissions in 2007 for 30 Chinese provinces. Total industrial BC emissions amount to 894 Gg in China in 2007, which is consistent with previous studies.[34,61,62] From the production perspective, Shandong ranks first with emissions of 79.7 Gg, followed by Henan (73.5 Gg), Shanxi (61.1 Gg) and Hebei (60.3 Gg). Provincial consumption-based BC emissions present a different distribution pattern, with 741 Gg (83%) emissions induced by domestic demand. This percentage is comparable to previous results on primary PM$_{2.5}$ and gaseous pollutants including SO$_2$ and NO$_x$.[25,63] Except for Shandong (contributing 64.7 Gg emission), the southern provinces, including Zhejiang (57.2 Gg), Jiangsu (55.0 Gg) and Guangdong (51.2 Gg), hold the top positions. Remarkably,

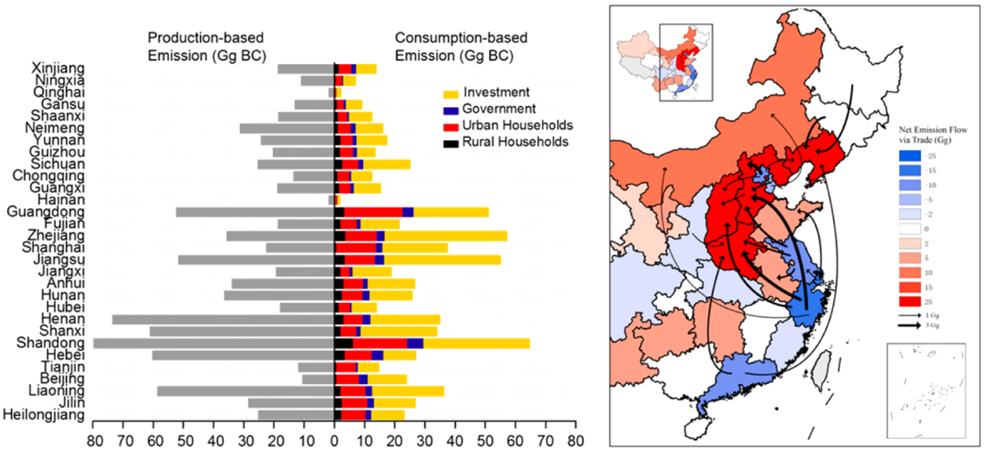

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Figure 2. (a) Comparison between production-based and consumption-based BC emissions. Consumption-based BC emissions are categorized into four types based on final consumption. (b) Largest net fluxes in "traded" BC emissions among 30 provinces. Color in the map indicates total net emission budget (emission imports minus exports) via trade. Red indicates an emission importer, i.e., more BC is emitted due to the interprovincial trade. Blue indicates an emission exporter. The arrows reflect typical cross-border net emission flows above 1 Gg in interprovincial trade. The thickness of the arrows indicates the relative magnitude of the net BC emissions transferred between provinces.

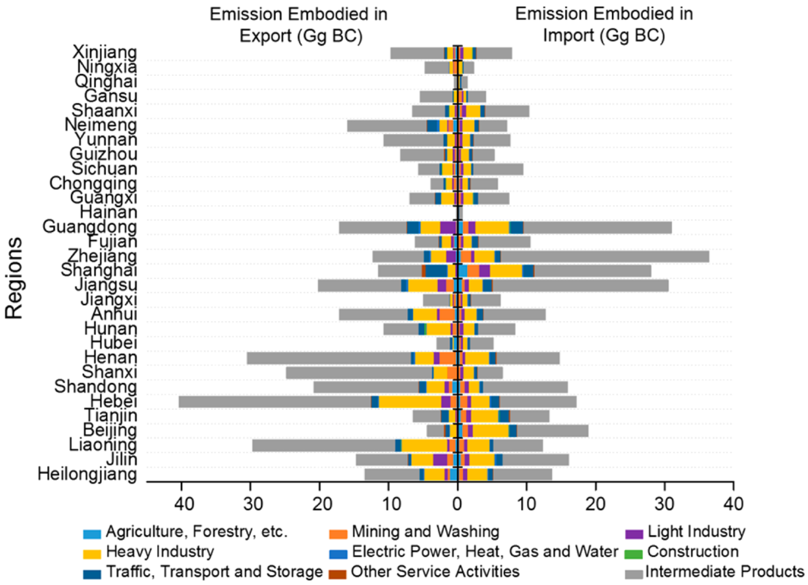

Figure 3. Sectoral BC emissions embodied in exported and imported products via interprovincial trade for 30 provinces; the 17 sectors in MRIO are sorted into 8 for clearer presentation and listed in Table S2. Intermediate products (dark gray) are embodied BC emissions used by industry.

the Yangtze River Delta contributes 20% of the total embodied BC emissions, although its domain area is less than 2% of the total area. Consumption-based emissions consist of emissions from four types of final consumption. Investment is the dominant motor driving industrial BC emissions for 29 provinces (the exception is Xinjiang), contributing approximately 40−70% of the total consumption-based BC emissions. Urban household consumption is the second largest driver of BC emissions, ranging from 15% in Shanxi to 43% in Tianjin. Government consumption and rural household consumption account for the remaining 15%.

The difference between production-based and consumption-based BC emissions indicates that emissions are transferred via trade. Figure 2(b) illustrates net emissions transfer through trade (only the largest fluxes between provinces are shown). Thirteen of 30 provinces are net emissions importers, and the other 17 provinces are net exporters. Net importers are mainly industry-dominant provinces such as Hebei (23.2 Gg), Shanxi (18.3 Gg), Liaoning (17.3 Gg) and Henan (15.7 Gg). Their industrial activities and associated emissions enhanced by trade support consumption across the country, particularly for a few developed provinces. Conversely, Zhejiang (24.1 Gg), Shanghai (16.4 Gg), Beijing (14.5 Gg) and Guangdong (13.8 Gg) are major BC exporters. They behave as exporters in trade with almost all other provinces, whereas the larger flows more often end up in Hebei and Henan.

The pattern of major flows is from southeastern China to the North China Plain (NCP) geographically and from developed to less-developed provinces economically. The demand-driven flows can be categorized into three types based on the economic strength of emission exporter and importer, with the dominant pattern being from a province with abundant capital to a province owning heavy industry (Figure 2b). First, the largest BC emissions transfer occurs from Beijing to Hebei (with 4.2 Gg BC emissions being relocated), followed by a flow of 4.1 Gg emissions from Zhejiang to Hebei. Second, shifts in emissions between contiguous provinces of comparable economic strength are also noticeable. They occur noticeably within the Yangtze River Delta and northeastern provinces, including Heilongjiang, Jilin, and Liaoning, which indicate intimate economic relationships between contiguous provinces. The typical emission flows in this category are from Shanghai to Jiangsu (1.2 Gg) and from Jilin to Liaoning (2.7 Gg). The third type of flow is from industrial provinces to resource-rich but less-developed provinces, suggesting the need for inputting fundamental raw materials for industrial activities. For example, 1.4 Gg of emissions are transported from Hebei to Shanxi. In general, trade-induced emission flows across China occur from south to north and coastal to inland, exhibiting a reversed source−receptor pattern to BC dispersion via atmospheric transport.

Apart from revealing emission flows from a regional aspect, MRIO can also explore the sectors that undertake the transfer of BC emissions in trade.[31] Figure 3 shows the sector-specific BC emission transfer embodied in interprovincial trade. At the national level, nearly half of these trade-relevant emissions are caused by the production of intermediate products. This ratio is even higher in Shanxi, Gansu, Qinghai, Xinjiang, and Yunnan, where more than 80% of the emissions are caused by the massive quantities of low-value-added raw materials and energy that are produced for export. By contrast, Zhejiang and Jiangsu have the highest proportion of intermediate goods from the import aspect because these provinces lack natural resources but are advanced in processing capabilities.

Moreover, final use accounts for the remaining 15%−55% of trade-embodied emissions, in which heavy industry (including the petro-chemical, nonmetallic mineral products, and metallic mineral products) plays a dominant role in most provinces due to its intensive energy consumption. In addition to heavy industry, exporting agricultural products induces salient BC emissions in Hubei and Sichuan provinces; mining and washing are responsible for most trade-relevant BC emissions in Shanxi and Liaoning provinces. However, for Jiangsu and Zhejiang, light industry such as textile and timber processing is key to generating BC emissions. With regard to imported products, less-developed provinces tend to have a higher proportion of emission output in high-value products of light industry, whereas the provinces scarce in energy and raw materials are likely to depend on products of mining and washing in other provinces.

Surface BC Concentrations from a Production and Consumption Perspective. By combining atmospheric BC transport and emission flow in trade, the source of surface BC concentrations can be classified according to their on-site emission region and the final consumer of the relevant products. With regard to whether BC is emitted locally or from other provinces, BC concentration can be classified as either local or domestic production. Meanwhile, from a consumption-based perspective, BC concentration can be referred to as local, domestic or foreign consumption. We specify "domestic consumption" as the consumption from the other 29 provinces, and "foreign consumption" as the internationally exported products from the province where production-based BC is generated. From an on-site emission perspective, 9 provinces (i.e., Hubei, Anhui, Jiangxi, Hainan, Guangxi, Chongqing, Guizhou, Shaanxi, and Qinghai) have a share of domestic-production concentration (i.e., originated from the other 29 provinces, namely the green, yellow, and light gray portions in Figure 4) over 50%. Others are typically within 30−50%, and the lowest (3%) is in Xinjiang. This indicates the importance of local emissions, but trans-boundary transport is also a noteworthy contributor. With regard to the key provinces for air pollution mitigation, Beijing has a proportion of domestic-production BC surface concentration equal to 42%, Tianjin 45%, Shanghai 22%, Jiangsu 39%, Zhejiang 41%, and Guangdong 40%. BC concentration originating from other provinces but induced by local consumption (i.e., the green portion in Figure 4) is very small (<3%) and mainly driven by neighboring provinces.

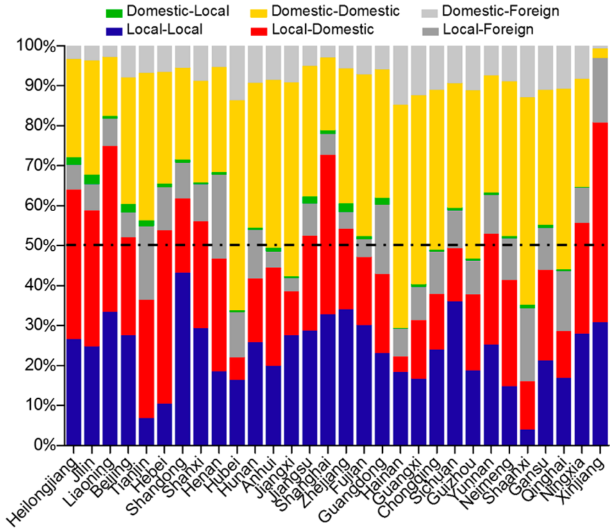

Figure 4. Contribution of surface BC concentration in 30 provinces from both production and consumption perspectives. Blue (local−local), red (local−domestic), dark gray (local−foreign) columns indicate the percentage of surface concentration in a province that is contributed by its own on-site emissions but induced by consumptions from its own province, the rest 29 provinces, and foreign countries, respectively. Comparably, green (domestic−local), yellow (domestic−domestic), and light gray (domestic−foreign) columns, respectively, indicate the percentage that is contributed by the emissions released in the other 29 provinces but induced by local, domestic and foreign consumptions. The dashed line at 50% marks the comparison between surface concentrations contributed by local and nonlocal on-site emissions.

From the consumption perspective, final demands within mainland China on average induce 82% of provincial surface BC concentration across China. Domestic-consumption-induced concentration (i.e., the red plus yellow portions in Figure 4) is greater than local-consumption-induced concentration (i.e., the blue plus green portions in Figure 4) for all provinces, which indicates the profound influence of inter-regional trade and trans-boundary transport on concentration. The proportion of BC concentration generated locally but induced by domestic consumption ranges from 4% in Hainan to 50% in Xinjiang and reflects the comparative industrial scales for supporting local living standards or for securing economic growth via exports. This proportion is above 30% for typical emission importers. Particularly, 43 and 41% of the concentration in Hebei and Liaoning, respectively, is contributed by the local emissions induced by domestic consumption. Meanwhile, for emissions-exporting provinces, such as Beijing, Zhejiang and Guangdong, this proportion falls

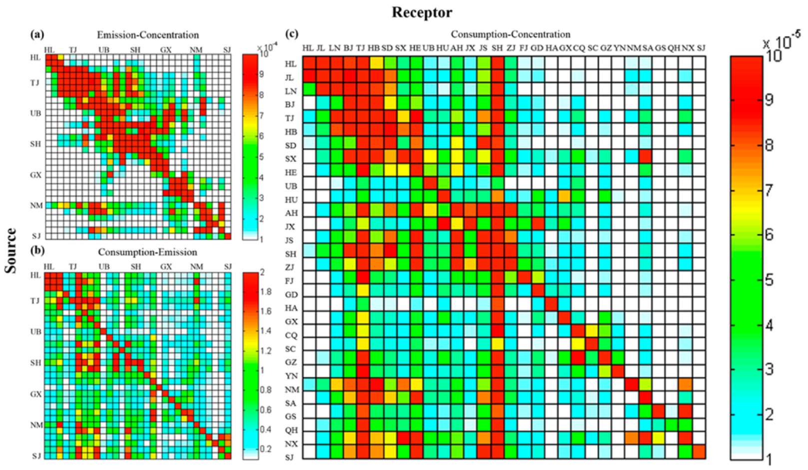

Figure 5. (a) Source-receptor relationship between provincial emission and BC concentration (area-weighted at the surface) among 30 provinces (unit: (μg·m^{-3})/(Gg·yr^{-1})). (b) Source-receptor relationship between provincial final consumption and on-site emission (unit: tons per billion Yuan). (c) Source-receptor relationship between provincial final consumption and surface BC concentration (unit: (μg·m^{-3})/(billion Yuan·yr^{-1})). Province abbreviations are HL, Heilongjiang; JL, Jilin; LN, Liaoning; BJ, Beijing; TJ, Tianjin; HB, Hebei; SD, Shandong; SX, Shanxi; HE, Henan; UB, Hubei; HU, Hunan; AH, Anhui; JX, Jiangxi; JS, Jiangsu; SH, Shanghai; ZJ, Zhejiang; FJ, Fujian; GD, Guangdong; HA, Hainan; GX, Guangxi; CQ, Chongqing; SC, Sichuan; GZ, Guizhou; YN, Yunnan; NM, Neimeng; SA, Shaanxi; GS, Gansu; QH, Qinghai; NX, Ningxia; SJ, Xinjiang (also, Table S1 in the Supporting Information).

to near 20%. However, Shanghai has a higher percentage (40%) of local−domestic concentration because of its considerable share of industry to support domestic consumption.

Source-Receptor Relationship from Combined Production and Consumption Perspectives. To combine the influence of atmospheric transport and interprovincial trade on surface BC concentrations and emissions, which is informative for judging the priority of cooperative action for pollution mitigation, we introduce three source−receptor indicators shown in Figure 5.

Figure 5(a) shows the emission-concentration relationship that is defined as the annual averaged surface BC concentration over a receptor resulting from a unit of emission in a source region (in (μg·m^{-3})/(Gg·yr^{-1})).[29] This atmospheric transport efficiency is calculated by dividing the source-produced concentration in a receptor by total annual emission in the source region. The colored chart shows a pattern in which a receptor is more sensitive to its own emissions than to domestic emissions and to upwind contiguously located sources than to remote ones. Normalized BC concentrations resulting from local emission range from 0.0047 (μg·m^{-3})/(Gg·yr^{-1}) in Xinjiang to 0.20 (μg·m^{-3})/(Gg·yr^{-1}) in Shanghai, which is mainly determined by the emission density. Nonlocal contributions to BC concentration from neighboring provinces is typically 1−2 orders of magnitude smaller than local emissions but more than 1−2 orders of magnitude larger than emissions from remote provinces. Provinces within Jing-Jin-Ji and the Yangtze River Delta share a close relationship in BC concentration through atmospheric transport. For instance, 1 Gg annual emission in Beijing and Hebei can increase the surface BC concentration in Tianjin by 0.022 and 0.019 μg·m^{-3}, respectively. Similarly, the bilateral influence between Jiangsu, Shanghai and Zhejiang ranges from 0.003 to 0.013 (μg·m^{-3})/(Gg·yr^{-1}).

Figure 5(b) shows the trade-induced consumption-emission relationship that is defined as the production-based BC emissions of a receptor associated with a unit of domestic consumption from a source (in tons of BC per billion Yuan), in which consumption includes the sum of four final consumption.[30] This BC intensity is calculated by dividing the source-induced BC emission in a receptor by total annual final consumption in the source region. Unlike Figure 5(a), in which the pattern of atmospheric transport exhibits a diagonal distribution, the consumption-emission graph reflects a column-like distribution, which indicates that massive amounts of BC imported from almost all other provinces via trade into some industry-dominant provinces (e.g., Hebei, Henan, and Liaoning). Normalized production-based BC emissions induced by local consumption range from 3.4 tons per billion Yuan in Tianjin to 53.4 tons per billion Yuan in Shanxi. Generally, this value is higher in provinces with massive energy consumption than in developed provinces with strict environmental laws. However, those developed provinces relocate significant amounts of emissions to other provinces. The proportion of emissions caused in other 29 provinces is comparable to the proportion of local on-site emissions in developed provinces. Every billion Yuan of consumption in Tianjin produces 5.6 tons of BC emissions in Hebei, 1.6 times its own local emission intensity. Similarly, Hebei and Henan receives 2.4 and 2.1 tons of BC emissions, respectively, for every billion Yuan of consumption in the Yangtze River Delta. Heilongjiang and Jilin shift 7.2 and 8.4 tons of BC emissions, respectively, to Liaoning for every one billion Yuan of consumption.

Figure 5(c) shows the consumption-concentration relationship, that is, annual averaged surface BC concentration in a receptor resulting from a unit of domestic consumption in a source (in (μg·m^{-3})/(billion Yuan·yr^{-1})), which considers the joint influence of trans-boundary transport and inter-regional trade on surface BC concentration together. This consumption influence efficiency is calculated by concentration in a receptor caused by on-site emissions in 30 provinces that are induced by a unit of annual consumption in a source. The pattern of the graph is a combination of Figure 5(a,b), showing both aggregated groups along the diagonal and column-like distribution of eminent contributions. The bilateral influence of domestic consumption within Jing-Jin-Ji and the Yangtze River Delta is more than 8 × 10^{-5} (μg·m^{-3})/(billion Yuan·yr^{-1}). Northeastern provinces also show intimate internal relationships regarding both atmospheric transport and interprovincial trade. Surprisingly, Tianjin and Shanghai are the two provinces most vulnerable to consumption-based emissions in other provinces despite being net BC exporters in interprovincial trade. The average surface BC concentration resulting from a unit of consumption in a nonlocal source region in Shanghai and Tianjin is 1.5×10^{-4} ($\mu g \cdot m^{-3}$)/(billion Yuan·yr^{-1}) and 1.4×10^{-4} ($\mu g \cdot m^{-3}$)/(billion Yuan·yr^{-1}), respectively. This phenomenon can be attributed to the considerably high BC concentration resulting from a unit of local-production emission and a prevailing proportion of local-production emission induced by domestic consumption than by local consumption. Although they are densely urbanized metropolises, Tianjin and Shanghai have a considerable scale of secondary industry, which induces a massive emission import (S3, Supporting Information). In addition, the source−receptor relationship between consumption and concentration is also noticeable within some emission exporters and importers in interprovincial trade. Per-billion Yuan annual consumption from a source province can lead to an approximately 8×10^{-5} $\mu g \cdot m^{-3}$ increase in BC concentration in a receptor province. In particular, BC concentrations in Liaoning and Hebei are largely affected by domestic-consumption emissions particularly from the Jing-Jin-Ji area and Shanxi, whereas the concentrations in Anhui and Jiangsu are affected by emissions from the Yangtze Delta area and Anhui. In Henan and Shandong, BC concentration is sensitive to consumption in all the source provinces mentioned above. These provinces may show a close relationship for air pollution control.

■ DISCUSSION

Using WRF/Chem modeling and environmental MRIO analysis, we quantified the source−receptor relationship of atmospheric transport and trade-induced geographical relocation of BC emissions. By combining the dual effects, we traced the influence from both producer and consumer perspectives on BC surface concentration for each province in China. The results can provide insights into collaborative efforts on air pollution control for policy makers.

The source−receptor relationship of physical BC transport among provinces is largely determined by both the amount of BC emissions and the direction of prevailing winds. Depending on locations, more than 20% of surface BC concentration may originate from a neighboring province, particularly for provinces such as Tianjin and Hubei, which are located downwind contiguously of major BC source provinces (such as Hebei and Henan). By contrast, provinces such as Hebei, Shanxi, Henan, Zhejiang, and Guangdong transport substantial BC pollution to their neighbor provinces. Notably, in inland China, where the north wind is dominant particularly in autumn and winter because of the influence of the Siberian High, BC transport typically occurs more usually from north to south. In southeastern coastal China, however, where airflow is driven by the Hawaiian High, BC transport occurs mainly northwestward during the summer. Combining these two factors, the proportion of surface BC concentration caused by the other provinces varies from 3 to 71%. Provinces such as Hubei, Shaanxi, and Hainan are vulnerable to nonlocal emissions because of their relatively small scale of industry compared with their neighbors with massive industrial production. Beijing, Shandong, Jiangsu, and other provinces with on-site emissions comparable to those of their neighbors are also sensitive to domestic-production emissions. Several provinces, such as Xinjiang, Liaoning and Shanghai, have a percentage above 75% of the local-production surface BC concentration owing to their location and the total amount of BC emissions, which indicates a local-production dominant situation.

Unlike atmospheric transport, which is driven by natural forces, domestic trade relocates BC in a different way. Beijing, Tianjin, Guangdong and the Yangtze River Delta are more likely to outsource BC emissions via interprovincial trade to industrial provinces including Hebei, Henan, Shanxi, and Liaoning. For developed provinces such as Beijing and Shanghai, consumption-based BC emissions can be double the production-based BC emissions, whereas in industry-dominant provinces such as Hebei and Shanxi, net BC emissions transferred via trade amount to approximately 30% of their production-based emissions. Surface BC concentration generated by local emission but induced by domestic consumption can account for more than 30% of the total concentration in these provinces. In addition, three northeastern provinces show tight economic connections, and Liaoning plays the major role of emissions importer. This imbalance in interprovincial trade may be due largely to the enormous disparity in wealth and economic structure among provinces.[64] Although emission transport via trade is bilateral, developed provinces are more likely to import low value-added commodities in the heavy industry, mining and washing, and agricultural sectors from less-developed provinces according to their mainstay industry, while exporting technology-containing commodities from light industry.

Combining these two aspects, it is reasonable to say that the patterns of atmospheric transport and interprovincial trade are the opposite of one another. Emission flows from Beijing and Tianjin to Hebei are transported in reverse to influence the local BC concentration. Similar patterns can also be observed from Shanghai to Jiangsu and Zhejiang. Moreover, consumption in developed provinces located along the southeastern coastline increases BC concentration and exerts adverse influence on air quality in those emission input industry-dominant provinces, mainly on NCP via trade. These phenomena may serve as a major motivation for the close cooperation between provinces on air pollution control as advocated in the “Law of the People’s Republic of China on the Prevention and Control of Atmospheric Pollution.” By introducing advanced technology from developed provinces to industry-dominant provinces and by taking cross-regional governance into consideration, the supportive provinces benefit in that their surface pollutant concentration caused by domestic-production emissions is reduced. Meanwhile, cooperation contributes to reducing surface BC concentration in the emission source, which may compensate for the pollutant transferred via interprovincial trade. Overall air quality in China can also be enhanced because downwind provinces suffer less from trans-boundary emissions. Generally, neighboring provinces have a more intimate relationship concerning both trans-boundary transport and interprovincial trade, which indicates their optimal prospects for joint efforts on mitigating air pollution. Two partly overlapping control zones for collaborative action on air pollution mitigation with prior concern are promoted according to our study. The Huabei control zone, led by the Jing-Jin-Ji area, together with Shandong, Shanxi, Liaoning, and Henan has a close relationship with respect to both trans-boundary transport and virtual transfer of emissions. Liaoning is also intimate with Jilin and Heilongjiang and may implement multilateral supervision action. Meanwhile, for Shandong and Henan, cooperation with the Yangtze River Delta (i.e., Jiangsu, Shanghai, and Zhejiang) and Anhui can also achieve enhanced efficiency in emission mitigation. These six provinces comprise the Huadong control zone.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b05989.

Provinces and sectors information, emission factors of BC for eight fuel types, sectoral distribution of provincial production-based BC emissions, and the evaluation of model simulation with observations. (PDF)

Simplified Chinese MRIO Table (2007) for 30 provinces and 17 sectors. (XLSX)

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Notes

The authors declare no competing financial interest.

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