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Interprovincial trade driven relocation of polycyclic aromatic hydrocarbons and lung cancer risk in China

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Highlights

- Interprovincial trade induced BaP cancer risk in China was estimated
- Virtual PAH emissions were transferred from western and central China to coastal regions
- Interprovincial trade contributed 42% to cancer risk via BaP exposure in 2007
- Cancer cases declined in coastal region and increased in western region
- Overall interprovincial trade reduced Chinese lung cancer cases

Abstract

Polycyclic aromatic hydrocarbons (PAHs) are a class of ubiquitous organic contaminants which poses an adverse health impact on environment and humans. This study assesses the PAHs environmental contamination and associated lung cancer risk attributable to interprovincial trade in goods and services in China. Virtual trade driven PAHs flow mainly from well-developed and industrialized provinces to less-developed provinces that provide energy and raw materials. In 2007, Shanxi (with a net PAHs outflow of 3743 tons) and Hebei (with a net PAHs outflow of 851 tons) account for 66.8% of total PAH emission outflow attributable to interprovincial trade. The largest single net PAHs flow was from Shanxi to Zhejiang (399 tons), followed by Shanxi to Jiangsu (371 tons), and Hebei to Zhejiang (194 tons). Our results also reveal a switching from outflow to inflow of industrial products with high PAH emissions in some provinces from 2007 to 2012 due to the changes in their industrial structure. The estimated incremental lifetime cancer risk (ILCR) based on modeled benzo[a]pyrene (BaP) concentrations and considering trade driven emissions shows that excess nonoccupational lung cancer cases associated with trade related industrial BaP emissions totaled 2176 in 2007, accounting for 42% of lung cancer cases induced by all Chinese BaP emissions. Without interprovincial trade, Chinese lung cancer cases would increase to 3677 in 2007, indicating that interprovincial trade reduces lung cancer cases in well-developed and populated coastal provinces and increases cases in less populated and less developed provinces, which reduces the overall number of Chinese lung cancer cases. It is recommended that well-developed provinces in eastern

and southern China should subsidize those inland provinces providing goods and services to eastern and southern China, in addition to the interprovincial trade.

Keywords: China, Interprovincial trade, PAHs, Lung cancer risk

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous organic contaminants in the environment which have raised widespread concerns given their toxicity and adverse effect on human health (Zhang et al., 2009b). Due to their physicochemical and mutagenic properties and carcinogenicity, the Convention on Long Range Transboundary Air Pollution Protocol in Persistent Organic Pollutants (POPs) of United Nations Economic Commission (UNCEC , http://www.unece.org/env/lrtap/pops_h1.htm) has included PAHs in the POPs list. As the largest emitter of anthropogenic air pollutants in the globe, China has been suffering from severe environmental contamination (Lin et al., 2014). Among those air pollutants, ambient PAH concentrations in many Chinese localities are considerably higher than those in developed countries, leading to high human exposure risks to these toxic chemicals (Xie et al., 2017). One of the most important human exposure pathways for PAHs is inhalation exposure. Globally, incremental lifetime lung cancer risk (ILCR) induced by ambient PAH exposure is 3.1×10^{-5} in 2007 (Shen et al., 2014). In particular, PAHs bound to $PM_{2.5}$ have been considered the major contributors to human health risks from $PM_{2.5}$ (Xie et al., 2017).

A large portion of atmospheric PAH emissions originate as a byproduct of industrial activities and burning of fossil fuels (Vardar et al., 2008). Over the past decades, rapid industrialization and urbanization have led to significant increases in Chinese PAH emissions. Previous studies have shown that industrial sources (all emission sectors listed in **Table S1**) accounted for approximately 35% of total PAH emissions in 2004 and 2007, second to biomass burning (approximately 65% of total PAH emissions in 2007) (Shen et al., 2013; Zhang and Tao, 2009). PAH emissions are characterized by substantial disparities between provinces or regions owing to the huge differences in resource and energy endowments, economic structures, population, and lifestyles in China (Xu et al., 2006; Zhang et al., 2007). Recent investigations have revealed that emissions of anthropogenic air pollutants are significantly redistributed among provinces due to interprovincial trade (Ling et al., 2019; Wang et al., 2019; Zhao et al., 2015). To secure and supply sufficient energy, goods, and services to support rapid industrial, economic, and social development in China, the interprovincial and interregional trade of goods and services increased substantially in recent decades. Extensive studies have demonstrated that the trade of goods and services has become an important driver of the release of air pollutants and greenhouse gases (Davis et al., 2011; Liu et al., 2019; Meng et al., 2018). It is necessary that integrate consumer perspective to consider the importance of environmental damage by air pollutants (Wang et al., 2017; Zhang et al., 2019). Trade plays a key role in overcoming resource bottlenecks. However, trade also creates the environmental inequality due to environmental linkages (Ling et al., 2019; Zhang et al., 2017). It has been found that

121 emissions of atmospheric pollutants have grown rapidly in some inland and less
122 developed provinces in Western China, but they have stabilized or decreased in coastal
123 and well-developed provinces and regions, for reasons related to regional economic
124 status and environmental policy (Zhao et al., 2015). Often, rapidly increasing air
125 emissions in less developed regions have been partly attributable to goods and services
126 trade operated to meet the rapidly rising energy and resource demands of the well-
127 developed provinces (Moran and Kanemoto, 2016; Ou et al., 2019; Wang et al., 2017).
128 Given the similarities in emission sources of PAHs and other air contaminants, we
129 would expect that increases in interprovincial trade could also act as an important
130 source of PAHs in China.

131 Extensive studies have been devoted to investigating emission of air pollutants and
132 greenhouse gases attributable to international and interregional trade using
133 environmentally extended multiregional input-output (MRIO) analysis and
134 corresponding climate, ecological, and health effects (Chen et al., 2016; Davis and
135 Caldeira, 2010; Yi et al., 2019). The significance of international and interregional trade
136 to the emission and redistribution of carbon dioxide, black carbon, $PM_{2.5}$ and its
137 precursors, and mercury has been assessed and quantified (Chen et al., 2018; Meng et
138 al., 2018; Zhang et al., 2017). Early studies have been carried out to assess
139 consumption-induced CO_2 emissions using input-output models (Davis and Caldeira,
140 2010; Feng et al., 2013). MRIO analysis was extended to quantify air pollutants
141 emissions, environmental contaminants, and corresponding health exposure risks
142 afterwards. Jiang et al. (2015) showed that export related $PM_{2.5}$ emissions caused

157,000 premature deaths, accounting for 12% of the national total induced by $PM_{2.5}$-related air pollution in China in 2007. Zhang et al. (2017) found that approximately 22% of total global premature deaths related to $PM_{2.5}$ pollution was associated with international trade in 2007. Wang et al. (2017) demonstrated that more than 50% of air pollutant emissions in China was related to interregional trade. Zhang et al. (2018) tracked the imbalance of economic benefits and environmental costs induced by the regional import/export, which caused greater emissions of air pollutants occurring in less developed regions. Ling et al. (2019) showed that sulfur dioxide (SO_2) emissions embodied in west-east energy transmission accounted for more than 40% of total SO_2 release in Northwestern China in the 2000s.

The influences of goods and services trade on PAH emissions, environmental contamination, and exposure risks are almost unknown. The present study explores the redistribution of PAH emissions and human exposure risks attributable to Chinese interprovincial trade. We employed the MRIO model to generate estimated trade driven PAH emissions in 2007 and 2012. As a case study, the CanMETOP (Canadian Model for Environmental Transport of Organochlorine Pesticides) was used to simulate atmospheric concentration of benzo[a]pyrene (BaP), a representative PAH compound often used as marker for carcinogenic PAHs (Li et al., 2012), by implementing BaP emission released from Chinese interprovincial trade in 2007 and 2012. Finally, BaP atmospheric concentrations in 2007 and 2012 were incorporated into a lung cancer risk model to assess human inhalation exposure risk.

2. Materials and methods

Sixteen PAHs recommended by the USEPA priority control list of toxic chemicals were analyzed (https://archive.epa.gov/epawaste/hazard/wastemin/web/pdf/pahs.pdf). To examine the transfer of trade driven PAH emissions among 30 provinces in mainland China, three models were implemented to quantify human health exposure to PAH attributable to interprovincial trade. We first generated PAH emissions embodied in interprovincial trade in 2007 and 2012 using an environmentally extended multiregional input-output (MRIO) model. We then performed multiple modeling scenario simulations using CanMETOP to simulate BaP concentration, the most toxic congener among 16 PAHs, in 2007 and 2012 with and without trade included, thereby to assess the contribution of trade to BaP environmental contamination and human exposure risk across China. Finally, using incremental lifetime cancer risk (ILCR) (Shen et al., 2014), we assessed trade induced cancer risks due to inhalation exposure to BaP in 2007 and 2012, respectively. We focused on these two years because the MRIO tables are available only in 2007 and 2012.

2.1 Production-based PAH emissions inventory

The production-based emissions inventory on a provincial basis, constructed from the anthropogenic emissions of each individual PAH congener from the 30 industrial sectors (**Table S1**) in 2007 and 2012, was established based on emission activities in a province and emission factors. Among industrial activities, energy consumption data for 30 Chinese provinces were collected from the 2008 and 2013 provincial statistical yearbooks of each province and the 2008 and 2013 China Economic Census Yearbook

(http://www.stats.gov.cn/tjsj/), which provided 2007 and 2012 energy production and consumption data. We aggregated the provincial energy consumption data into 30 sectors to conform with the Chinese MRIO table for 2007 and 2012 to generate provincial PAH emissions embodied in interprovincial trade (Liu et al., 2012; Mi et al., 2017). The PAH emission factors for different energy types (e.g., coal, petroleum, and natural gas) were obtained from the literature (Shen et al., 2013; Xu et al., 2006; Zhang et al., 2009a), as shown in **Table S2**.

Production-based PAH emissions from energy consumption in each province are given by:

$$E_i^a = \sum_{k=1}^{n} EA_{i,k}^a \times EF_{i,k}^a \quad (1)$$

where $EA_{i,k}^a$ denotes the *k*th energy consumption in the *a*th province from *i*th sector, and $EF_{i,k}^a$ is the corresponding emission factor. For the investigation of PAH emissions induced by interprovincial trade, we only took sectoral PAH emissions from national economic industry (**Table S1**) into account because only industrial emissions can "flow" among different provinces and regions subject to provincial/regional trade (Meng et al., 2018; Yi et al., 2019; Zhang et al., 2019). For the convenience of subsequent discussions, we regrouped 30 provinces into 8 Chinese regions (**Fig. S1**, **Table S4**) and 30 emission sub-sectors into 8 emission sectors (**Table S1**). The reconstructed production-based BaP emissions from four sectors including industry, transport, agriculture, and energy production in China in the present study are 462 and 369 tons in 2007 and 2012, respectively, agreeing with the PKU-BaP annual emissions from the four sectors at 445 tons in 2007 and 356 tons in 2012.

2.2 MRIO analysis of PAH emissions attributable to interprovincial trade

Trade driven PAH emissions in each province of China were estimated using the environmentally extended multiregional input-output (MRIO) model (Chen et al., 2018; Liu et al., 2019; Wang et al., 2019). The MRIO analysis has been extended to address the industrial production flows and air pollutants emissions caused by global and regional trade (Chen et al., 2018; Yi et al., 2019; Zhang et al., 2019). Considering the sectoral economic output in each province, the MRIO model estimates the sectoral output produced in one province and consumed in another province can be expressed by

$$\boldsymbol{x}^{a} = \boldsymbol{A}^{aa}\boldsymbol{x}^{a} + \sum_{b\neq a}\boldsymbol{A}^{ab}\boldsymbol{x}^{b} + \boldsymbol{y}^{aa} + \sum_{b\neq a}\boldsymbol{y}^{ab} \tag{2}$$

where a and b indicate province a (producer) and b (consumer); $\boldsymbol{x}^{a}$ and $\boldsymbol{x}^{b}$ represent the output of province a and province b, respectively; $\boldsymbol{y}^{ab}$ represents the final demand (investment, government consumption, urban household consumption, and rural household consumption) from province a to province b, and $\boldsymbol{A}^{ab}$ represents the intermediate requirements from province a to province b. The vectors with m provinces in Eq. (2) can be expanded as

$$\boldsymbol{x} = \begin{pmatrix} \boldsymbol{x}^{1} \\ \boldsymbol{x}^{2} \\ \boldsymbol{x}^{3} \\ \vdots \\ \boldsymbol{x}^{m} \end{pmatrix},\ \boldsymbol{y} = \begin{pmatrix} \sum_{b}\boldsymbol{y}^{1b} \\ \sum_{b}\boldsymbol{y}^{2b} \\ \sum_{b}\boldsymbol{y}^{3b} \\ \vdots \\ \sum_{b}\boldsymbol{y}^{mb} \end{pmatrix},\ \boldsymbol{A} = \begin{bmatrix} \boldsymbol{A}^{11} & \boldsymbol{A}^{12} & \cdots & \boldsymbol{A}^{1m} \\ \boldsymbol{A}^{21} & \boldsymbol{A}^{22} & \cdots & \boldsymbol{A}^{2m} \\ \vdots & \vdots & \ddots & \vdots \\ \boldsymbol{A}^{m1} & \boldsymbol{A}^{m2} & \cdots & \boldsymbol{A}^{mm} \end{bmatrix}. \tag{3}$$

When solved from total output, Eq. (2) can be rewritten as

$$\boldsymbol{x} = (\boldsymbol{I} - \boldsymbol{A})^{-1}\boldsymbol{y} \tag{4}$$

where $(\boldsymbol{I} - \boldsymbol{A})^{-1}$ is the Leontief inverse matrix.

Based on PAH production emission inventory (described in section 2.1), we

estimate sector specific consumption-based PAH emissions for Chinese provinces by combining Eq. (4). We use following relationship to calculate consumption-based PAH emissions attributable to trade flow:

$$E_c = \widehat{EI}[(\boldsymbol{I} - \boldsymbol{A})^{-1}\boldsymbol{y}] \tag{5}$$

where E_c is consumption-based PAH emissions, *EI* is emission intensity for 30 sectors (see **Table S1**) in 30 Chinese provinces (**Table S4**), and $\widehat{EI}$ is the diagonalization of *EI*. Sector specific PAH emission intensities (*EI*) can be obtained as sectoral production-based emissions divided by sectoral outputs for each province.

The production-based emissions from one region can be decomposed into the components induced by goods and services consumed in local and other regions (Eq. 6). The consumption-based emissions can be divided into components generated in local and those importing from other region, driven by the regions' consumption activities (Eq. 7).

$$E_p^a = E_c^{aa} + \sum_{\substack{b=1 \\ b\neq a}}^{30} E_c^{ab} \tag{6}$$

$$E_c^a = E_c^{aa} + \sum_{\substack{b=1 \\ b\neq a}}^{30} E_c^{ba} \tag{7}$$

where E_p^a and E_c^a are production-based and consumption-based PAH emissions in province *a*, respectively. E_c^{aa} represents PAH emissions induced by final demand from province *a* produced locally. E_c^{ab} stands for PAH emissions induced by final demand from province *b* produced in province *a*, and vice versa. The difference between production-based emissions and consumption-based emissions in each province indicates the net transfer of PAH emissions attributable to interprovincial trade.

A positive difference can be interpreted as a net outflow, and a negative value indicates a net inflow of PAH emissions.

2.3 BaP emissions gridding

Here, we choose BaP, the most toxic congener among 16 PAHs listed by the USEPA, and to evaluate its impact on human health subject to interprovincial trade. To input the trade related provincial BaP emissions into CanMETOP model grids at a $0.25° \times 0.25°$ latitude/longitude resolution, provincial BaP emissions were converted into gridded emissions. When interprovincial trade was taken into consideration, gridded BaP emissions with $0.25° \times 0.25°$ latitude/longitude resolution (E_{TRADE}^{g}) were obtained by adding up the gridded BaP emissions on a grid resolution of $0.1° \times 0.1°$ latitude/longitude resolution from the PKU-BaP inventory (http://inventory.pku.edu.cn/home.html), which accounts for major industrial sectors (coke production, transportation, and industry). If interprovincial trade was not considered, BaP emissions without trade can be calculated as

$$E_{NOTRADE}^{g}=\sum_{i=1}^{n} E_{TRADE}^{g,i} \times \left(1-\frac{E_{T}^{i,a}}{E_{TRADE}^{i,a}}\right) , \quad (8)$$

where $E_{TRADE}^{g,i}$ is gridded BaP emissions considering interprovincial trade from *i*th sector (coke production, transportation, and industry). $E_{TRADE}^{i,a}$ is BaP emissions considering interprovincial trade from *i*th sector in province *a*. $E_{T}^{i,a}$ is net BaP emissions, defined by $E_{ex}-E_{im}$, where E_{ex} and E_{im} are the BaP emission embodied in interprovincial export and import, respectively, which can be obtained from MRIO model and production-based PAH emission calculated by Eq. (1). Since PAH emissions from residential biomass burning occurs locally and is not involved in

the interprovincial trade, PAH emissions from biomass burning was not included in this investigation.

2.4 CanMETOP modeling of trade related BaP air concentration

The CanMETOP atmospheric transport model for POPs was used to simulate the tempo-spatial distribution of atmospheric concentration of BaP in a $0.25°$ latitude $\times$ $0.25°$ longitude grid across China and surrounding areas (**Fig. S2**) (Ma et al., 2003; Ma et al., 2004). The model included 3-D atmospheric advection, eddy diffusion, dry/wet deposition, gas-particle partitioning, and degradation processes in air, soil, and water (Ma et al., 2003). Other details are referred to the **Text S1** of Supporting Information.

To assess the influence of interprovincial trade driven PAH emissions on PAH environmental contamination and human health exposure risk, two modeling scenarios were implemented for our numerical investigations. The first scenario, referred to as the TRADE run, takes interprovincial trade into account. Interprovincial trade related BaP emission are implicitly included in the production-based BaP emission inventory (E_{TRADE}, **Table S5**), which includes major industrial sectors (coke production, transportation, and industry). The second scenario, referred to as NO_TRADE run, used BaP emissions without taking interprovincial trade into account (E_{NO_TRADE}, **Table S5**), calculated by Eq. (8). This formula indicates that BaP emissions were released locally with no reference to any interprovincial trade activities. The same model configurations (physicochemical and meteorology) were employed in the two simulation scenarios, as shown in the **Table S5**. The impact of interprovincial trade on BaP environmental

contamination and human health risk was assessed quantitatively by the fraction of BaP air concentration from the TRADE run to that of the NO_TRADE run, calculated by $\left[\left(C_{TRADE} - C_{NO\_TRADE}\right)/C_{TRADE}\right] \times 100$, where C_{TRADE} and C_{NO_TRADE} are the BaP air concentrations from TRADE and NO_TRADE simulations, respectively.

2.5 Lung cancer risk assessment

As mentioned previously, given strong carcinogenic properties, modeled ambient atmospheric concentrations of BaP were employed to assess gridded human inhalation exposure to trade related PAH across China, using the ILCR model (Shen et al., 2014; Xu et al., 2018). ILCR is the product of the cancer slope factor (CSF), the lifetime average daily dose (LADD), and the overall susceptibility (SUS) (Shen et al., 2014; Xia et al., 2013). Among these three parameters, LADD is associated with respiratory rate, body weight and lifetime, SUS with genetic susceptibility (GeneSus), ethnicity adjusted factor (EAF) and age sensitivity factor (ASF). Hence, ILCR (unitless) can be defined as

$$ILCR = CSF \times LADD \times SUS$$
$$= \sum_{\alpha}\left[CSF \times \frac{\left(C \times IR_{\alpha,\beta} \times n_{\alpha,\beta}\right)}{BW_{\alpha,\beta} \times LE} \times \left(GeneSus \times EAF \times ASF_{\alpha}\right)\right] \quad (9)$$

where the subscripts α and β stand for age and gender, C (mg/m^3) is modeled BaP air concentration, IR (m^3/day) is the inhalation rate, n (yr) is the exposure duration, BW (kg) is the body weight, LE is the average life expectancy (70 years). We wish to note that there are other factors causing lung cancer, such as smoking and coking. In fact, both smoking and cooking are major sources generating and releasing PAHs. In Eq. (9), the ethnicity adjusted factor (EAF) is implemented in to excludes lung cancer

incidences caused by smoking (Shen et al., 2014). In addition, the cancer slope factor (*CSF*) in Eq. (9) was obtained as the maximum likelihood estimate based on epidemiological data collected from the studies for PAH induced lung cancer incidents occurring in coke oven workers, using a multistage type mode.

In Eq. (9), the *CSF* of 3.14 kg(body weight)·day/mg for BaP was adopted (Xia et al., 2013). The life span of each age category for different gender in China, defined as the exposed year *y*, was collected from China Statistic Yearbook (2019). *EAF* was taken as 0.86 for Asian, and *ASF* values were taken as 10, 2, and 1 for the three age groups of <2, 2–16, and >16 years, respectively (Shen et al., 2014). *IR* were calculated based on oxygen consumption associated with energy expenditures for different age groups and gender, which can be found in Shen et al. (2014).

To quantify the effect of provincial trade on human inhalation exposure to BaP, the fraction of ILCR from the TRADE run to that from NO_TRADE run was estimated by $[(ILCR_{TRADE} - ILCR_{NO\_TRADE}) / ILCR_{TRADE}] \times 100$, where $ILCR_{TRADE}$ and $ILCR_{NO\_TRADE}$ were the ILCR calculated under the TRADE scenario and NO_TRADE scenario, respectively.

3. Results

3.1 Virtual transfer of PAH emissions via interprovincial trade

Fig. 1 shows a comparison between production-based and consumption-based PAH emissions in 30 Chinese provinces derived from the MRIO analysis in 2007 and 2012. There are approximately 37.8% and 38.0% of total national PAH emissions (including

all PAH emission sources, e.g. residential, deforestation/wildfire) were emitted via the production of goods and services in 2007 and 2012, respectively. Total production-based emissions from all sectors of 30 Chinese provinces were 40120 and 30851 tons, of total PAH emissions, of which 27886 and 29748 tons were emitted from the production of goods and services ultimately consumed in different provinces in 2007 and 2012, accounting for 69.5% and 96.4% of total production-based emissions, respectively.

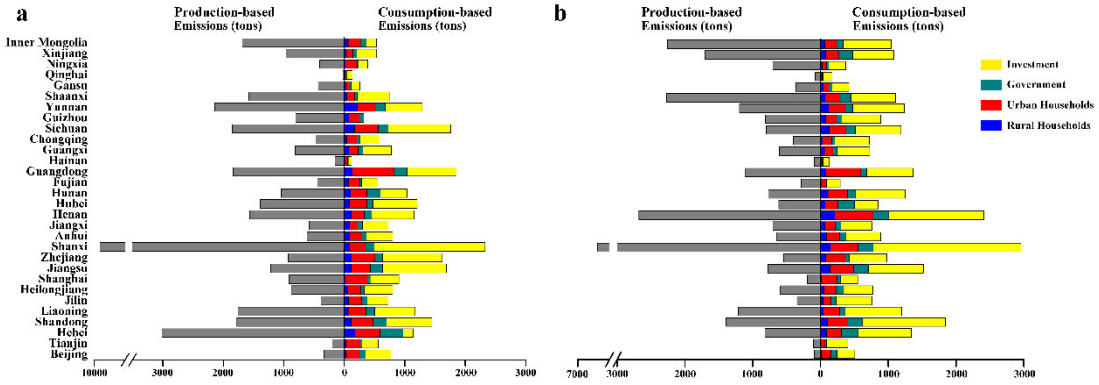

Fig. 1. Comparison between production-based and consumption-based PAH emissions (tons) in 30 Chinese provinces in 2007 (**a**) and 2012 (**b**).

Among 30 provinces, the highest production induced PAH emissions (9911 tons in 2007 and 6691 tons in 2012) occurred in Shanxi, and were approximately three times higher than those in the second PAH emissions province (Hebei Province (3016 tons) in 2007, Henan Province (2677 tons) in 2012). In 2007, higher consumption-based PAH emissions were found in well-developed Chinese provinces: Guangdong (1854 tons), Jiangsu (1691 tons), and Zhejiang (1618 tons). With an area of less than 2% of the total area of China, the Yangtze River Delta (YRD, including Jiangsu and Zhejiang provinces, and Shanghai) in Eastern China accounted for 15.1% of total consumption-based PAH

emissions. Among 30 emission sectors, coking is the most important source of PAH emissions (**Fig. S3**). Since simple beehive coke oven (i.e. indigenous coke) production activities were banned and almost ceased by 2011 in China and replaced by large-scale regular coke ovens (Xu et al., 2018; China Coal Industry Yearbooks, http://www.stats.gov.cn/tjsj/). The latter reduces the PAH emission factors by several orders of magnitude compared with beehive coke ovens (**Table S3**). As a result, the production-based PAH emissions reduced in 2012 as compared to that in 2007. From the perspective of final consumption, investment is the dominant engine driving consumption-based PAH emissions in 26 provinces (except for Hebei, Inner Mongolia, Guizhou, and Ningxia) in 2007, contributing approximately 59.8% (IR, 56.1%-68.3% interquartile range) to total consumption-based PAH emissions. In 2012, the proportion of consumption-based PAH emissions due to investment was 54.5% (IR, 46.7%-60.7%) in 30 provinces. Urban household consumption was the second largest source of consumption-based PAH emissions, accounting approximately for 24.8% (IR, 20.7%-33.9%) in 2007 and 19.5% (IR, 16.8%-22.5%) in 2012 of the total consumption induced emissions, respectively.

The difference between production-based and consumption-based PAH emissions indicates virtual emissions transferred via trade. For provinces where services and light industries are major industrial components, consumption-based emissions were greater than production-based emissions because the former were largely dependent on products or energy from provinces with higher density of PAH emitting industry, specifically, provinces characterized by energy, heavy industry, and materials

manufacturing (Zhao et al., 2015). **Fig. 1** shows consumption-based PAH emissions in well-developed provinces and municipalities (Beijing, Tianjin, Shanghai, and Chongqing) in Eastern and Southern China overwhelmed or were similar to production-based emissions in 2007 and 2012. Conversely, in those provinces characterized by the export of energy, mineral resources, and heavy industrial products, production-induced PAH emissions were higher than consumption-induced emissions. Comparing **Fig. 1a** with **1b** we can also identify a switching from outflow to inflow in some provinces from 2007 to 2012, such as Hebei, which was attributable to the changes in their industrial structure, such as the reduction of investment in the coking industry. In fact, the proportion of GDP in the secondary industry in Hebei Province declined but increased in the tertiary industry from 2007 to 2012 (Hebei Economic Yearbooks (http://tjj.hebei.gov.cn/hetj/tjsj/jjnj/)). During this period, the proportion of coal-fired power generation in energy production and consumption also decreased.

The production-based and consumption-based BaP emissions in 30 Chinese provinces in 2007 and 2012 are illustrated in **Fig. S4**. As expected, the provincial distribution of BaP emissions is almost identical to 16 PAH emission as shown in **Fig. 1**. BaP production-based emissions occurred mostly in central and western provinces in 2012, such as Shanxi, Henan, Shaanxi, and Inner Mongolia. Compared with 2007, total PAH and BaP emissions in 2012 decreased in almost entire eastern and central China and increased in western China, especially in Inner Mongolia, Xinjiang, Shaanxi, and Yunnan. This shift could be attributed to the relocation of heavy and energy industries from well-developed eastern China to less developed western China due to more strict

402 regulation for the air emission reduction in the east and abundant energy and mining
403 resources in the west (Ling et al., 2019).

404 **Fig. 2** illustrates net PAH emission transfers among 30 provinces embodied in
405 Chinese interprovincial trade in 2007 (**Fig. 2a**) and 2012 (**Fig. 2b**). Nine of thirty
406 provinces in 2007 are net emission exporters, among which Shanxi had a net outflow
407 of 3743 tons of PAH emissions, followed by Hebei (851 tons), Inner Mongolia (761
408 tons), and Shaanxi (487 tons). In 2012, the number of net emission exporting provinces
409 reduced to seven among which Shanxi was still the largest PAH emission exporter with
410 a net outflow of 3527 tons whereas Shaanxi (1123 tons) became the second largest
411 emission exporter, followed by Inner Mongolia (892 tons). Most of these provinces are
412 less developed with low populations and characterized by heavy and energy industries.
413 Their industrial and trade activities provide industrial products to meet the demand of
414 other regions, particularly for well-developed provinces and regions with strong PAH
415 virtual emission inflows in 2007 and 2012, respectively, including Zhejiang (1097 tons
416 in 2007 and 557 tons in 2012), Jiangsu (972 and 845 tons), Guangdong (934 and 815
417 tons), and Beijing (546 and 425 tons). These well-developed provinces and
418 metropolises behave similarly to trade related PAH importers with greater emission
419 inflows, often to Shanxi, Hebei, and Shaanxi Provinces. Overall, these results confirm
420 higher consumption-based PAH emissions in provinces located in the Eastern and
421 Southern seaboards of China, and higher production-based PAH emissions in inland
422 provinces, particularly those in Central and Western China. The spatial distribution of
423 total PAH emissions and emissions from industry sources in 2007 are shown in **Fig. S5**,

collected from PKU-PAH inventory (Shen et al., 2013). Higher PAH emissions can be observed in Shanxi Province, suggesting that industry emissions, especially the emissions from coking industry, have made a considerable contribution to total PAH emissions. We further estimated the emission fraction of total PAH emissions compared with emissions from industrial sources (**Fig. S5c**). As shown, large emission fraction values occur in Western China, suggesting that PAH industrial emissions dominated in these western provinces, characterized by abundant energy and mineral resources. The spatial distribution of total and industrial BaP emission is shown **Fig. S6**, which, again, exhibits the spatial similarities with total PAH emission as shown in **Fig. S5**.

The demand-driven emission fluxes can be categorized into two major types based on economic strength and emission outflow and inflow. The dominant emission transfer pattern is the emission flows from less developed provinces with abundant energy and mineral resources to provinces with abundant capital or light industry. As seen from **Fig. 2**, as a typical case, the largest trade driven PAH emission flux exported from Shanxi to Zhejiang with the relocation of 399 tons of PAH emissions in 2007, followed by Shanxi to Hebei (373 tons), and Inner Mongolia to Jiangsu (80 tons). Higher PAH emission flows are from provinces with heavy industry to provinces with abundant capital, such as Hebei, which is characterized by the largest steel industry, with outflows from Hebei to Zhejiang (194 tons), and from Hebei to Beijing (115 tons). Overall, the largest emission outflow was from Shanxi, which accounted for 21.7% of total Chinese PAH emission outflows. Among the provinces with the richest coal reserves, Shanxi had the largest number of beehive coke ovens and the highest PAH emission densities

from 1982 to 2010 (Xu et al., 2018). **Fig. 2** suggests that the industries with high PAH emissions tend to be transferred to the provinces in northwestern China from 2007 to 2012 and this part of China has become a main PAH emission exporter.

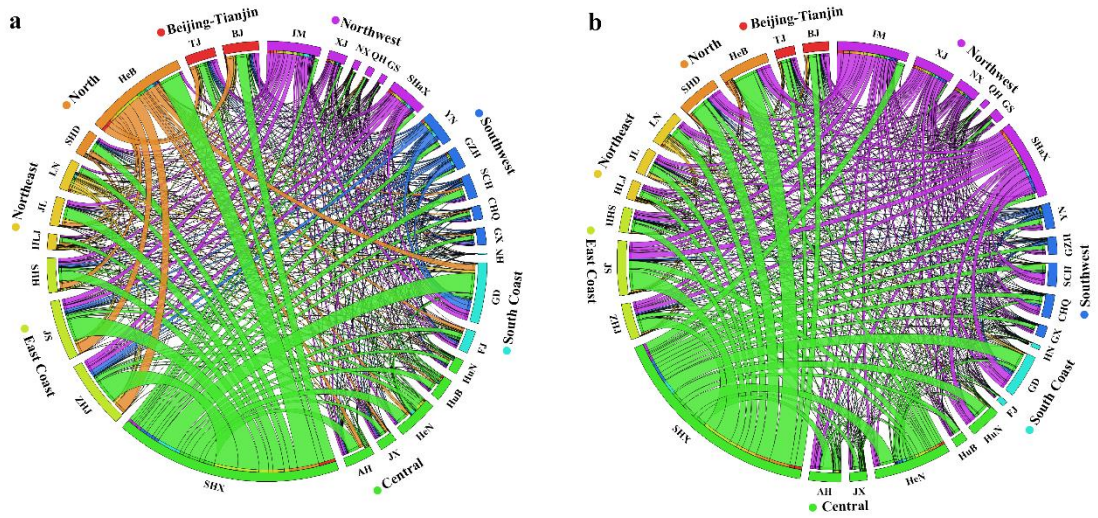

Fig. 2. PAH emission transfers attributable to interprovincial trade within China in 2007 (**a**) and 2012 (**b**). Note: The graph is drawn by the Circos procedure (http://mkweb.bcgsc.ca/tableviewer/visualize/). The 30 outer circular arcs represent 30 provinces and the 8 colors of outer circular arcs represent 8 regions aggregated from 30 provinces (**Table S4**). The length of each circular arc indicates the magnitude of PAH emissions transfer in this province. The strips connected with outer circular arcs represent PAH emissions outflows embodied in this province transfer to other provinces and the width of the strips represent the magnitude of PAH emissions transfer.

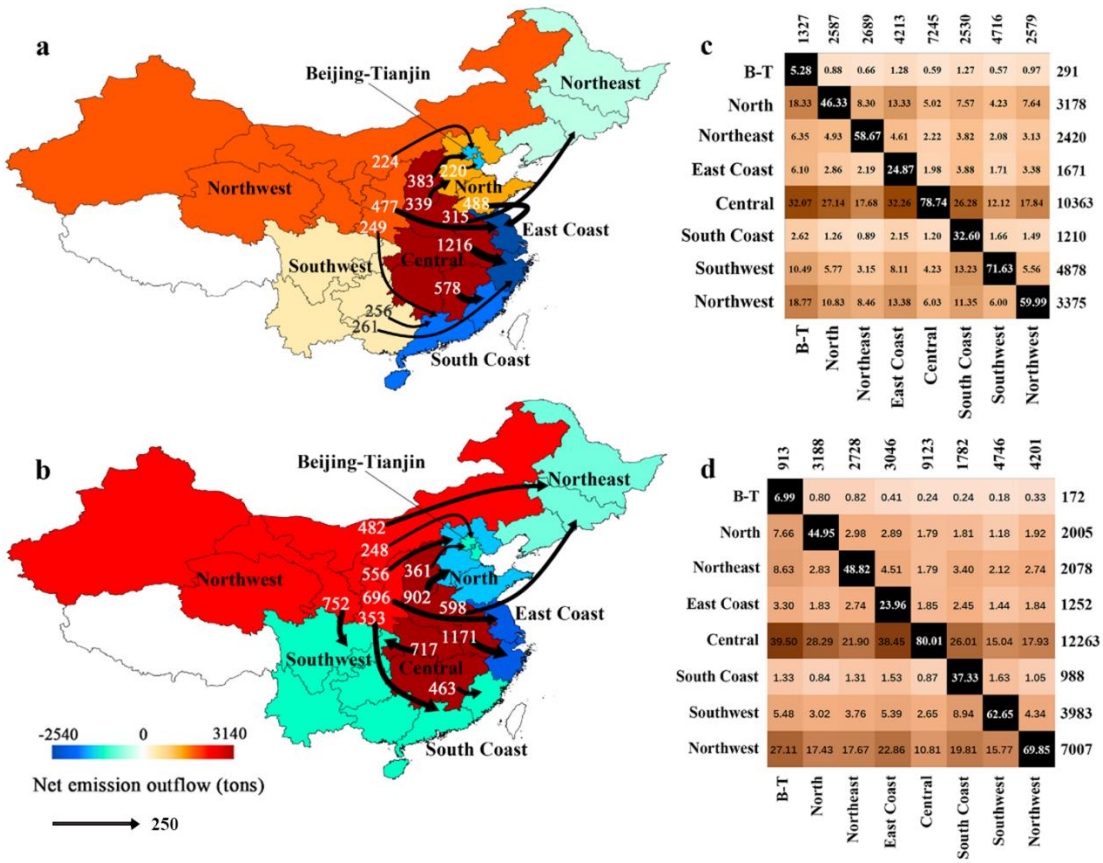

Fig. 3. Left panel shows the largest net interregional fluxes of PAH emission embodied in regional trade in 2007 (**a**) and 2012 (**b**). The red color indicates the regions with net PAH outflows and the blue color for net PAH inflows attributable to interregional trade. Arrows represent net emission flows in interregional trade and numbers indicate flows. The block charts for PAH emission transfer on the right panel (**c** and **d**) show the proportion of PAH emissions in an area where the emissions could be caused by goods and services consumed in this area and other regions in 2007 (**c**) and 2012 (**d**). The dark cell in the diagonal displays the proportion of PAH air release from goods and services produced and consumed locally. The values on the top axis are total PAH emissions (tons) in each region caused by goods and services consumed in that region and other regions. The values on the right axis are total PAH emissions caused by goods and services consumed in each region.

Fig. 3a and **3b** illustrate virtual flows of PAH emissions attributable to interregional trade in 2007 and 2012, respectively. In general, one can observe an eastward transport pattern for virtual PAH emission fluxes, manifesting as product-induced emissions from energy and resource abundant Western and Central China with heavy industries to the Eastern seaboard to meet the demands of the well-developed coastal regions. **Fig. 3c** and **3d** provide more details about the relationships among emission flows in the eight

regions in 2007 and 2012, showing the proportion of PAH emissions in each single region compared to total PAH emissions over the eight regions due to each region's final demand for goods and services. As seen, Central and Northwestern China produced more PAH emissions to meet final demand from other regions. For example, PAH emissions in Central and Northwestern China accounted for 32.1% and 18.8% of total PAH emissions, respectively, in the Beijing-Tianjin metropolises in 2007 due to final demand for goods and services in the two megacities. As a result, increasing interprovincial trade seemed to promote virtual PAH emission transfers from well-developed coastal and urban agglomerations to less-developed inland regions in Central and Western China. Comparing the virtual emission flows between 2007 and 2012 one can identify some differences in the flow pathways and magnitudes, which is again attributed to the changes in the industrial structures and demands to the products with high PAH emissions in China.

Figure S3 further illustrates provincial PAH emissions from 8 emission sectors (**Table S1**) embodied in interprovincial trade in 2007 and 2012. In the majority of provinces with abundant energy and mineral resources, the PAH emissions embodied in the outflow of goods and services were overwhelmed by coking and heavy industry. Especially, in Shanxi, Hebei, Henan, and provinces in northwestern China, except for Qinghai Province, which has the smallest volume of economic activity in China, the PAH emissions embodied in the outflow of goods and services from coking and heavy industry contributed more than 75% of total PAH emission outflow (**Fig. S3b** and **3d**). In well-developed provinces and regions of eastern and southern China, the PAH

emissions embodied in the outflow of goods and services were mainly from traffic and transport. For example, in Beijing, Shanghai, Hainan, and Fujian, the PAH emissions embodied in the outflow of good and services from traffic and transport accounted more than 80% of total PAH emissions outflow. In contrast, in well-developed provinces and regions of eastern and southern China, the inflow of PAH emissions from the coking and heavy industry far outstripped outflow of PAH emissions.

3.2 BaP atmospheric contamination induced by trade

BaP was selected as a representative PAH congener due to its strong carcinogenic nature in our modeling investigation of PAH environmental contamination and risk exposure. The CanMETOP model was employed to simulate Chinese BaP atmospheric concentrations using two model scenarios in 2007 by inputting BaP emissions with and without trade included (**Table S5**). **Fig. S7** displays modeled 2007 annually averaged ambient BaP particle and gas phase concentrations from the TRADE scenario at 1.5 m height above the ground surface using the PKU-PAH (BaP) inventory. Higher BaP concentrations have been found in Central and Southwestern China, primarily in Shanxi, Henan, Hebei, Sichuan, Guizhou, and Hunan Provinces, indicating strong industrial activity in these provinces, resulting in higher BaP emissions (**Fig. S7**). **Fig. S8b** illustrates modeled annually averaged BaP air concentrations estimated using NO_TRADE scenario runs, which exhibit a similar spatial pattern to that of the TRADE run (**Fig. S8a**), indicating that export and import of industrial products are linked with industrial activities. It should be noted that NO_TRADE scenario simulated BaP concentrations were higher in Eastern and Southern China than those estimated using

the TRADE run. This suggests that, if BaP emissions attributable to the interprovincial trade were taken into account, trade related BaP virtual emissions in those regions, mostly in well-developed provinces with lower BaP emissions, would increase due to the demands of Eastern and Southern China for goods and services imported from less-developed regions. This can be seen more clearly in **Fig. S8c** which compares the fraction of BaP air concentration from the TRADE run to that of the NO_TRADE run. Negative fractions can be identified in Eastern and Southern China, and positive values are seen in Central and Western China, except for Qinghai Province, which has the smallest volume of economic activity in China and more depends on imports from other regions.

Generally, similar to the emission difference as mentioned previously, a positive difference for ΔC_S $\left(C_{TRADE} - C_{NO\_TRADE}\right)$ shows a net export of PAH and a negative value indicates a net import of PAH. Our results clearly show that well-developed Eastern and Southern China are PAH export regions, their export being induced by their demand for goods and services from Central and Western China, and vice versa.

3.3 Lung cancer risk to BaP embodied in interprovincial trade

Fig. 4 shows that the spatial patterns of ILCR in 2007 from the two model scenarios do not differ significantly, but the NO_TRADE run yielded somewhat higher ILCR in Eastern and Southern China. Higher population-weighted ILCR can be discerned in Henan Province (**Fig. S9**) with the highest population in China, followed by populated megalopolises, including Beijing and Tianjin (Eastern China), Guangzhou and Fuzhou in Southeastern China, Shenyang and Harbin in Northeastern China, and cities in the

Sichuan Basin. The higher ILCR in populated regions and cities is attributed to spatial superimposition of the high cancer risk and high population density, which enhances human health risk due to inhalation exposure to BaP. The spatially averaged ILCRs for lung cancer risk caused by Chinese ambient BaP, estimated from the TRADE and NO_TRADE runs, were 6.29×10^{-6} (IR, 1.22×10^{-8}-1.41×10^{-6} interquartile range) and 8.49×10^{-6} (IR, 1.19×10^{-8} - 1.64×10^{-6}) in 2007, respectively. Comparing the modeling results with actual Chinese production-based emissions (TRADE scenario) suggests that interprovincial trade reduced ILCR by 2.2×10^{-6} (approximately 26%) (**Fig. 4c**), which avoided 1502 (CI95: 818-2187) lung cancer cases in 2007 (**Fig. S10a**). The coastal regions and megacities gained the greatest health benefits from interprovincial trade, including Beijing with a reduction of 7.2×10^{-5} in ILCR (**Fig. 4c**) and 94 avoided lung cancer cases (**Fig. S10a**), the YRD with a reduction of 3.5×10^{-4} in ILCR (**Fig. 4c**) and 884 avoided lung cancer cases (**Fig. S10a**), and the Pearl River Delta (PRD) with a reduction of 9.5×10^{-6} in ILCR (**Fig. 4c**) and 310 avoided lung cancer cases in 2007 (**Fig. S10a**). The spatial patterns of ILCR from two model scenarios in 2012 are displayed in **Fig. S11**. The spatially averaged ILCRs estimated from the TRADE and NO_TRADE runs in China are 4.56×10^{-6} (IR, $3.35. \times 10^{-8}$-1.62×10^{-6}) and 4.94×10^{-6} (IR, 2.61×10^{-8}-1.38×10^{-6}), respectively. Overall, the interprovincial trade reduced ILCR by 0.38×10^{-6} (**Fig. S11c**), which avoided 486 (CI95: 287-686) lung cancer cases in 2012 in China (**Fig. S10b**). Due to the reduction in PAH emission, the lung cancer risk due to exposure to PAH in China decreased from 2007 to 2012. During both 2007 and 2012, the extra lung

565 cancer cases were identified in northwestern China and the avoided lung cancer cases
566 were found mostly in the Eastern seaboard regions of China. This is because
567 production-induced BaP emissions occurred mostly in energy and mineral resource
568 abundant Western and Central China, where there is a relatively low population density
569 compared to the Eastern and Southern seaboards of China where consumption induced
570 emissions dominated (**Fig. 1**). On the other hand, BaP contamination and health burdens
571 in Central and Western China might be, to some extent, deteriorated by increasing BaP
572 emissions via exported goods and services to Eastern and Southern China due to the
573 goods and services demands from these well-developed regions. For example, Shanxi
574 and Hebei Provinces had 49 and 32 extra lung cancer cases attributable to
575 interprovincial trade, respectively, where high emission intensive goods and industrial
576 products were manufactured to meet the demands of eastern provinces or exported raw
577 materials used to produce final demanded goods. To more clearly discern the effect of
578 provincial trade on inhalation exposure to BaP, the fraction of ILCR from the TRADE
579 run compared to that from NO_TRADE run was calculated (**Fig. 4d**). In agreement
580 with the fractions of modeled BaP concentrations from the TRADE compared to those
581 from the NO_TRADE run, negative fractions are seen in the well-developed Eastern
582 and Southern seaboard, indicating that these regions benefited from interprovincial
583 trade by importing, rather than producing, goods and services from less-developed
584 provinces with rich resources and heavy industries, which reduced virtual cancer risk.
585 Positive fractions can be seen in less-developed Central and Western Chinese provinces
586 (except for Qinghai), particularly the energy and mineral abundant provinces who

exported their products to Eastern and Southern China to meet demand from this well-developed region, enhancing the health risk attributable to BaP in the atmosphere. This result demonstrates that flows towards Western and Central China with rich natural resources and low population have potentially improved energy and mineral resource utilization efficiency; as a result, they have mitigated adverse health impacts from ambient PAHs for China as a whole. However, a huge health burden was imposed on Western and Central China, and weak production technology and underdeveloped medical conditions in Western and Central China exacerbated the negative impact on western and central China.

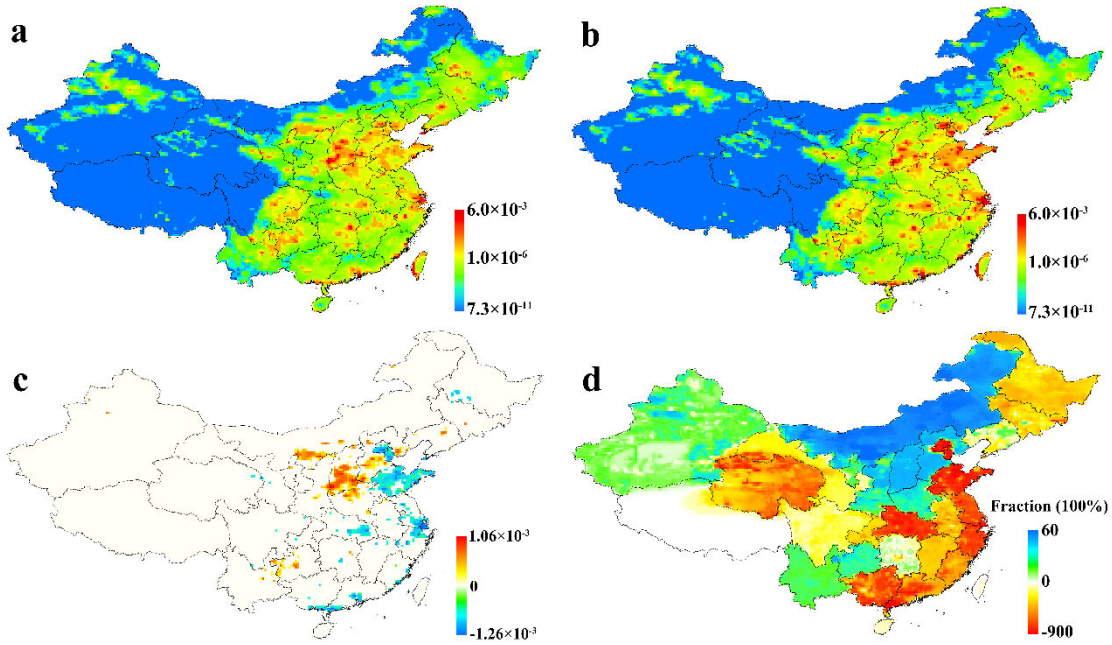

Fig. 4 (**a**) Modeled gridded incremental lifetime cancer risk (ILCR, Unitless) due to human exposure to ambient BaP concentrations from the TRADE run; (**b**) same as **Fig. 4a** but for the NO_TRADE run; (**c**) differences in ILCR between the TRADE and NO_TRADE runs; (**d**) fraction of ILCR from the TRADE run compared to that from NO_TRADE run, estimated by $[(ILCR_{TRADE} - ILCR_{NO\_TRADE})/ILCR_{TRADE}]\times100$.

Fig. S12 shows the cumulative frequency of log-transformed ILCR (logILCR)

604 caused by simulated ambient BaP concentrations with (TRADE run) and without
605 (NO_TRADE run) interprovincial trade. The logILCR exhibits a bimodal distribution.
606 It should be note that 2007 populations with excess lung cancer risk frequencies greater
607 than 10^{-5} and 10^{-4} caused by BaP industry emissions were reduced from 10.68% and
608 1.48% to 9.5% and 1.12% due to the interprovincial trade. In other words, with the
609 trade, the lung cancer risk for the Chinese population to BaP declined by 11% and 24%,
610 respectively, confirming that overall public health due to human exposure to PAHs can
611 be improved via interprovincial trade. However, for the less developed Shanxi Province,
612 lung cancer cases calculated from the TRADE and NO_TRADE runs were 142 (CI95,
613 67-217) and 93 (CI95, 45-140), indicating that ILCR attributable to interprovincial
614 trade increased approximately 35%. Knowing that those less developed but energy and
615 mineral resource abundant provinces provided industrial products and goods to well-
616 developed provinces in Eastern and Southern China, the net health benefits from PAHs
617 emissions in Eastern and Southern seaboard of China probably were at the cost of
618 negative environmental and health impacts in less-developed provinces and regions,
619 potentially resulting in environmental inequity in less-developed areas (Guo et al., 2012;
620 Zhang et al., 2018).

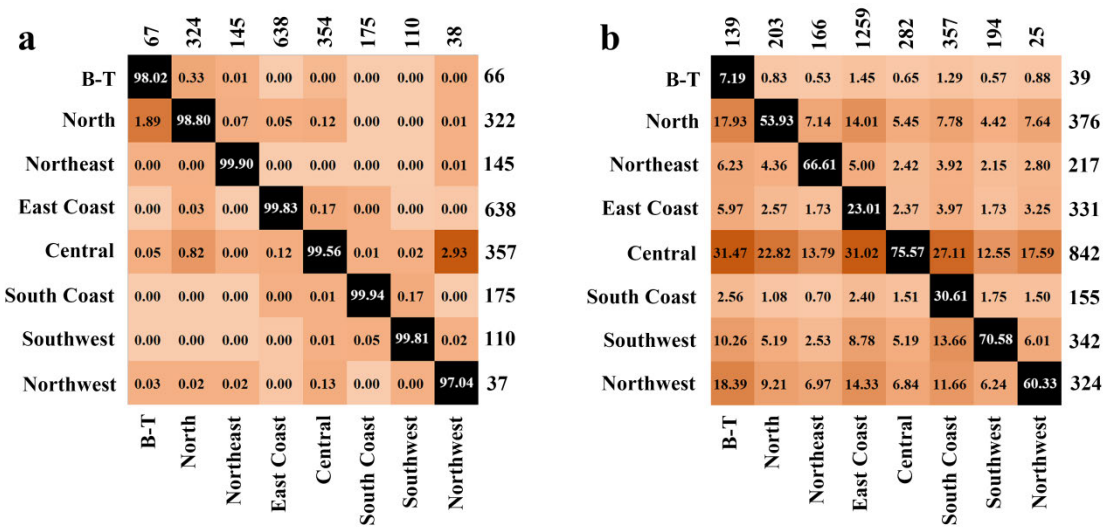

Fig. 5. Same as **Fig. 3c** and **3d** but for proportion of lung cancer incidents due to human exposure to BaP associated with atmospheric transport (**a**), and with interregional trade (**b**) in 2007. Values in each block in the two charts are the fractions of lung cancer incidents (%) in the regions denoted by the bottom axis, attributable to BaP atmospheric transport (**a**); and to goods and services (**b**). The number of total lung cancer incidents corresponding to the region denoted in the bottom axis is indicted in the top axis; and the number of lung cancer incidents caused directly due to BaP emitted in each region (**a**) or indirectly emitted from consumption of products in each region, produced in that region or other regions (**b**), is listed in the right axis.

Fig. 5a shows the percentage of lung cancer cases in each region due to BaP emissions in that and other regions in 2007. These results indicate that lung cancer cases related with atmospheric transport were mainly induced by neighboring regions. For example, BaP emissions from North China contributed to 1.89% of lung cancer cases in Beijing-Tianjin. **Fig. 5b** illustrates the percentage of lung cancer cases in each region due to consumption-based BaP emissions in that and other regions. The share of lung cancer cases in a given region related to goods and services consumed in other regions varies from 0.53% in the less-developed Northwestern China to 31.5% in the developed region of Beijing-Tianjin. Transboundary health risks related to interregional trade are much greater than those induced by atmospheric transport. In contrast to the physical transport of BaP in the atmosphere, interregional trade exerts a great influence on

inhalation exposure risks in regions where production of BaP emissions occurs far from where goods and services are finally consumed. In other words, interregional trade provides a more direct and efficient pathway for BaP and their health risks, as can be seen in **Fig. 6** illustrating the virtual flow of the number of avoided and extra lung cancer cases embodied in interprovincial trade across China. **Fig. 5b** shows that the well-developed eastern regions get the greatest health benefits from interregional trade. On the other hand, health burdens were exacerbated by less developed inland regions. For example, the final consumption for goods and services in Beijing-Tianjin, the east coast, and the south coast contributed to 31.5%, 31%, and 27.1% of lung cancer cases induced by exposure to BaP in Central China, respectively. Thus, unlike atmospheric transport, which is driven by physicochemical and environmental factors, domestic trade relocates PAH emissions in a different way. The demand and consumption of goods and services also contributes to the risk of exposure to PAHs.

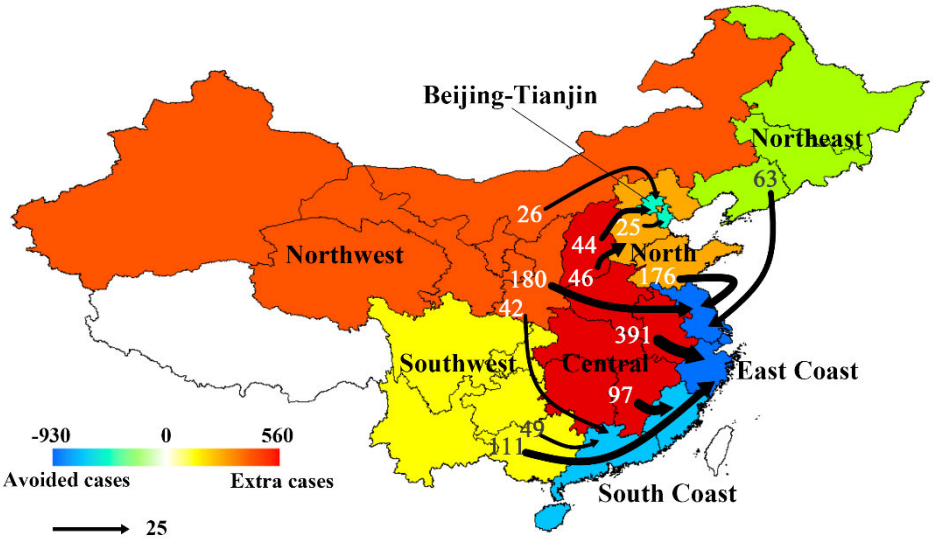

Fig. 6. Geographical distribution and primary virtual flow of avoided and extra lung cancer cases embodied in interprovincial trade.

4. Discussion

In general, in the interprovincial energy and mineral resources trade, well-developed eastern China is located in the downstream of supply chains whereas central and western China is located at the top of these supply chains. The latter plays a leading role in a "transmission channel" of energy and mineral resources products from energy and resource abundant central and western China to eastern China, which yielded a large surplus of PAH emissions in northwestern and central China. Accordingly, BaP contamination and health burdens in central and western China increased due to the goods and services demands from these well-developed regions. It is expected that such supply chains and "transmission channel" would not be altered in forthcoming years. To retard PAH contamination and human exposure to PAHs, those provinces or regions with higher PAH emissions, such as Shaanxi, Hebei, Yunnan, Inner Mongolia, and other energy and resources abundant provinces in central and western China, should have aggressively reduce local PAH emissions according to their respective demands to goods and services. However, if a large portion of PAH emissions from those inland provinces would be attributed to the demands of the good and service products from well-developed eastern and southern China, the PAH emissions have to be maintained on a high level in the inland provinces. As aforementioned, before 2011, coke production was one of the major industrial PAH emission sources and contributed significantly to PAHs pollution in central China. With the replacement of beehive coke ovens by large-scale regular coke ovens subject to the Coal Law in China (Xu et a., 2017), PAH emissions in Shanxi province, central China, which outflowed the largest

682 PAHs to east China, was reduced markedly, as seen in **Table S2**, **Figs. S3** and **S4**. The
683 ban of beehive coke in China also benefited significantly the 62% decline in cumulative
684 non-occupational excess lung cancer cases from 1982 to 2015 in China, particularly in
685 Shanxi province (Xu et al., 2017). This suggests that official activities could play a
686 crucial role in the production-based emission abatement in those less developed
687 provinces. In this sense, our results raised an issue for the environmental equity between
688 consumers and producers of goods and services. Concerns for the environmental equity
689 and energy justice were just recently raised (Ling et al., 2019, Wang et al., 2019).
690 Measures and strategies have been proposed to reduce the environmental inequity and
691 energy injustice hidden in China's inter-provincial and inter-regional trade, including
692 the enhancement of the prices of energy and raw material products, accounting for
693 pollution cost-benefit approach in entire supply chain rather than direct PAH emission
694 only, and promoting subsidies from central government and provinces located in the
695 downstream of the supply chains to those less developed provinces at the top of supply
696 chains (Xu, et al., 2017; Ling et al., 2019; and Wang et al., 2019). In addition to fossil
697 energy, northern and northwestern China are also the regions with most abundant
698 renewable and clean energy resources in China, such as solar energy and wind energy
699 (Ling et al., 2019). Applications of these renewable energy resources should be
700 promoted and supported continuously and increasingly in these provinces by all level
701 government agencies.

702 Our emissions and CanMETOP model are subject to uncertainties from different
703 sources. These uncertainties might come from emission factors, errors in emission

sources, economic data in input/output table (Chen et al., 2016). Usually, the largest uncertainty always was traced back to emission inventories. PAHs examined in this study are emitted from the same types of sources reported by Shen et al. (2013) who estimated the uncertainties of PAH emission in developing countries ranging from 34% to 61% in 2007. The uncertainties in CanMETOP primarily come from emission input and processes in air, soil, and water, but were very difficult assessed. It has been estimated that model uncertainties were approximately 20%-25% (Huang et al., 2016), which was lower than an atmospheric chemistry model (Lin et al., 2014). The raw data used in Chinese provincial MRIO model are derived primarily from official statistics. Due to statistical error, the MRIO tables themselves have inherent uncertainties. Zhang et al. (2017) added a 13% uncertainty of production-based emission due to the uncertainty related to MRIO itself in their investigation to transboundary health impacts of air pollution induced by international trade.

5. Conclusion

Aiming at filling knowledge gaps in the effects of goods and services trade on PAH emissions, environmental contamination, and exposure risks, the present study constructed PAH emissions embodied in the interprovincial trade. A coupled atmospheric transport, multimedia exchange, and lung cancer risk model was employed to assess quantitatively the environmental fate and lung cancer risks induced by the trade-related PAH relocation. Our results revealed that interprovincial trade played a more important role in PAH environmental fate than the atmospheric transport of PAH,

indicating that trade is an important pathway for PAH relocation and transport. We found that interprovincial trade reduced overall cancer risk in Chinese population because the import of goods and service in well-developed Eastern China from less-developed Western and Central China decreased PAH emissions in populated Eastern China, but increased PAH emissions in less-populated Western and Central China, overall reducing PAH inhalation exposure risk in entire China. It should be noted that here we only simulated the atmospheric transport and lung cancer risk of BaP embodied in interprovincial trade. If all 16 PAH congeners were taken into account in the cancer risk assessment, we would anticipate higher lung cancer cases associated with PAH interprovincial trade. The dissimilarity of nature resources determines largely interprovincial trade of goods and services between well-developed and less developed eastern and western China, which might exacerbate the burden on the environment and human health between these two parts of China. Our results could help policy-makers to implement proper measures to control or mitigate PAH emissions in China. The virtual PAH flow and relocation associated with the interprovincial trade obtained in this study could also help to implement regional cooperative programs in both well-developed eastern China and less-developed western China to take their individual responsibilities in PAH emissions and risk reduction.

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Notes
The authors declare no competing financial interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at the JCLP website.

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