The Slowdown in Global Air Pollutant Emissions and Driving Factors

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Summary

Fine particles ($PM_{2.5}$) causes millions of deaths and has attracted increasing attention in recent years. The socioeconomic drivers of the dynamic global emissions are not fully understood. We quantify the driving factors of primary particle emissions and oxidized precursor emissions from 2004-2011. The results indicate that the global growth rates have slowed because of improvements in energy intensities and production efficiency; however, different types of air pollutants show uneven regional changes. Although the emissions in Asia are driven by the increasing demand for certain products, improvements in the emission intensity of coal represent the main factor offsetting SO_2 and NO_x emissions, whereas emission controls implemented for industrial processes have largely contributed to reducing primary $PM_{2.5}$ emissions. The net emissions embodied in East Asia's exports to developed countries declined while that to developing countries increased rapidly. The analysis creates opportunities to involve both producers and consumers in co-mitigation of various pollutants.

Introduction

Fine particulate matter pollution ($PM_{2.5}$, i.e., particulate with a diameter less than 2.5 micrometres) that consists of primary particle emissions and those generated through the

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oxidation of precursor emissions (e.g., sulphur dioxide (SO_2) and nitrogen oxides (NO_x)) is considered a threat to human health[1], the environment[2] and the global climate[3,4]. In 2010, $PM_{2.5}$ pollution was associated with 3.3 million premature deaths worldwide, predominantly in Asia[5,6]. Over the past 50 years, $PM_{2.5}$ emissions generated from developed countries have decreased dramatically because of the implementation of a variety of environmental measures[7,8], whereas in developing countries, such as China and India, $PM_{2.5}$ emission levels have increased substantially because of the rapid expansion of coal-based industries[9,10]. Increased emissions in emerging markets have been driven by economic growth and rapid industrialization. However, few efforts have been made to quantify the contributions of the various socioeconomic drivers to the increase in $PM_{2.5}$ emissions[11].

Compared with long-lived CO_2 emissions (which can remain in the air for hundreds of years after emission), $PM_{2.5}$ is short lived and it presents an average lifetime about 1-2weeks [12]. Therefore, the adverse environmental and health impacts of $PM_{2.5}$ are greatest within emission-producing regions and in nearby downwind regions, and reducing such emissions could directly mitigate the related effects without a time lag. Thus, identifying the driving factors underlying $PM_{2.5}$ emissions could immediately contribute to addressing air pollution and related health issues. Such a scenario is not the case with CO_2 pollution because cumulative CO_2 emissions from the preindustrial era have led to global warming, and current measures to decarbonize CO_2 emissions can only slow down the increases in CO_2 accumulation and associated warming effects.

In addition, considerable $PM_{2.5}$ emissions in emerging economies have been linked to consumers worldwide [13-15]. These transboundary air pollutants are associated with shifting air pollution and health impacts and the climate forcing of aerosols[16-18]. Thus, quantifying the underlying socioeconomic factors that affect these change of emissions (driven by both local and foreign consumption) throughout the global supply chain is crucial for targeting clean-up efforts to reduce global and regional $PM_{2.5}$ emissions and alleviate trade-related climate and health impacts[16].

$PM_{2.5}$ pollution has both primary and secondary sources[19], and both sources significantly contribute to regional air quality issues. The former mainly include primary particulate emissions generated from coal combustion activity, diesel vehicle use, and industrial processes, whereas the latter involve the formation of aerosols from precursors, such as SO_2, NO_x[20], non-methane volatile organic compound (NMVOCs), and ammonia (NH_3). Sulphate and nitrate are large components of ambient $PM_{2.5}$[21], and they contribute significantly to the development of severe haze episodes in eastern China[22]. Anthropogenic NMVOCs are mainly released from combustion activities and industrial processes (e.g., transport, heating, the use of organic chemicals, paints, etc.)[23]. Anthropogenic NH_3 is mainly emitted from fertilizers, livestock wastes and some industrial activities [10,24]. Black carbon (BC) and organic carbon (OC) are important components of primary $PM_{2.5}$, and they are mainly emitted through the incomplete combustion of fossil fuels and biomass. BC profoundly enhances global warming by absorbing solar radiation and reducing surface albedo levels,[25] whereas OC (except for brown carbon) cools the atmosphere by scattering radiation[26]. In addition to carbonaceous aerosols, trace metals also contribute to the $PM_{2.5}$ mass and have effects on human health[27,28] and ocean fertilization[29]. Due to data availability, this study considered both primary and secondary sources of $PM_{2.5}$ pollution and did not consider trace metals.

This study aims to investigate the socioeconomic drivers of global $PM_{2.5}$ emission sources, including BC, OC, other primary $PM_{2.5}$, SO_2, NO_x, NH_3 and NMVOC emissions from industrial activities and residential household. Beginning with the newly developed inventories (see section ***Production-related air pollutant emissions inventory***), we present country- and sector-specific analyses of annual emissions from both production and consumption perspectives and quantify shifts in air pollutants that occur via international trade by using multi-regional input-output (MRIO) analysis. MRIO analysis enables to link producers and consumers worldwide and analyse the effects of trade on global and regional energy consumption[30], CO_2 emissions[31], material consumption[32], biodiversity[33], mercury emissions[34], and other environmental issues[35,36]. See section ***Multiregional input-output analysis*** for details. We quantify the contributions of the various factors to global and regional changes in emissions between 2004 and 2011 through structural decomposition analysis (SDA), a tool that has been widely used to evaluate the contributions of different drivers to overall changes in U.S. CO_2 emissions[37], global greenhouse gas emissions[38] and global energy consumption[39]. See sections ***Structural decomposition analysis*** for details.

Results

Contributions to the changes in emissions from 2004 to 2011

Air pollutant emissions in this study include those generated from sources in all industrial sectors and via residential energy consumption. Industrial sectors (see details in Table S1) include all economic activities (e.g., agricultural, manufacturing and processing sectors and services), which can be linked to the consumers in the interregional trade. Emissions from residential households (e.g., from cooking and heating) represent the direct emissions from residential energy consumption.

Figure 1 shows the contributions of ten factors to changes in industrial emissions from 2004 to 2007 and from 2007 to 2011 for five aerosol and precursor species (see results for residential emissions in Figure S2). Global emissions (except for SO_2) have continued to rise over the two studied periods despite individual and governmental mitigation efforts. However, the emission growth rates decreased for all species except NH_3 from 2007 to 2011 compared with that from 2004 to 2007.

Levels of primary species, i.e., BC, OC, and other primary $PM_{2.5}$, increased slightly more rapidly from 2004 to 2007 with growth rates of 9.0%, 7.5% and 7.7%, respectively (Figure S1). However, the growth rates of precursor species have been more modest in comparison, with values of 3.1% for NMVOCs, 3.6% for NH_3, 4.0% for SO_2 and 5.7% for NO_x. From 2007 to 2011, NH_3 was the only species presenting a higher growth rate relative to that of 2004 to 2007. The increase in estimated NH_3 levels can be attributed to increased vehicle use in developing regions[10].

Emission intensity reductions have been the main force for offsetting global air pollution emissions from both industrial activities and residential household. Instead focusing on overall changes in emission intensity, we further decompose it to show contributions of emission factor, energy mix, energy intensity and process emission intensity (i.e., production efficiency) changes. This analysis reveals that energy efficiency gains and process emission intensity decline have contributed almost all emissions reduction. With all other factors remaining constant, overall emission intensity changes curbed 10%-20% of the industrial emissions

(Figure 1 and S1) and 20%-30% residential emissions from 2004 to 2007 (Figure S2). These efforts have mainly been offset by a progressive increase in per capita consumption and population growth. From 2004 to 2007, changes in per capita consumption (dark and light blue, green) drove industrial emissions by approximately 20%. Import structures (purple) represent the share of imports in the total consumption in each sector, and their effects are largely attributed to differences in the emission intensity levels between regions. Import and consumption structures have different effects across air pollutants because of differences in emission sources. The consumption structure (yellow) change from 2004 to 2007 contributed to emissions reductions (but not for NH_3), whereas import structure changes contributed to a reduction in NMVOCs and an increase in the volume of other air pollutants.

From 2007 to 2011, the annual growth rates for all air pollutants except for NH_3 decreased largely because of emission intensity changes. The contribution of change in production efficiency and energy mix were much larger during 2007-2011 than that during 2004-2007. Per capita consumption and population changes had similar but stronger effects on emission changes during this period. However, the shifting consumption structure had the opposite effect during these two periods and led to increases in global $PM_{2.5}$, NO_x and SO_2 emission levels from 2004 to 2007 by approximately 1% and decreases in these emission levels by approximately 1% from 2007 to 2011 (yellow). Structural changes in imports (purple) had less of an impact on emission growth (except for NO_x) from 2007 to 2011. To further understand the factors that shaped these emission changes, we analysed the changes in regional emissions and quantified the contributions of various drivers.

Global drivers of regional emissions

The net emissions embodied in trade have tended to increase slowly and have even decreased in East Asia. The difference in production-based (i.e., emissions directly released within territorial boundaries) and consumption-based (emissions related to goods and services consumed in each region[31]) emissions is the net emissions embodied in trade. An increasing number of reports have indicated that emissions released by local producers can be considerably driven by global consumers[13,16,17]. However, the trend has changed slightly. From 2004 to 2007, the net emission transfer changes in East Asia were 0.34 Tg for $PM_{2.5}$, -1.1 for SO_2, -0.03 Tg for NH_3 and -0.3 for NMVOCs, and from 2007 to 2011, these values decreased substantially to 0.07 Tg for $PM_{2.5}$, -4.2 Tg for SO_2, -0.6 Tg for NH_3 and -1.6 for NMVOCs.

Figure 2 shows the contributions of different driving forces to production- and consumption-based emissions in ten aggregated regions for 2004 to 2007 and 2007 to 2011 (see the detailed results for all 129 countries and definitions of the ten regions in the supporting Table S2). In all regions, increasing consumption levels per capita have constituted the main driving force behind increases in air pollutant emission levels. East Asia experienced the largest increases in regional $PM_{2.5}$, NO_x, NH_3 and NMVOCs emissions from 2004 to 2011, which accounted for 72% ($PM_{2.5}$), 142% (NO_x), 33% (NH_3) and 82% (NMVOCs) of the global emission changes, respectively. The SO_2 emissions in East Asia increased by 2.5 Tg from 2004 to 2007 and decreased by 4.4 Tg from 2007 to 2011. However, all emissions in South Asia exhibited slight increasing trends from 2004 to 2011. Infrastructure investments served as the main driver in East and South Asia and increased regional air pollutant emission levels by 40% and 20%, respectively, from 2004 to 2011 (while other factors remained constant). From a sectoral perspective (Figure S4), demand for construction, machinery and equipment, and transport are the largest drivers for East Asia, whereas the demands for metal, food products and energy are also considerable drivers for South Asia, especially in terms of increasing SO_2 emissions. Construction and machinery and equipment are primary sectors of capital formation (i.e., investment) and characterized by the use of energy- and emissions-intensive supply chains. The services and dwellings sector is fairly emissions-intensive from a consumption perspective because demands from service sectors drive the production of emissions-intensive products (e.g., electricity).

Figure 2 also reveals the considerably negative contributions of emission intensity to all emissions, especially SO_2. The SO_2 emissions in East Asia declined from 2007-2011, which demonstrates that improvements in overall emission intensity levels (i.e.,) is an effective method of controlling emissions under rising demand. We further decompose the emission intensity to emission factor/ end-of-pipe control, energy mix and energy efficiency gains. As shown in Figure 2, improvements in the emission factor and production efficiency constituted the main mechanism offsetting the SO_2 and NO_x emissions in East Asia. Substantial primary

$PM_{2.5}$ emissions are generated from industrial processes, such as cement and lime production[40], and can be reduced by 10-99.9%[41] through the use of control devices. Emissions controls in industrial processes have resulted in a large decrease in primary $PM_{2.5}$ (including BC and OC) emissions in East Asia if other factors were constant (Figures 2 and S5). Energy mix has not greatly affected the air pollutant emissions, except that in Sub-Saharan Africa almost all emissions reduction during 2004-2007 were linked to energy efficiency gains in East Asia, which was an accelerator of emissions growth during 2007-2011. Moreover, energy intensity of coal in South Asia also slightly contributed to increased SO_2, NO_x and NMVOC emissions. East and South Asia gained fewer environmental benefits from reducing emissions from oil use, which has been an important factor underlying the reduced emissions in North America and Western Europe in recent years (Figure S5).

Import structures had less of an effect on the growth of air pollutant emissions in East Asia from 2007 to 2011 compared with the effect from 2004 to 2007. This finding indicates that the global trade structure tends to contribute to air pollutant mitigation in China. An import structure is defined by the share of imports in the total consumption in each sector. Import structure changes can influence emissions because of considerable differences in embodied emission intensity levels in different countries (up to a factor of ten)[13]. Indeed, when a country replaces domestically produced products with imported products that have higher embodied emission intensities, this change in the import structure can increase air pollutant emissions globally and vice versa[42]. Combined with emission intensity improvements, import structure changes have tended to contribute less to emission growth in East Asia (Figure 2).

Consumption structures also had a negative effect on all species with the exception of NH_3 from 2007 to 2011. Changes in the consumption structure of a country can reflect changes in the behaviours of its consumers. The per capita consumption and total population increased from 2004 to 2011 in all regions. These negative effects of consumption structures on regional or global emissions do not suggest that consumers bought fewer emission-intensive goods but rather that the use rate of less emission-intensive products increased compared with the use rate of high emission-intensive products. Considerable contributions of consumption structures to NH_3 emissions can be partly attributed to the rising demand for vehicle fleets[12,38]. Thus, these consumption patterns should be reconsidered, especially in developing countries. Most developing countries (especially China) are experiencing rapid patterns of urbanization, and the migration of rural residents to cities could lead to significant increases in energy consumption and associated emission levels as well as the demand for energy-intensive products[43]. In early phases of a transition when per capita consumption is low, consumption patterns that benefit personal health while also limiting air pollutant emissions may be used. Additionally, population growth increases air pollutant emissions, especially in Sub-Saharan Africa, and this trend is expected to continue in the future [44].

Socioeconomic factors also contribute to changes in transboundary emissions between producers and consumers. Figure 3 shows the change in transboundary primary $PM_{2.5}$, SO_2 and NO_x emissions from 2004 to 2011 caused by international trade (results for NH_3, NMVOCs, BC and OC can be found in Figures S6-S7). The results suggest that rapid increases in emissions were largely driven by domestic consumption activities, but export-related emission growth was also notable, for example, 20.4% (453 Gg) of the total emission growth in East Asia from 2004 to 2011.While another 1,777 Gg was driven by the local consumption. Moreover, the increased emissions embodied in trade in East Asia from 2004 to 2011 were overwhelmingly driven by consumption in developing countries. Nearly all net emissions embodied in exports from East Asia to Latin America and the Caribbean (LAM), the Middle East and North Africa (MNA), Southeast Asia and the Pacific (PAS), and Sub-Saharan Africa (SSA) and South Asia (SAS) increased. However, in developing regions, such as LAM, MNA, South-PAS, and SSA, the growth in emissions embodied in imports outpaced the growth in emissions embodied in exports. These production differences were generally balanced by additional production and the release of $PM_{2.5}$ emissions in East Asia (Figure 3).

Emissions outsourced from developed regions, such as North America, Western Europe and the Pacific OECD-1990 countries, to developing regions have decreased substantially (Figure 3). However, the net emission transfers from developing regions to other developing regions (EAS, LAM, MNA, SSA, SAS, and PAS) increased by 676 Gg, and these combined changes represent a virtual redistribution of emissions from developed regions to developing regions. Similar patterns are found for SO_2 and NO_x (Figure 4), BC and OC (Figure S6) and NH_3 and NMVOCs (Figure S7). As a notable change, China has made significant strides toward reducing exported SO_2 emission levels; however, the exported SO_2 emission levels of South Asia have experienced considerable growth and account for 24% of the total SO_2 increase for this period (6,053 Gg, Figure 3). Moreover, the net emission transfer (emissions embodied in exports minus emissions embodied in imports) in East Asia declined for SO_2, NH_3, $NMVOC_S$, BC and OC from 2004 to 2011.

Discussion

We find that East and South Asia were the most significant regional contributors to the increase in air pollutant emissions between 2004 and 2011. These contributions were largely driven by increasing consumption levels and partly offset by emission intensity improvements. The per capita investment change in the East Asia region was a substantial driver of global emissions between 2004 and 2011 (most emissions were released locally), as capital investment increased market demand and led to the large-scale expansion of the production of cement, lime, steel and other emission-intensive processed materials[45,46]. For example, cement production levels in China increased from 970 million tons in 2004 to 2099 million tons in 2011[47] at an annual increase of 12%. Almost all of the cement produced was consumed domestically[48]. Because of the negative effects of emission control on industrial processes, more investments and attention should be devoted to promoting the use of effective control measures in these industries to offset rising demand. Decrease of emission factor of coal use represented the main contributor to emission reductions (especially for SO_2 emissions) in East Asia from 2007 to 2011. These improvements are highly related to the incorporation of flue gas desulphurization (FGD) systems because such installations in Chinese electricity plants increased from 12% in 2005 to 82.6% in 2010[49]. Energy mix slightly contributed to the air pollutants emissions during 2004-2007 and was negligible during 2007-2011. However, the recent Chinese policy directive to cap coal at 4 billion metric tonnes requires its proportion in the energy mix to decrease from 68% in 2015 to around 55% by 2020[50], which implies energy mix will contribute further reduction in air pollution emissions in the coming years.

Improvements in emission intensity levels also contributed to a reduction of approximately 20-40% residential household emissions from 2004 to 2011 if other factors remained constant (Figure S2). Developing regions would substantially benefit from efforts made to improve household emission intensities. An estimated 3 billion people worldwide rely on the use of highly polluting traditional solid fuels for household cooking and heating purposes[51]. Solid fuels (including biomass and coal) used in developing regions for household heating and cooking have much larger emission factors than fossil fuels. Potential strategies for mitigating solid fuels include the use of cleaner stoves, such as advanced fan stoves that use pelletized biomass, and cleaner end-use fuels, such as natural gas, liquefied petroleum gas (LPG), and electricity[52].

International cooperation to address transboundary air pollutions, such as Long-Range Transboundary Air Pollution (LRTAP) Convention [53] should also include the 'leakage' of emission via international trade, especially the emission transfers between developing regions. Economic globalization has caused goods and services to flow across the world and has enhanced international trade. There is increasing evidence that reflects the effect of global trade on environment [54]. Over the past two decades, global trade has tripled from US$6.2 trillion to US$19 trillion[55]. A shift in the trade structure also occurred, and trade is of relevance to certain policy issues (e.g., emission transfers between countries through international trade). For example, consumption in other developing regions has increased air pollutant emission levels in East Asia by 10-20%, whereas consumption in developed countries has had a decreasing impact on emission levels (Figure 3). Overall, the shifts of air pollutants from developed regions to developing regions have declined, whereas the shift in emissions from Latin America, the Middle East, South Asia, Southeast Asia and the Pacific to East Asia and South Asia has increased rapidly. Along with intraregional trade, trade flows and investments among developing regions are expected to continue to grow rapidly as evidenced by the establishment of several institutional architectures, such as the South-South Cooperation Fund, the Asian Infrastructure Investment Bank and China's One Belt One Road strategy. The growing links between developing countries is likely to play a critical role in determining the magnitude and regional distribution of future global emissions [56]. China should upgrade its export mix with developing trade partners and avoid acting as a factory for steel, plate glass, cement, chemicals and other emission-intensive products to meet growing demand in developing regions.

Regional cooperation is required to accelerate emission reduction and maximize health and environment benefits. Although air pollutant emissions from developed regions (e.g., the USA) have declined, consumers in developed countries still contribute to considerable emissions and also related health impacts in developing countries (e.g., India and China[13,57]). In 2007, in particular, 21%, 32% and 36% of the top three $PM_{2.5}$ emissions sectors (mineral products (2.5 Tg), electricity (1.9 Tg) and ferrous metals (1.5 Tg), respectively) were associated with demand from foreign countries. Although China's economy-wide emission intensity levels declined steeply (approximately 30-60%) over this period, a large gap was observed when these levels were compared with the trends for developed countries (Figure S8). The emission intensity of primary $PM_{2.5}$ in China is more than 10 times that of the USA. A considerable opportunity is available to improve the emission intensity of the Chinese economy, especially for the sectors of coal mining and electricity, for which more energy efficient technologies and equipment can be employed.

Experimental Procedures

Production-related air pollutant emissions inventory

The production-based air pollutant emissions inventory (F_{Pr}) must be used to conduct an MRIO analysis. This inventory assigns emissions to locations where air pollutant emissions are physically produced. Production-based BC[58], OC[59], primary $PM_{2.5}$[60], SO_2, NO_x[9] and NH_3[10] levels are based on production activities (energy statistics or material production) and databases of currently used technologies[61] (Table S3). NMVOC emissions, which are mainly emitted from solvent use and as fugitive emissions from fuels, are ascribed to different industry sectors as shown in Table S4. The emission factors for 2011 were assumed to be equivalent to those observed throughout 2010; therefore, the emissions for 2011 were derived from the emissions for 2010 based on the activity rates (energy consumption, per capita income, etc.)[62-64].

In this study, we adopted global high-resolution PKU inventories as our primary datasets[60]. These data are based on detailed sub-national fuel consumption data when available[65]. The PKU inventories are based on a global 0.1° × 0.1° fuel combustion dataset (PKU-FUEL-2007, covering up to 78 fuel combustion processes and 223 countries/territories) and an updated emission factor dataset for different air pollutants[60,65,66]. In this study, sector- and production-based inventories of global emissions were developed for 2004, 2007 and 2011 based on 129 countries/regions (supplementary data) and 57 industry sectors (Table S1 in the SI). The method used to combine the PKU inventories and GTAP database is shown in the SI.

The air pollutant emissions considered in this study include those generated from production sectors and from non-commercial residential energy consumption. Production sectors include all economic activities (e.g., agricultural, manufacturing and processing sectors and services), which contribute approximately 60% of the primary $PM_{2.5}$ (55% of BC and 25% of OC), 96% of SO_2, 95% of NO_x, 85% of NH_3 and 70% of NMVOC emissions. Emissions from non-commercial residential households (e.g., from cooking and heating) represent direct emissions from residential energy consumption. Industrial and non-commercial residential emissions are driven by different factors and should be regulated through different measures. Changes in industrial and residential emissions are decomposed separately, and the corresponding results are shown in Figures 1, S1 and S2. Unless stated otherwise, emissions reported in the manuscript correspond to industrial emissions generated from an identifiable production sector and do not include those related to non-commercial residential emissions.

Multiregional input-output analysis

Environmental input-output analyses (EIOs)[67] have been widely used to illustrate economy-wide environmental repercussions (i.e., $PM_{2.5}$ emissions in this study) triggered by economic activities. Economic globalization entails the geographical separation of producers and consumers. This separation can redistribute energy consumption and air pollution emissions associated with the production of traded goods and services and alter global and regional $PM_{2.5}$ emission levels[13,68]. In the present study, we used the MRIO model to build global consumption-based emissions inventories for 2004, 2007 and 2011. For a global economy with N regions (in practice, most regions included in the present analysis are individual countries) and M industries, the global extended MRIO table has three main components: 1) emissions resulting from the production of goods and services, which are referred to as production-based emissions (F); 2) monetary flows of goods and services and the interdependence between suppliers and consumers along the supply chain (**Z**); and 3) the final demand for commodities (**Y**, monetary unit).

The mathematical expression of the entire economy with N producers is as follows:

$$\begin{pmatrix} \mathbf{X}^1 \\ \mathbf{X}^2 \\ \mathbf{X}^3 \\ \vdots \\ \mathbf{X}^N \end{pmatrix} = \begin{pmatrix} \mathbf{A}^{11} & \mathbf{A}^{12} & \mathbf{A}^{13} & \cdots & \mathbf{A}^{1N} \\ \mathbf{A}^{21} & \mathbf{A}^{22} & \mathbf{A}^{23} & \cdots & \mathbf{A}^{2N} \\ \mathbf{A}^{31} & \mathbf{A}^{32} & \mathbf{A}^{33} & \cdots & \mathbf{A}^{3N} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ \mathbf{A}^{N1} & \mathbf{A}^{N2} & \mathbf{A}^{N3} & \cdots & \mathbf{A}^{NN} \end{pmatrix} \begin{pmatrix} \mathbf{X}^1 \\ \mathbf{X}^2 \\ \mathbf{X}^3 \\ \vdots \\ \mathbf{X}^N \end{pmatrix} + \begin{pmatrix} \sum_s \mathbf{Y}^{1r} \\ \sum_s \mathbf{Y}^{2r} \\ \sum_s \mathbf{Y}^{3r} \\ \vdots \\ \sum_s \mathbf{Y}^{Nr} \end{pmatrix} \quad (1)$$

where X^r is a vector of the total economic output of each sector in region r, i.e., $\mathbf{X}^r = (X_1^r \quad X_2^r \quad X_3^r \quad \cdots \quad X_m^r)^T$; $\mathbf{A}^{qr}$ is the matrix of technical coefficients, in which the columns reflect inputs from industries in region q required to produce one unit of output from each sector in region r; and $\mathbf{Y}^{qr}$ is a vector of the final products supplied from each sector in region q for region r. Based on this framework, air pollutants associated with consumption in each region ($\mathbf{F}_C{}^r$) can be calculated as follows:

$$\mathbf{F}_C{}^r = \mathbf{h}(\mathbf{I} - \mathbf{A})^{-1} \times \mathbf{Y}^r \quad (2)$$

where $\mathbf{Y}^r$ is the vector $Y^r = \left(y^{1r} \quad y^{2r} \quad y^{3r} \quad \cdots \quad y^{Nr}\right)^T$, h is a vector of the emission intensity (emissions per unit of industry output) for each economic sector, I is the identity matrix, and $(\mathbf{I}-\mathbf{A})^{-1}$ is the Leontief inverse matrix. By allocating the direct and indirect emissions to the final consumer demand, we derived a global consumption-based emissions inventory for each of the three years[13]. Notably, we traced all emissions associated with consumed goods back to the original sources of emissions, even when products were transferred through other countries/regions or were intermediate constituents in a multiregional supply chain.

The MRIO model endogenously calculates the domestic output as well as the regional output resulting from the international trade of intermediate products. Trade, economic input-output (by sector), GDP, population, and energy consumption data for each region were obtained from Version 9 of the Global Trade Analysis Project (GTAP), which compiles primary data from the voluntary contributions of each region[69].

Structural decomposition analysis

A decomposition analysis was used to analyse the factors driving emission changes. Two decomposition methods are available: the index decomposition analysis (IDA) method and the SDA method. These methods have been widely used to evaluate the contributions of different driving forces to overall changes in regional emissions. SDA can be used to identify a range of production and final consumption effects that the IDA approach fails to identify[70], and it can also be used to assess direct and indirect effects affecting the entire supply chain[71]. In this study, we conducted an SDA to quantify the contributions of several drivers of global and national air pollutant emissions within an MRIO framework. The SDA method allows for the compartmentalization of changes in global emissions based on changes in constituent sectors.

Emissions associated with products and services that are produced in region r but consumed in region s ($\mathbf{F}^{rs}$) can be expressed as follows:

$$\begin{aligned}\mathbf{F}^{rs} &= h^{r}(\mathbf{I}-\mathbf{A})^{-1}\mathbf{Y}^{s} \\ &= \sum_{i}\sum_{j}\sum_{k}\frac{E_i^r}{\mathrm{x}_i^r}L_{ij}^{rk}\frac{fd_j^{ks}}{fd_j^s}\frac{fd_j^s}{fd^s}\frac{fd^s}{P^s}P^s \\ &= \sum_{i}\sum_{j}\sum_{k}e_i^r L_{ij}^{rk}D_j^{ks}C_j^s H^s P^s\end{aligned} \quad (3)$$

where $\mathbf{F}^{rr}$ is a vector of elements that represent emissions produced locally based on the final consumption when r=s; $\mathbf{F}^{rs}$ denotes the emissions released in region r related to the crossregional final products consumed in region s when $r \neq s$; h^{r} is a vector of the corresponding direct emission intensity in region r but is zero for all other regions; E_i^r denotes air pollutant emissions from sector i in region r, which includes emissions from fossil fuel (Ef) and a process emission component (Ep); x_i^r is the total output of sector i in region r; L_{ij}^{rk} is an element of the Leontief matrix that denotes the input from sector i in region r required for one dollar of output in sector j in region k; $\mathbf{fd}^s$ is the final demand; fd_j^s is the demand for products in sector j in region s; fd_j^{ks} denotes the final demand for products in sector j in region s imported from region k; and P_s represents the regional population in years. As Ef is region, sector and fuel specific, for each fuel type the emissions in each sector are estimated as the product of the fuel combustion and a time emission factor. We can then further decompose fuel use Ef into an emission factor, an energy mix and an energy intensity component[45], that is

$$\begin{aligned}\mathbf{F}^{rs} &= \sum_i \sum_j \sum_k e_i^r L_{ij}^{rk} D_j^{ks} C_j^s H^s P^s \\ &= \sum_i \sum_j \sum_k \sum_h \left(I_{i,h}^r M_{i,h}^r T_i^r + Ep_i^r \right) L_{ij}^{rk} D_j^{ks} C_j^s H^s P^s\end{aligned} \tag{4}$$

Thus, as is shown in Equation (4), the total change in $\mathbf{F}^{rs}$ is determined by quantifying the contributions of the nine driving factors described above.

1) $I_{i,h}^r = (Ef)_{i,h}^r / (En)_{i,h}^r$ (emissions from fuel combustion) is the emission intensity of fuel type h in sector i region r, reflecting the integrated emission factor of coal (e.g., coke, cleaned coal), oil and gas. For example, The shift from coke to anthracite would also change the emission factor. $(En)_{i,h}^r$ is the used fuel type h in sector i region r.

2) $M_{i,h}^r = (En)_{i,h}^r / (En)_i^r$ (emissions from fuel combustion) is energy mix which is defined as the use shares of the different fuel types (coal, oil and gas) in sector i region r. $(En)_i^r$ is the total fossil fuel use sector i region r.

3) $T_i^r = (En)_i^r / x_i^r$ is energy intensity in sector i region r and measures the energy consumption per unit of output, which indicates the energy efficiency.

4) $(Ep)_i^r = (En)_i^r / x_i^r$ (emission intensity-process emission) measures air pollutant emissions per unit of output from sector i in region r. This factor reflects technological efficiency gains, energy mix changes, etc.

5) L_{ij}^{rk} denotes the input from sector i in region r required for one dollar of output in sector j in region k, and it is further split into the sectoral emission intensity level and trade structure based on five sectors.

6) $D_j^{ks} = fd_j^{ks} / fd_j^{s}$ (trade structure) is the share of the final demand for products in sector j in region s imported from region k.

7) $C_j^{s} = fd_j^{s} / fd^{s}$ (consumption structure) is the share of the demand in region s for products in sector j.

8) $H^{s} = fd^{s} / P^{s}$ (per capita consumption) measures the economic growth effect.

9) P_s represents the population effect.

Thus, regional production-based emissions can be calculated as follows:

$$F^{r} = \sum_{i}\sum_{j}\sum_{k}\sum_{s} e_i^{r} L_{ij}^{rk} D_j^{ks} C_j^{s} H^{s} P^{s} \qquad (5)$$

Similarly, regional consumption-based emissions can be calculated as follows:

$$F^{s} = \sum_{i}\sum_{j}\sum_{k}\sum_{r} e_i^{r} L_{ij}^{rk} D_j^{ks} C_j^{s} H^{s} P^{s} \qquad (6)$$

Emissions from residential household energy consumption (F_d) are determined and divided into direct residential emissions in country r per dollar of consumption (U^r), consumption (H^r) and population (P^r).

$$F_d = U^{r} H^{r} P^{r} \qquad (7)$$

For production-based emissions, the regional air pollutant emissions between two points in time (indicated by subscripts 0 and 1) can be expressed as follows: $\Delta F^{r} = F_1^{r} - F_0^{r}$. However, a unique solution to the decomposition is not available[48,72]. For m factors, the number of potential complete decompositions without any residual terms is equal to $m!$[72]. Because our analysis focuses on ten factors, we follow the methods of previous studies and use the average of so-called polar decompositions as an approximation of the average of all $m!$ decompositions[72]. A full description of the SDA method is provided in the Supplementary Information (SI) section.

Uncertainty

Uncertainties in the decomposition analysis are related to errors in production-based emissions inventories and multiregional input-output (MRIO) calculations because of inaccuracies in national economic statistics and data harmonization[73]. The uncertainty analysis of production-based emissions inventories used in this study was conducted using a Monte Carlo simulation. Variations in source strengths, emission factors, control technology efficiencies, compliance rates, and coal ash content levels and fractions were considered. A Monte Carlo simulation which varies these parameters of emission inventory indicated that the emission factor dominated the uncertainty [9,10,60,66]. The detailed comparison can be found in our published papers[9,10,60,66]. Table S5 also shows comparison between anthropogenic $PM_{2.5}$ emissions of this study and those of previous study for some Asian countries. Moreover, the MRIO calculations contributed additional errors due to uncertainty in economic data[74,75]. Generally, GTAP data are widely accepted and used as a reputable source for MRIO analyses[16,17,74]. For example, Qita et al.[76] demonstrated that the MRIO calculation show a high reliability. The uncertainty of SDA, such as the sign reversal problem[77] discussed by previous study has been partly addressed by further decomposition of the Leontief's multiplier. Additionally, as there is no unique solution for the decomposition but constrained by the extremely large dataset, the SDA in this study used polar decomposition as an approximation of the average of all *6*!=720 decompositions[72,78], and therefore may introduce a certain amount of uncertainty. Lots of efforts are appreciated to address these uncertainties and limitations in the near future.

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Data and Software availability. All the original data can be obtained from given data sources. The data are processed in MATLAB 2016. All datasets generated during this study are available as supplementary data and also in the figshare repository with the identifier https://figshare.com/account/home#/projects/67259.

Author Contributions

J.M., J.L. and D.G., designed the study. Q.Z., T.H., and W.M. provided the emission inventory. J.M. conducted the multi-regional input–output analysis and structural decomposition analysis. J.M., D.G, and Z.L. interpreted data. J.L., Z.M., and Z.L. coordinated and supervised the project. J.M. led the analysis and writing. J.M., H.Y., K.Y., J.L., D.G., Z.L, Z.M., D.C., X.W., and S.T. contributed to the writing of manuscript.

Declaration of Interests

The authors declare no competing interests.

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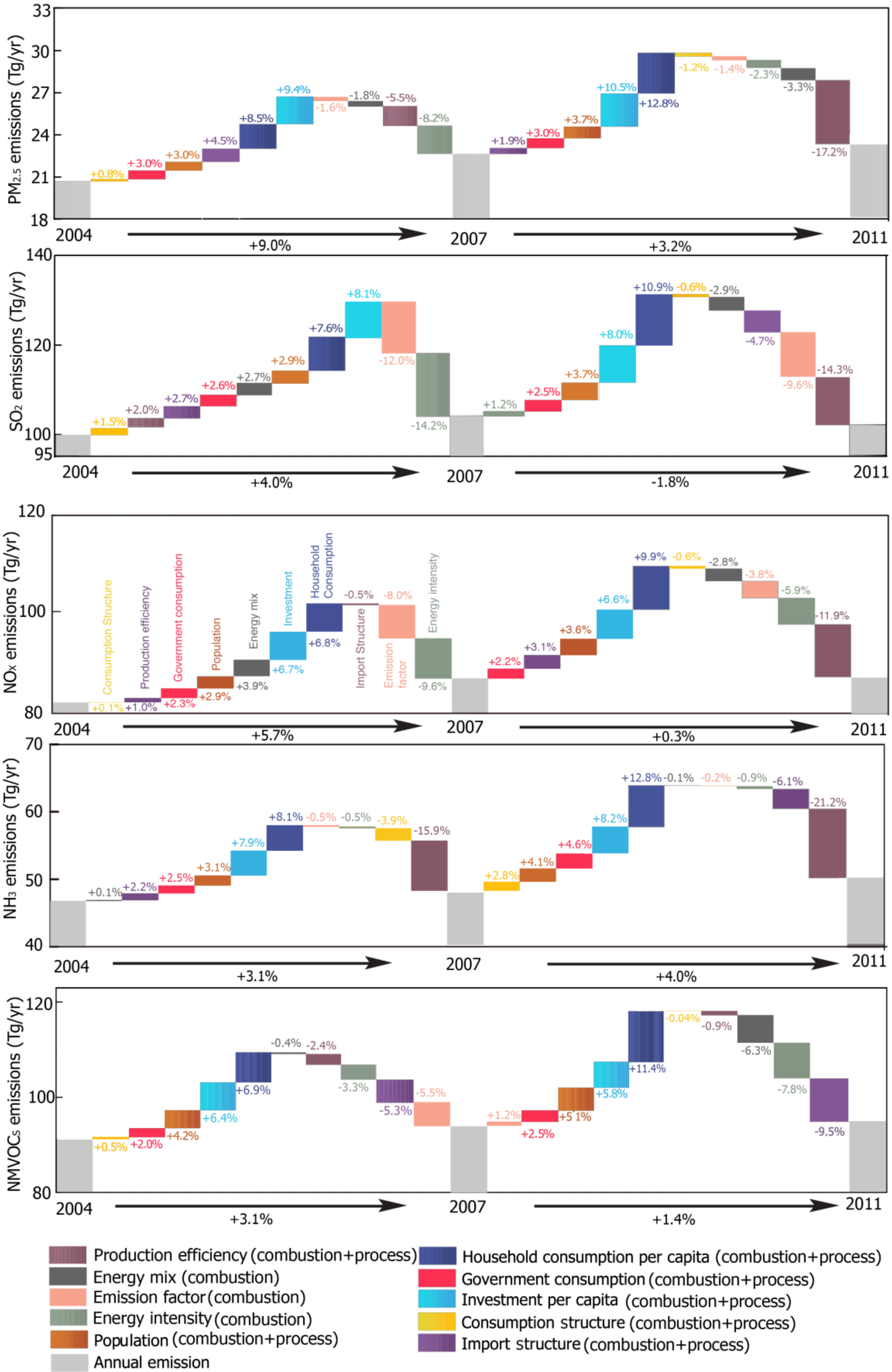

Figure 1. Contributions to the changes in industrial primary $PM_{2.5}$, SO_2, NO_x, NH_3 and $NMVOC_S$ emission levels from 2004 to 2007 and from 2007 to 2011. The percentage provided for each coloured bar represents the contribution of each factor to the change in air pollutant emissions for the two periods.

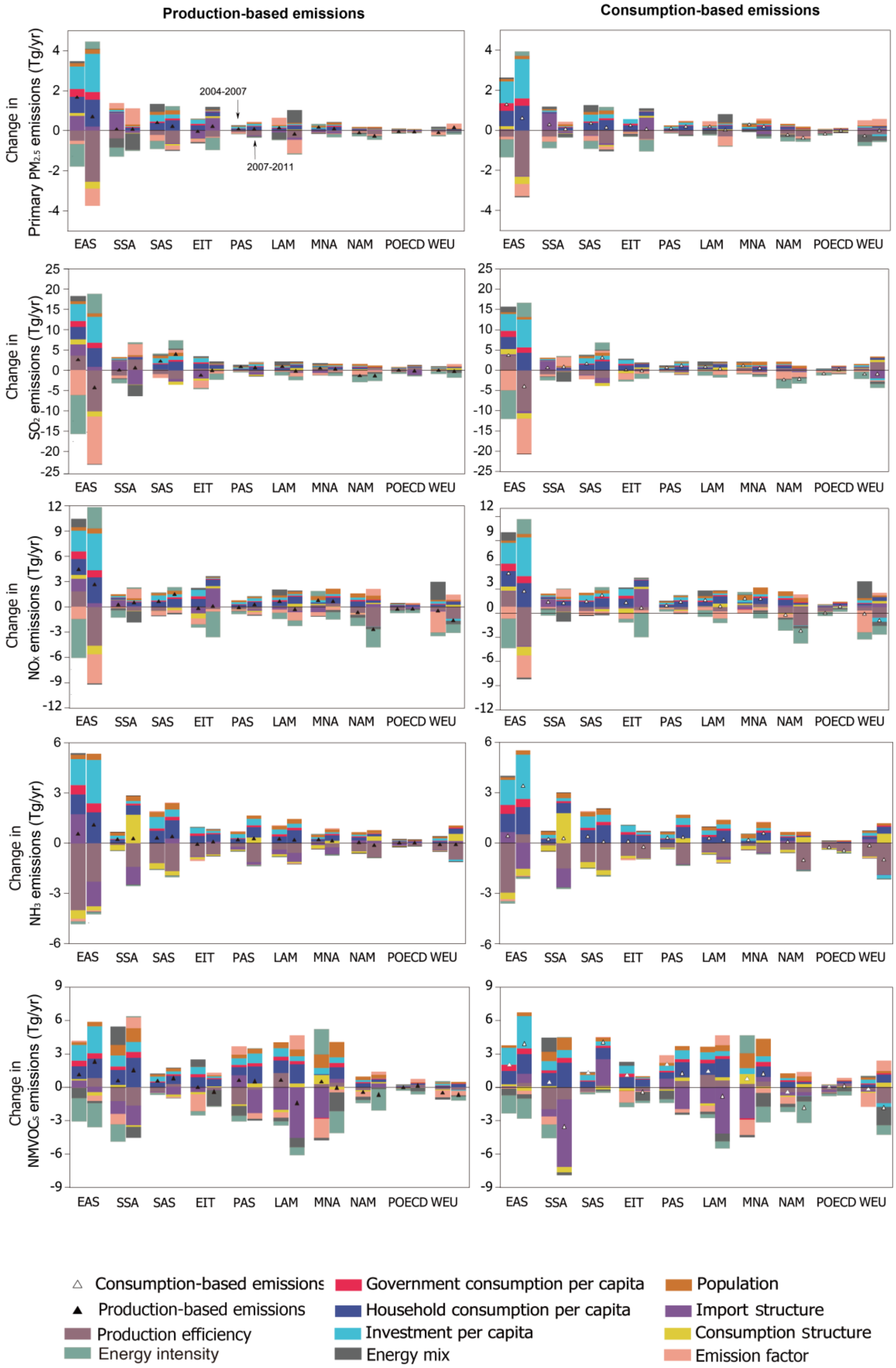

Figure 2. Contributions to the changes in regional air pollutant emissions (Tg/yr) for the two periods (i.e., from 2004 to 2007 and from 2007 to 2011). Regional codes: EAS: East Asia (mainly

China); SSA: Sub-Saharan Africa; SAS: South Asia (mainly India); EIT: Economies in Transition (including Eastern Europe and the former Soviet Union); PAS: Southeast Asia and the Pacific; LAM: Latin America and the Caribbean; MNA: the Middle East and North Africa; NAM: North America (USA and Canada); POECD: Pacific OECD-1990 countries (including Japan, Australia, and New Zealand); and WEU: Western Europe.

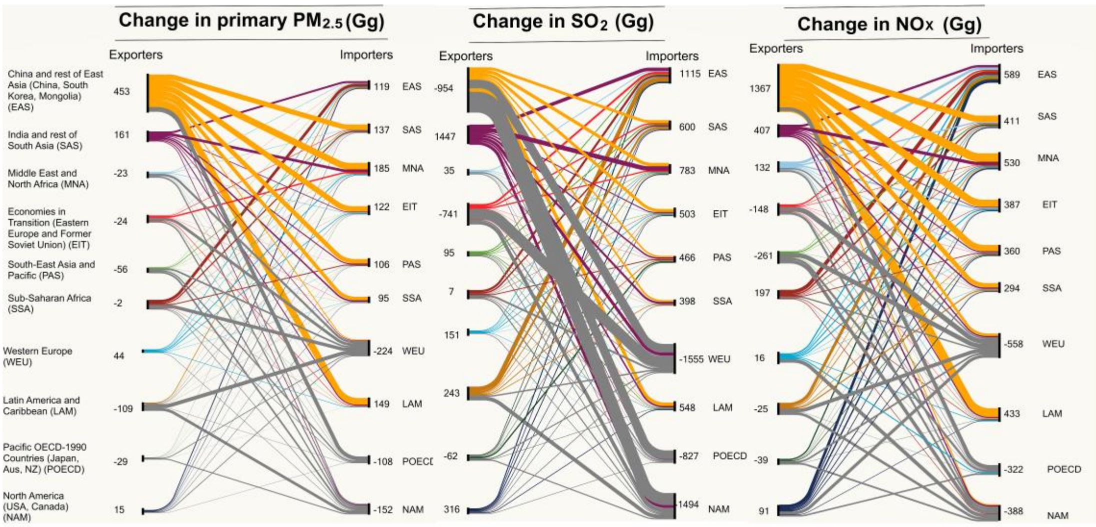

Figure 3. Changes in transboundary primary $PM_{2.5}$, SO_2 and NOx emissions from 2004 to 2011 via international trade. The flows between exporters and importers represent the change of emissions embodied in two regions from 2004 to 2011. Flow widths on the left and right represent emissions (in Gg) produced by regions and those induced by consumers, respectively. Grey flows denote flows with negative values.