Global sulfur dioxide emissions and the driving forces

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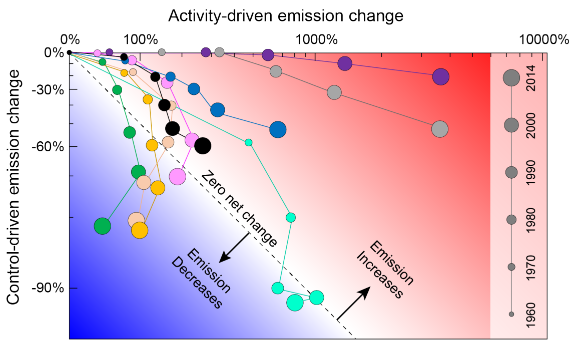

Abstract

The presence of sulfur dioxide (SO_2) in the air is a global concern because of its severe environmental and public health impacts. Recent evidence from satellite observations shows fast changes in the spatial distribution of global SO_2 emissions, but such features are generally missing in global emission inventories that use a bottom-up method due to the lack of up-to-date information, especially in developing countries. Here, we rely on the latest data available on emission activities, control measures, and emission factors to estimate global SO_2 emissions for the period 1960–2014 on a 0.1° × 0.1° spatial resolution. We design two counterfactual scenarios to isolate the contributions of emission activity growth and control measure deployment on historical SO_2 emission changes. We find that activity growth has been the major factor driving global SO_2 emission changes overall, but control measure deployment is playing an increasingly important role. With effective control measures deployed in developed countries, the predominant emission contributor has shifted from developed countries in the early 1960s (61%) to developing countries at present (83%). Developing countries show divergency in mitigation strategies and thus in SO_2 emission trends. Stringent controls in China are driving the recent decline in global emissions. A further reduction in SO_2 emissions would come from a large number of developing nations that currently lack effective SO_2 emission controls.

Introduction

Atmospheric emissions of sulfur dioxide (SO_2) are of great concern because of their adverse impacts on the climate and human health[1,2]. Through decades of efforts to reduce SO_2 emissions, developed countries have made remarkable progress in mitigating SO_2 pollution[3,4], with considerable benefits for regional to global environments[4,5]. Most developing countries, however, either have not adopted effective strategies or have just started to do so in recent years[6]. Only after 2000, for example, did China start to impose strict controls on SO_2 emissions[7], although China's pace in emission reduction has accelerated since 2013 following the issuing of the Air Pollution Prevention and Control Action Plan[8]. India and many other developing countries, on the other hand, have seen continuing increases in SO_2 emissions over the past decades because of the growth of electricity demand and an absence of effective regulations on emission controls[9]. Observations from space imply that India is overtaking China as the largest SO_2 emitter in the world[10]. Such divided trends in SO_2 emissions by country have led to large-scale rapid changes in the spatial distribution and hotspots of global SO_2 emissions in recent years[11,12], which have not been well captured by global emission inventories due to a lack of detailed information[11].

Several studies and official reports have provided bottom-up estimates of SO_2 emissions, although there have been discrepancies in their estimated values (**Table S1**). Nevertheless, it is clear that SO_2 emissions mainly originate from fossil fuel consumption, especially for power generation, and industrial activities, including petroleum refining and metal smelting[13,14]. Fuel quality, energy mix, industrial structure, and control measures are determinants of SO_2 emissions in a given country[15], and these determinants are ultimately associated with the socioeconomic development status of the country[6]. Studies have found an inverted-U shaped relationship between SO_2 emissions and socioeconomic development levels, indicating that SO_2 emissions generally follow the Environmental Kuznets Curve (EKC) as the economy grows[16,17]. Given recent changes in developing countries, however, differences in the SO_2 emission trajectories between developed and developing countries are less clear.

Here, we provide the up-to-date version of a global SO_2 emission inventory developed by the research group at Peking University (i.e., PKU-SO_2, freely available at http://inventory.pku.edu.cn/). PKU-SO_2 is compiled by the bottom-up method, spans the period 1960–2014, and incorporates the latest information available for developing countries, especially for China, on activities, emission factors, and control measures (see

Methods). Additional international fuel trade information was also considered to potentially reduce the emission uncertainties induced by the spatiotemporally varying sulfur content of fossil fuels[18]. Based on PKU-SO_2, we present in this paper a comprehensive assessment of global SO_2 emissions regarding the source profiles, spatial distribution, and temporal trends. We compare our assessment with previous studies and discuss the potential reasons for the discrepancies between studies. To address the factors driving the changes in SO_2 emissions over the study period, we introduce two counterfactual scenarios to decompose the contributions of emission activities and sulfur-control measures on historical emission trends. We then investigate the relationship between SO_2 emissions and socioeconomic development status by different groups of countries, using per-capita gross domestic product (*GDP*cap) as an indicator. These analyses based on the newly compiled emission data should have important policy implications for SO_2 pollution mitigation.

Methods

Emission inventory development

A bottom-up method was used to calculate country-level emissions as follows:

$$Emis = \sum_i A_i \times EF_i$$

where A_i and EF_i represent the emission activity and the emission factor (*EF*, defined as the mass of pollutants emitted per unit of emission activity) for source *i*, respectively. A total of 75 emission sources were considered in the emission inventory, which included eight major sectors and six fuel types (see **Table S2** for detailed source information). The emission activity data were collected from the International Energy Agency (IEA)[19] for power generation, industry combustion, transportation, and residential sectors except for China, for which the residential energy consumption was taken from an updated residential energy dataset based on a recent national questionnaire survey[20,21]. The activity data of non-combustion industries were provided by the U.S. Geological Survey (USGS)[22] and the U.S. Energy Information Administration (USEIA)[23]. Information on the dry matter burned as agriculture waste and in wildfires and deforestation was provided by the Global Fire Emissions Database (GFED) at a monthly resolution[24].

The *EF* data were collected and treated differently among emission sources. For the hard coal and oil consumed in power and industrial sectors, the *EF*s were directly derived from a newly compiled *EF* dataset for SO_2, detailed in a previous study[18]. This *EF* dataset considered the effects of both international fuel trade on country-specific sulfur contents and control measures (e.g., flue gas desulfurization or FGD, sulfur removal from petroleum refinery) [18]. The *EF*s of lignite were calculated by country based on the country-specific sulfur and ash contents of lignite collected from the literature (see **Table S3**) together with the FGD promotion rate according to our recent study[18]. For the transportation sector, linear regression was adopted to predict the spatiotemporal variation in *EF*s based on a collection of 125 *EF* measurements obtained from the literature (**Table S4**). For nonferrous metal smelters (i.e., nickel, lead, copper, and zinc) and natural gas production, the *EF*s in uncontrolled conditions were originally calculated using a mass-balance method with information obtained from the USGS and USEIA[22,23] and were further calibrated by the accompanying production of byproducts (e.g., sulfuric acid)[22,25] to obtain the final *EF*s. For other sources, which account for small fractions of the total SO_2 emissions, constant *EF*s from previous studies were adopted. The country-level emissions were spatially allocated into 0.1° × 0.1° grid cells by source using high-resolution energy data[26] as a surrogate. Emissions from the residential sector were then temporally resolved by month using grid-specific monthly profiles generated in a previous study[27].

Decomposition of driving forces on SO_2 emissions

In addition to the real case, we developed two counterfactual scenarios to quantify the influences of two major drivers on SO_2 emissions, i.e., emission activity and control measure. The emission activity considered not only the changes in the magnitude of energy consumption and industrial production but also the changes in sulfur content induced by international fuel trade. We set 1960 as the starting point, and by holding control measures constant as in 1960, we quantified the net influences of emission activity on emission changes from 1960 to 2014. Similarly, the influences of sulfur-control measures were quantified by keeping the sulfur-control rates during the study period constant as those in 1960 when the rates were equal or close to zero. The effects of these two drivers constituted unique trajectories of historical SO_2 emissions for both individual countries and the globe, which allowed us to compare the importance of these two drivers in specific regions and periods.

Uncertainty analysis

A 10000-time Monte Carlo simulation was performed to address the uncertainty in the emission estimates. The overall uncertainty stemmed from uncertainties in the *EF*s, activity data, and technology division (including control measures). The distributions of the *EF*s were subject to log-normal distributions. Activity data and technology split were assumed to be uniformly distributed. Following previous studies[28,29], the standard deviation was set as 50% for technology division, 5% for fuel consumption in the power and industrial sectors, 15% for transportation, 20% for indoor biomass fuels, 30% for outdoor biomass burning, and 10% for all the other sources. The uncertainties were presented here as the medians and the interquartile ranges (i.e., the interval between the 25th and 75th percentiles) of the emission values given by the Monte Carlo simulation.

Results and discussion

Global emissions and source profiles in 2014

With consideration of global fuel trade, the global total SO_2 emission from all sources (excluding volcanic emissions) was estimated as 105.4 Tg y^{-1} (95.8~119.8 Tg y^{-1}) in 2014, with a predominant contribution from anthropogenic sources (98%). Our inventory features not only the incorporation of the international fuel trade but also a high spatial resolution (0.1° latitude × 0.1° longitude), a long-term temporal coverage (1960-2014), and detailed source information (76 sources). **Table S5** compares the emission estimates in this study with several past global SO_2 emission inventories in their latest years. Generally, our estimates are in line with these previous studies. For example, the total anthropogenic emission was estimated to be 111.7 Tg y^{-1} in 2014 by CEDS[13] and 102.4 and 106.9 Tg y^{-1} in 2010 by EDGAR and HTAP, respectively[30,31], compared with 109.2 Tg y^{-1} (99.0~124.2 Tg y^{-1}) and 105.4 Tg y^{-1} (95.9~119.8 Tg y^{-1}) in 2010 and 2014, respectively, in our study. Despite the overall similarity, substantial differences were found in the source profiles, spatial distributions, and temporal trends. Potential reasons for these differences are discussed in detail in the following sections.

We estimated that in 2014, 43% of the total anthropogenic emissions were from power plants (**Fig. S1**), followed by industry (35%) and international shipping (16%). Developed and developing nations showed little difference in the aggregated sectoral profiles but large differences in the fuel profiles in specific sectors. Both groups of nations showed dominant contributions of the power and industry sectors and contributions of <10% from all other sectors together (emissions from international shipping were not assigned to specific countries). Residential emissions accounted for a higher contribution in developing countries, as coal was still a leading fuel type for household cooking and heating in many developing countries[20]. The combustion of hard coal and oil accounted for 78% of the global total emission. Note that these important SO_2 sources were strongly influenced by the international fuel trade, which was detailed in a previous study[18]. Developing countries showed a larger share (68%) of emissions from coal combustion than developed countries (47%). The combustion of biomass fuel, a major contributor to many other atmospheric pollutants (e.g., $PM_{2.5}$[29] and PAHs[28]), contributed to less than 2% of the total SO_2 emission. Omitting emissions from shipping, the source profiles of the SO_2 emissions in this study were compared with the global and regional estimates from CEDS in 2014 (the latest year of CEDS estimates available) (**Fig. S2**). Our global estimate largely agreed with that of CEDS but showed a lower contribution from developing nations. Our estimate of China's total anthropogenic SO_2 emission, for example, was 22.8 Tg y^{-1} (20.1~25.8 Tg y^{-1}) in 2014, which was close to the estimate reported by the Multi-resolution Emission Inventory for China (MEIC) (20.4 Tg y^{-1})[8] but was 38% lower than the CEDS estimate (36.5 Tg y^{-1}). The major differences with CEDS are in the industrial sector and are likely attributed to the different FGD promotion rates adopted.[13,18] The FGD penetration in this study was based on the reported data from China statistical yearbook on environment[32], which was generally larger than previous ones[7]. In recent years, China has made considerable efforts to control air pollution from industrial sector[33]. A recent study has suggested that the industrial sector was the leading driver of the declined SO_2 emission in China since 2010[8].

Spatial distribution

Fig. 1 shows the spatial distribution of all-source SO_2 emissions (excluding those from aviation) in 2014 at a spatial resolution of 0.1°×0.1°. Among all countries, China (especially in the eastern part) and India exhibited the highest emission densities, mainly due to the intensification of coal consumption and metal smelting; these two countries accounted for 58% of coal consumption and 44% of metal smelting worldwide[19]. Emissions from the top 20 coal-fired power plants in China accounted for 6% of China's total SO_2 emissions, which is equivalent to the annual emission of Brazil. Relatively high emission densities were also found in Eastern Europe, Middle Europe, and the eastern United States due to large emissions from power generation and industrial sources. These high-emission regions usually coincided with dense population, implying that population density could play an important role in determining the spatial variation of total SO_2 emissions. Per-capita emissions varied substantially both within and across countries (**Fig. 1b**).

Densely populated places, such as eastern China, the peninsula of India, Europe, and the eastern United States, showed lower per-capita emissions (< 10 kg cap^{-1}) than the global average (12.3 kg cap^{-1}, 11.2~14.0 kg cap^{-1}).

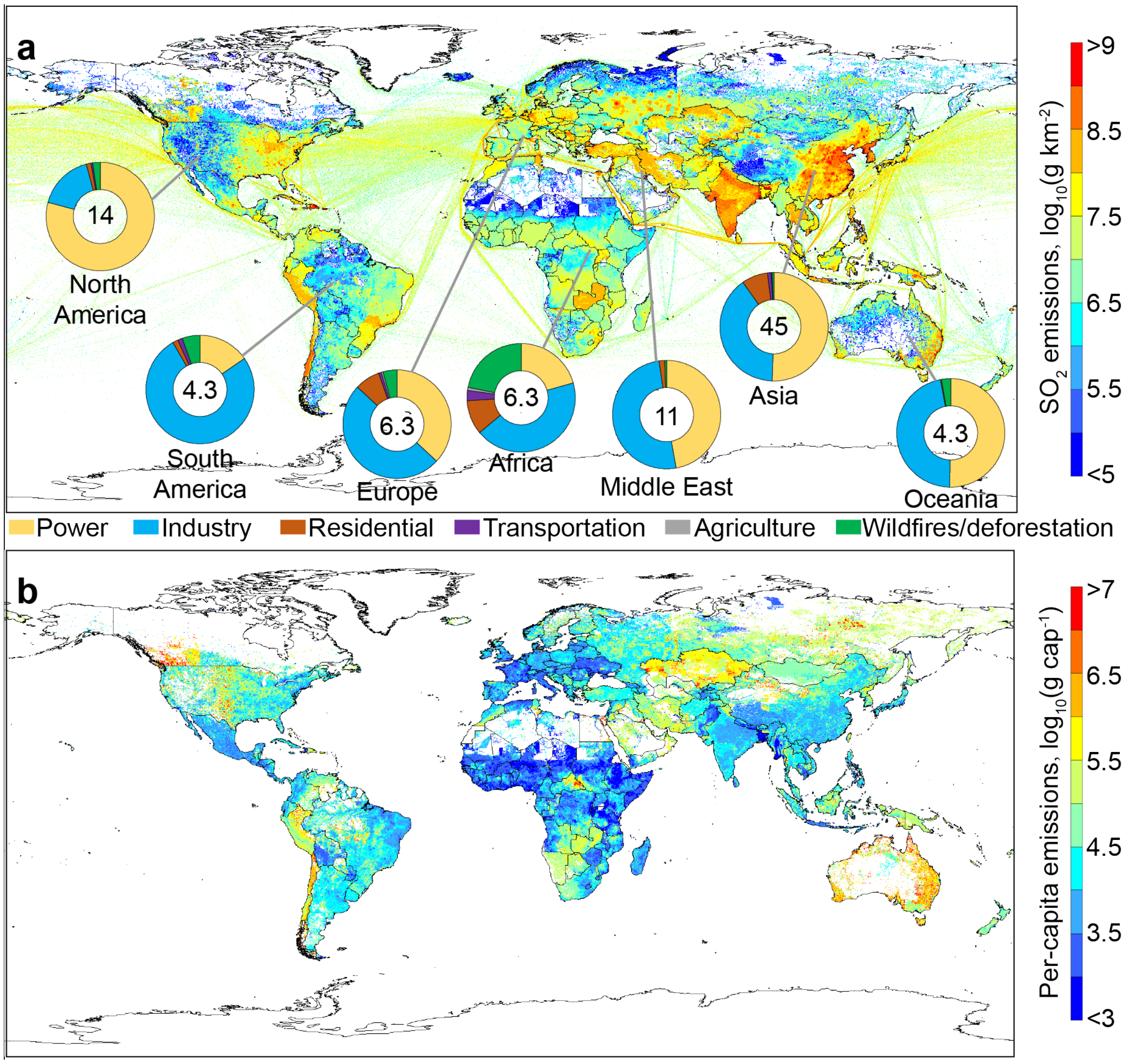

Fig. 1 Geographical distributions of (a) annual all-source SO_2 emissions (excluding aviation emissions) and (b) per-capita emissions in 2014. The embedded diagrams in panel (a) show the source profiles for 7 regions. The world shapefiles were obtained from Esri (ArcGIS Hub, *Countries WGS84*. June 21, 2015. http://www.arcgis.com/home/item.html?id=30e5fe3149c34df1ba922e6f5bbf808f).

According to **Fig. 1a**, the frequency distribution of gridded SO_2 emission densities, as shown in **Fig. S3**, was more leptokurtic compared to $PM_{2.5}$ and other incomplete combustion byproducts (e.g., BC, PAHs, and CO).

This difference reflected different geographic features of their dominant sources. The unimodal distribution of SO_2 emissions indicates a more concentrated spatial distribution compared with other pollutants with multimodal distribution. SO_2 emissions mainly came from point sources (e.g., power generation and industrial sources), while one of the major sources of incomplete combustion products was the combustion of solid fuel in rural households, of which the spatial distribution was more scattered. As a result, SO_2 emissions were agglomerative in space, with reduced spatial continuities, compared to $PM_{2.5}$ (the Moran's I is 0.50 for $PM_{2.5}$ and 0.32 for SO_2).

The inventory showed differences in both the total emissions and the source profiles among continents (**Fig. 1a**). For example, Asia contributed 62% of the global terrestrial SO_2 emission; Europe and Africa only contributed 8% and 5%, respectively. In Asia, power generation and industry were the predominant contributors, accounting for 50% and 42% of Asia's SO_2 emission, respectively. In Europe, power generation contributed 38% of its total emission, which was less than industrial sources (50%), due primarily to a lower dependency on thermal electricity powered by fossil fuels (42%) compared to many other regions of the world (e.g., 67% in the United States, 75% in China, and 81% in India). In Africa, wildfires and deforestation contributed a large fraction of SO_2 emissions (21%).

Fig. S4 shows the geographical distributions of sectoral emissions in 2014. By choosing the sector with the highest contribution in each grid cell, we derived the sectors most responsible for SO_2 emissions on a 0.1°×0.1° grid (**Fig. S4**), which provided more detailed spatial information on the source profiles and could be especially beneficial to emission control management. We found that although emissions from power and industrial sectors dominated China's total emission, the residential sector was still a major source in some parts of the northern and western China due to the lower intensity of industrial activities in these regions.[34] This pointed to the different emission structures between rural and urban areas, as shown in **Fig. S5**. Globally, rural areas released over 65% of the SO_2 emissions into the atmosphere. Likewise, the per-capita emissions for rural residents (15.1 kg cap^{-1}, 13.7~17.2 kg cap^{-1}) were higher than those for urban residents (9.1 kg cap^{-1}, 8.3~10.3 kg cap^{-1}), especially for developed countries with less rural populations (31.4 kg cap^{-1} on average, 28.6~35.7 kg cap^{-1}). Further investigation showed that SO_2 emissions in rural areas were still dominated by power generation and industrial sources, in contrast with emissions of incomplete combustion products, such as PAHs, which also showed a higher per-capita emission for rural residents compared to their urban counterparts but was mainly caused by high emissions from the rural residential consumption of coal and biomass[28]. Relatively small differences were found between developed and developing countries in the per-capita emissions attributed to power generation. It was estimated that per-capita coal consumed in power generation was 61% higher in developed countries than in developing countries,[19] which offset the effects of better controls on emission reduction in developed countries.

Temporal trends of SO_2 emissions

Fig. 2 shows the global trend of SO_2 emissions and the contributions from major sectors, fuel types, and regions. The global emission increased at an average rate of 2.4 Tg y^{-1} (1.2~3.7 Tg y^{-1}) during the period 1960–1975, peaked at 136 Tg y^{-1} (122~153 Tg y^{-1}) in the late 1970s, and started to decrease in the 1980s, with an inverse trend between 2000 and 2007 as a result of increasing coal consumption in China. This emission trend was very different from the emission trends of $PM_{2.5}$ and many other pollutants for which the emissions largely continued rising over the last several decades[29,35,36]. Between 1960 and the late 1970s, global coal consumption increased by 135%[19], but sulfur controls during this period were only adopted in a limited number of countries (e.g., Japan and the United Kingdom). The rising energy demand, especially in developed countries, drove the increase in global SO_2 emissions. Since the late 1970s, FGD technology has been scaled up in many developed countries as an efficient solution to reducing SO_2 emissions[37]. For example, the cumulative capacity of FGD units increased from 0.8 GW in 1975 to 21.1 GW in 1990 in the United States[38]. With policy restrictions, the expanding deployment of sulfur mitigation measures gradually offset the SO_2 emissions that were otherwise increasing due to the growing energy demand, and the global emission started to decline. Thanks to the catch-up of control measure deployment in developing countries in the 2000s[8], the declining trend continued after an 8-year reversion between 2000 and 2007. The declining trend is likely to continue for years because of the regulations and measures proposed and taken in China and other developing countries[39]. For example, China banned imports of coal with high ash and sulfur in 2015 and the sale of high-sulfur diesel used by tractors and ships in late-2017, both of which are expected to further contribute to the global SO_2 emission reduction in the coming years[40].

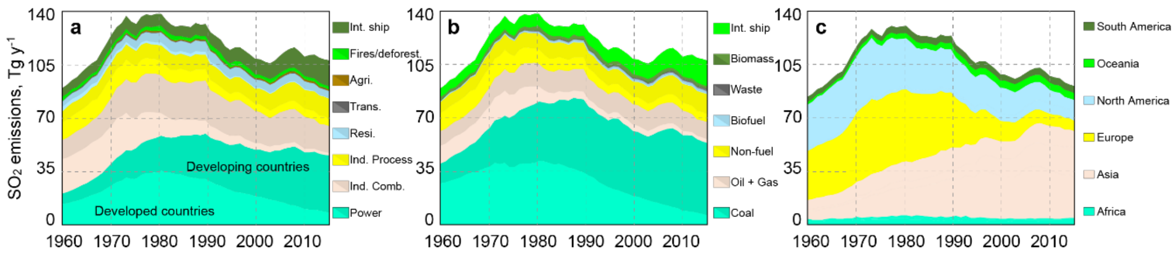

Fig. 2 Temporal trends of annual SO_2 emissions by sector (a), fuel type (b), and region (c). The light and shaded areas indicate emissions from developed and developing countries, respectively. Emissions from international shipping, which mainly originated from oil combustion, are illustrated separately in **Fig. 2b** because they cannot be allocated to individual countries.

During the last 55 years, SO_2 emissions revealed substantial shifts in terms of emission sectors, sources, and regions. As shown in **Fig. 2a**, emissions from industrial combustion, a primary contributor in the 1960s, started to decline in the early 1970s due to the transition of major fuel types from coal to electricity and gas[19]. On the other hand, emissions from power plants increased rapidly from 20.5 Tg y^{-1} (18.5~22.8 Tg y^{-1}) in 1960 to 58.4 Tg y^{-1} (52.8~64.9 Tg y^{-1}) in 1980 and were driven by the fast growth of electricity demand. The contribution of power generation to global total emissions has remained constant at approximately 45%since the mid-1980s and has slightly decreased recently (since 2005). The recent decrease was partly caused by the growing control efforts in developing countries[41]. The residential sector only accounted for < 8% of the total emissions during the entire study period. Notably, the contribution of shipping emissions increased from 6% in 1960 to 16% in 2014. Recent studies have showed that controlling sulfur in ship oil can yield considerable health and climate benefits[42,43].

In terms of the fuel types (**Fig. 2b**), coal contributed the largest emissions to the global total, with a peaking contribution of 63% in the mid-1980s. The decreasing trend since then was a joint result of FGD promotion in power plants and the transition from coal-powered to electricity-powered industries[19]. Given economic and technological gaps, the emission trends in developed and developing countries differed essentially. The total emissions from developed countries peaked at 70.2 Tg y^{-1} (63.4~78.4 Tg y^{-1}) in 1973 and have continuously declined since then, primarily driven by the promotion of control measures in power generation (e.g., FGD) and industries (e.g., sulfur recovery technology in petroleum refining and smelting). At the same time, emissions from developing countries gradually increased until very recently because of the increase in energy consumption and the expansion of the metal industry. For example, copper production in China increased by a factor of 10 between 1990 (560 Gg) and 2014 (6500 Gg)[22]. As a result, the predominant emission contributor has shifted from developed countries in the early 1960s (61%) to developing countries at present (83%), which has also led to large-scale spatial changes over time. As displayed in **Fig. 2c**, the relative contribution from Asia has increased rapidly since 1960, though it has leveled off in recent years, while the contributions from North America and Europe have decreased over time.

The emission trends reported by this study compared with other studies (**Fig. S6**). The comparison showed general agreement between studies except EDGAR 4.3.2 and Lamarque et al.,[44] of which the reported emissions were lower than the others (**Fig. S6**). We found that the estimation differences for large developing countries such as China and India can be substantial between datasets in the recent years, likely due to poor data availability in these countries. By incorporating more recent information, our estimation suggests that due to the increasing adoption of strengthened control measures, SO_2 emissions in China is decreasing, which represents the major reason for the recent decline in global SO_2 emissions. This decreasing trend agreed well with the up-to-date emissions reported by MEIC[8].

In addition, we compared our emission estimates of China and India with top-down emission estimates derived from satellite inversion (**Fig. S7**). Large differences by factors of 2 ~ 3 were found among the top-down estimates. Since all the top-down estimates were constrained by observations of the Ozone Monitoring Instrument (OMI) aboard the Aura satellite, such large differences could be mostly attributed to the different models and retrieval algorithms used[45,46]. Despite the large uncertainties, generally decreasing trends were found in China since 2006, which was in consistent with our results (**Fig. S7a**). While for India, those top-down estimates were considerably lower than the estimates in this and most other bottom-up inventories, although all showed continuously increasing trends (**Fig. S7b**). The large disparity could be due to the uncertainties from various sources in the bottom-up estimation, such as EFs and energy consumption[28,29] but could also be caused by the quality of the satellite observations, which can be contaminated by a variety of factors such as cloud cover and land property, and the uncertainties in the retrieval and inversion algorithms in the top-down estimation could also play a role[46]. Future studies were called for to narrow down the uncertainties in both emission estimation approaches and reduce the disparity between the two estimates.

Decomposition analysis of global and regional SO_2 emission trends

In addition to the emission estimates using the real-world data (called the "real case"), we designed two counterfactual scenarios to decompose the historical SO_2 emission trends into the changes in 1) emission activities (i.e., energy consumption and structure, industrial production, trade-induced changes in sulfur contents or SCs) and 2) end-of-pipe control measures (i.e., FGD in power and industrial boilers, control measures in the road transportation, sulfur removal in petroleum refinery, metal smelter, and natural gas production). Based on the baseline emissions in 1960, the relative emission changes induced by emission activities were quantified by holding the control measures constant and changing the emission activities over time as in the real case; the relative emission changes induced by control measure deployment were calculated by holding the emission activities at their 1960 levels and changing the control measures over time as in the real case. **Fig. 3** illustrates the SO_2 emission trajectories along the two dimensions (i.e., emission activities and control measures) on global and regional scales from 1960 to 2014. Emission activity growth and control measure deployment were competing factors affecting the historical SO_2 emission trend. From a global perspective, emission activity growth played a more important role (**Fig. 3**), but substantial differences in the trajectories were found among regions/countries. Before 1970, activity was the leading force for both developed and developing countries, and the emission trajectories extended mostly along the direction of the activity's dimension (the horizontal axis in **Fig. 3**). After 1970, however, the trajectories started to divide: developed countries redirected their trajectories along the control measure's dimension towards the low end; developing countries such as India and China continued to move along the activity's dimension and did not show significant impacts from control measure deployment until recently (**Fig. 3**). The redirection in developed countries was caused by a combination of stringent emission controls, suppressed fossil fuel use, and the overseas outsourcing of raw industrial materials. For example, coal use (in energy count) and nonferrous metal production in the United States decreased by 29% and 18% from 2000 to 2014, respectively[19,22]. For most developing countries, by contrast, activity remained the predominant driver to decadal SO_2 emission changes with slight differences among countries due to different sulfur control efforts. For example, China exhibited a decrease of 52% in emissions due to control measure deployment, compared with 20% in India (**Fig. 3**). Note that both countries showed a similar emission increase induced by activity changes (**Fig. 3**). Even in some developing countries (e.g., Chile in **Fig. 3**), we found rapid transition in the emission trajectories from being activity-dominant (the red area in **Fig. 3**) to control-dominant (the blue area), as in developed countries, leading to a fast decrease in SO_2 emissions.

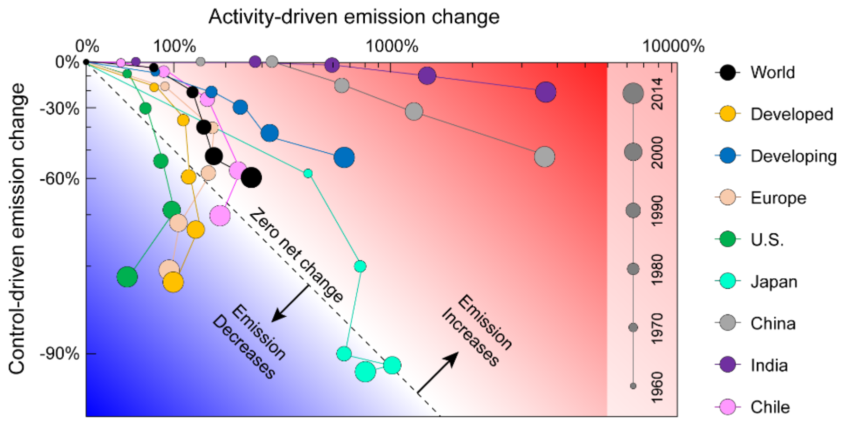

Fig. 3 Trajectories of SO_2 emissions driven by emission activity and control measures from 1960 to 2014. Each trajectory shows the emission ratios of a specific year relative to the base year (1960) driven by activity (*x*-axis) and control measures (*y*-axis). The axes are on a logarithmic scale. The dash line indicates equal forces from the two drivers. The red area indicates stronger impacts of activity growth compared to control measure deployment on emissions; the blue area indicates the opposite.

Environmental Kuznets Curves of SO_2 emissions

To better elucidate the varied SO_2 emission trajectories among countries, we derived the country-level per-capita SO_2 emissions and investigated the relationship between per-capita emissions and socioeconomic development. Although developing countries dominated global SO_2 emissions (84%), the average per-capita emissions in developed countries (13.7 kg cap^{-1}, 12.4~15.3 kg cap^{-1}) were 11% higher than those in developing countries (12.1 kg cap^{-1}, 10.8~13.8 kg cap^{-1}), which was mainly attributed to the much higher per-capita energy consumption in developed countries—the average per-capita consumptions of coal and electricity in developed countries were 66% and 350% higher than those in developing countries, respectively[19]. Stringent controls offset the impact of this high energy consumption on per-capita emissions in developed countries[47]. It was estimated that in 2014, the average removal rate of SO_2 emissions from developing countries only reached the level in developed countries of more than two decades ago. The global SO_2 emissions would be reduced by 41% if emission control measures in developing countries were as advanced as those in developed countries.

Fuel mix and control strength evolve with socioeconomic growth, leading to disparities in SO_2 emissions among countries. **Fig. 4** shows the relationships between log-transformed per-capita SO_2 emissions and socioeconomic levels as identified by *GDP*cap (in 2010 constant USD) in 2014[48]. Excluding those countries/regions with populations of less than 0.1 million, we found an inverted U-shaped relationship, i.e., an EKC[16], of SO_2 emissions with *GDP*cap. By using an empirical quadratic regression, the turning point of SO_2 emissions was identified at *GDP*cap = 30000 USD when the per-capita emission reached its highest level. It should be noted that the EKC and the turning point for SO_2 emissions were based on a global perspective, which could vary significantly in individual countries and regions. Directly applying the relationship to certain countries could induce strong biases. For example, China's *GDP*cap reached 30,000 USD in 2014, but its per-capita SO_2 emission started to decline in 2006, eight years earlier than the turning point indicated by the global EKC. This result is consistent with previous studies that suggested that the EKC patterns of SO_2 emissions differ by country or even city[49]. Some studies have further used the EKC trajectories to develop global emission inventories[50,51]. With a specific focus on SO_2 emissions, we summarized and compared the estimated EKC patterns of different studies (**Table S6**). The global turning point of our study (*GDP*cap = 30,000 USD) was close to the estimate by a recent study (24,300 USD)[52], but both studies suggested much earlier turning points than past studies that relied on data prior to 2000 (in those studies, the estimated turning points were all at *GDP*cap > 150,000 USD). Note that before 2000, sulfur control measures had not been extensively applied in developing countries. This disparity between recent and past studies indicated a potential shift in the global EKC pattern over time. Further investigation revealed that different studies showed consistent turning points for OECD countries (**Table S6**), which fell between 11,000–16,500 USD but were divided for non-OECD countries (the OECD/non-OECD classification was used here to match the classification used in previous studies). For example, our study showed the turning point of non-OECD countries at *GDP*cap = 79,000 USD, which was approximately 50% lower than a previous estimate using 1960–1990 emission data[16]; another estimate using 1984–2000 data did not show a turning point at all[53], indicating that it may occur at a very high *GDP*cap level. Such a difference in the EKCs of non-OECD countries was also observed when using our emission data along. By comparing the EKCs derived from our data in different emission years, we found that the *GDP*cap level (instead of the per-capita SO_2 emission) associated with the turning points of the non-OECD countries decreased from 490,000 USD to 79,000 USD between 1970 and 2014. This decrease ultimately drove the global turning point from 150,000 USD to 30,000 USD during the same period. Using the long-term data, we further examined the changes in per-capita emissions with *GDP*cap by country. **Fig. S8** illustrated the per-capita trends in eight representative countries, showing that the turning points for the developing countries appeared much earlier than developed countries in terms of *GDP*cap.

The mechanism underlying the temporal shift in the global turning point and the spatial difference across countries can be likely explained by a "late mover" advantage associated with the transmission of emission control strategies and technologies between countries/regions[35,54]. For example, decadal experience and knowledge gained from air pollution mitigation in Europe and North America has helped China to adopt efficient strategies for sulfur emission controls in recent years[55], noting that the GDP_{cap} triggering the turning point in China (2,000 USD) was much lower than the GDP_{cap} triggering the turning points in these developed nations (e.g., 21,700 USD for USA and 18,054 USD for UK) (**Fig. S8**). The turning point in India were found even earlier (1,550 USD) (**Fig. S8**). Meanwhile, China is outsourcing its experience to less developed countries, for example, in Southeast Asia.[56] To evaluate the impacts of the "late mover" advantage on global SO_2 emissions, we designed a 'no late mover' scenario assuming that developing countries in 2014 strictly adopted the same sulfur controls as those historically taken by developed counties at the same GDP_{cap}. It was found that the total global SO_2 emission would have been 20% higher in the "no late mover" scenario than was in the real case, highlighting the importance of the "late mover" effect on global SO_2 emission reduction. Nationally, the 'late-mover' advantage in developing countries reduced the per-capita emissions by 42% on average but with large variation across countries (2% ~ 96%). In China, especially, the 'late-mover' advantage was estimated to reduce SO_2 emissions by 76% in 2014. This analysis may facilitate a better understanding and provide an easier forecast of future SO_2 emission trends in countries across different socioeconomic development levels.

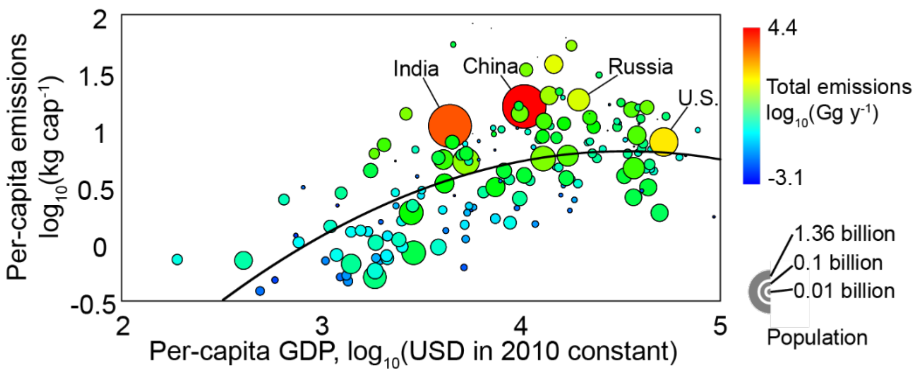

Fig. 4 Dependency of per-capita SO_2 emissions on per-capita GDP for all countries in 2014. The sizes and colors of the circles indicate the country population and total SO_2 emissions, respectively.

Based on a newly compiled SO_2 emission inventory, this study elucidated the spatiotemporal variations of global SO_2 emissions and the underlying driving forces. According to our estimation, the recent trend of SO_2 emissions was subject to a remarkable decrease as a result of both enduring control efforts by developed countries and an increasing awareness of sulfur control in some developing countries. Future trends in global SO_2 emissions will depend on how developing countries act in response to their rapid economic growth and urgent demands for clean air. The world is globalizing, and so is the air pollution issue[57]. Our analysis showed that developing countries benefited from globalization through the transmission of emission control strategies and technologies from developed countries, suggesting that regional and global cooperation is an important way to reduce SO_2 emissions in the genuinely global era.

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Supporting Information

Source profiles of SO_2 emissions (Fig. S1), comparison between CEDS and this study (Fig. S2), the statistical distribution of gridded SO_2 emissions in comparison with other pollutant emissions (Fig. S3), sector-resolved geographical distribution of SO_2 emissions (Fig. S4), rural-urban emission differences (Fig. S5), inter-comparison of global and regional SO_2 emission inventories (Fig. S6). Comparisons of emissions in China and India between top-down estimates and this study (Fig. S7). Historical EKCs for selected countries (Fig. S8).

Differences of SO_2 emissions reported by multi-sources (Table S1), summary of EFs used in this study (Table S2), SCs and ACs of lignite (Table S3), regression model of EFs for motor vehicles (Table S4), global SO_2 emission comparisons (Table S5), summary of turning point of EKC for SO_2 emissions (Table S6).

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