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Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3.1

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Abstract. A long-term historical emission inventory of air and climate pollutants in East, Southeast, and South Asia from 1950-2015 was developed as the Regional Emission inventory in ASia version 3.1 (REASv3.1). REASv3.1 provides details of emissions from major anthropogenic sources for each country and its sub-regions and also provides monthly gridded data with 0.25° × 0.25° resolution. The average total emissions in Asia during 1950-1955 and from 2010-2015 (growth rates in these 60 years) are as follows: SO_2: 3.15 Tg, 42.4 Tg (13.5); NO_x: 1.83 Tg, 47.6 Tg (26.0); CO: 62.2 Tg, 319 Tg (5.13); non-methane volatile organic compounds: 9.14 Tg, 61.8 Tg (6.77); NH_3: 7.99 Tg, 31.3 Tg (3.92); CO_2: 1.12 Pg, 18.3 Pg (16.3); PM_{10}: 5.76 Tg, 28.4 Tg (4.92); $PM_{2.5}$: 4.52 Tg, 20.3 Tg (4.50); black carbon: 0.751 Tg, 3.38 Tg (4.51); and organic carbon: 2.62 Tg, 6.92 Tg (2.64). Clearly, all the air pollutant emissions in Asia increased significantly during these six decades, but situations were different among countries and regions. Due to China's rapid economic growth in recent years, its relative contribution to emissions in Asia has been the largest. However, most pollutant species reached their peaks by 2015 and the growth rates of other species was found to be reduced or almost zero. On the other hand, air pollutant emissions from India showed an almost continuous increasing trend. As a result, the relative ratio of emissions of India to that of Asia have increased recently. The trend observed in Japan was different from the rest of Asia. In Japan, emissions increased rapidly during 1950s-1970s, which reflected the economic situation of the period; however, most emissions decreased from their peak values, which were approximately 40 years ago, due to the introduction of regulations and laws for air pollution. Similar features were found in the Republic of Korea and Taiwan. In the case of other Asian countries, air pollutant emissions generally showed an increase along with economic growth and motorization. Trends and spatial distribution of air pollutants in Asia are becoming complicated. Datasets of REASv3.1, including table of emissions by countries and sub-regions for major sectors and fuel types, and monthly gridded data with 0.25° × 0.25° resolution for major source categories are available through the following URL: http;//www.nies.go.jp/REAS/.

1 Introduction

With an increase in demand for energy, motorization, and industrial and agricultural products, air pollution from anthropogenic emissions is becoming a serious problem in Asia, especially due to its impact on human health. In addition, a

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significant increase in anthropogenic emissions in Asia is considered to affect not only the local air quality, but also regional, inter-continental, and global air quality and climate change. Therefore, reduction in air and climate pollutants emissions are urgent issues in Asia (UNEP, 2019). Short-Lived Climate Pollutants (SLCPs), which are gases and particles that contribute to warming and have short lifetimes, have been recently considered to play important roles in the mitigation both air pollution and climate change (UNEP, 2019). SLCPs such as black carbon (BC) and ozone are warming agents, which cause harm to people and ecosystems. A decrease in the emissions of BC and ozone precursors from fuel combustion led to the decrease of other particulate matter (PM) species, such as sulfate and nitrate aerosols. Even though this is a positive step for human health, it has a negative effect on global warming as sulfate and nitrate aerosols act as cooling agents in the troposphere. Therefore, to find effective ways to mitigate both air pollution and climate change, accurate understanding of the current status and historical trends of air and climate pollutants are fundamentally important.

Recently, Hoesly et al. (2018) developed a long-term historical global emission inventory from 1750 to 2014 using the Community Emission Data System (CEDS). This data set is used as input data for the Coupled Model Intercomparison Project phase 6 (CMIP6). The Emission Database for Global Atmospheric Research (EDGAR) also provides global emissions data of both air pollutants and greenhouse gases, with the current version 4.3.2 ranging from the period between 1970-2012 (Crippa et al., 2016). The EDGAR is used as the default data of input emissions for the Task Force on Hemispheric Transport of Air Pollution phase 2 (HTAPv2) (Janssens-Maenhout et al., 2015). For SLCPs, the European Union's Seventh Framework Programme project ECLIPSE (Evaluating the Climate and Air Quality Impact of Short-Lived Pollutants) developed a global emission inventory based on the GANS model. Current version 5 provides gridded emissions for every five years from 1990 to 2030 and also from 2040 to 2050 (Stohl et al., 2015). However, data from Asia in global emission inventories are generally based on limited country specific information. For the Asian region, several project-based emission inventories are developed, such as Transport and Chemical Evolution over the Pacific (TRACE-P) field campaigns (Streets et al., 2003a, b) and its successor mission, that is Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) (Zhang et al., 2009). Recently, the MIX inventory (mosaic Asian anthropogenic emission inventory) was developed as input emission data sets for the Model Intercomparison Study for Asia (MICS-Asia) Phase 3 by a mosaic of up-to-date regional emission inventories. The MIX inventory is also a component of the HTAPv2 inventory (Li et al., 2017a). For national emission inventories, numerous studies, research papers, and reports have been published. MEIC (Multi-resolution Emission Inventory for China) developed by Tsinghua University is a widely used emission inventory database for China (Zhang et al., 2009; Li et al., 2014; Zheng et al., 2014, Liu et al., 2015) and is included in the MIX inventory. Zhao et al. (2011, 2012, 2013, and 2014) developed recent and projected emission inventories of air pollutants in China. In addition, research papers for regional emission inventories of China were also published recently (Zhu et al., 2018; Zheng et al., 2019). In the case of India, Garg et al., (2006) developed a historical emission inventory of air pollutants and greenhouse gases from 1985 to 2005. For recent years, Sadavarte and Venkataraman (2014) developed multi-pollutant emission inventories for industry and transport sectors and Pandey et al. (2014) developed the same for domestic and small industry sectors for the same time period, that is 1996-2015. For Japan, several project-based emission data sets were developed, such as the Japan

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Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC 2012a, b, c; 2014), East Asian Air Pollutant Emission Grid Database (EAGrid) (Fukui et al., 2014), and emission data sets for Japan's Study for Reference Air Quality Modeling (J-STREAM) (Chatani et al., 2018). In addition, there are studies for other countries and regions, such as the Clean Air Policy Support System (CAPSS) for Republic of Korea (Lee et al., 2011), Thailand (Thao Pham et al., 2008), Indonesia (Permadi et al., 2017), and Nepal (Jayarathne et al., 2018; Sadavarte et al., 2019). However, these regional and national emission inventories in Asia are available for a limited period, with data of the past missing.

The authors of this study have been devoted in developing the REAS series. First version of REAS (REASv1.1) were developed by Ohara et al. (2007), which accounted for actual emissions during 1980-2003 and projected ones in 2010 and 2020. Kurokawa et al. (2013) updated the inventory in REASv2.1, which focused on the period between 2000-2008 when emissions in China drastically increased. REASv2.1 is used as the default data of the MIX inventory. In this study, a long historical emission inventory in the Asian region from 1950-2015 has been newly developed as REAS version 3.1 (REASv3.1). This study provides methodology, results and discussion of REASv3.1. Section 2 gives the basic methodology, including collecting activity data, settings of emission factors and removal efficiencies, and spatial and temporal allocation of emissions to create monthly gridded data sets of REASv3.1. In Section 3.1, trends in air pollutants emissions in Asia are described in detail. Spatial and temporal distributions are overviewed in Section 3.2. Section 3.3 compares the results of REASv3.1 with other emission inventories. Uncertainties of REASv3.1 are discussed in Section 3.4. Finally, summary and remarks are presented in Section 4.

2 Methodology and data

2.1 General description

Table 1 summarizes the general information of REASv3.1. REASv3.1 focuses on the long historical trends of air pollutants emissions in Asia. Target species include the following major air and climate pollutants: SO_2, NO_x, CO, non-methane volatile organic compounds (NMVOC), NH_3, PM_{10}, $PM_{2.5}$, BC, organic carbon (OC), and CO_2. CH_4, and N_2O that were included in REASv2.1 are not in the scope of this version. CH_4 is one of important components of SLCP and will be considered in the next version. The target years are from 1950 to 2015. The start year was chosen to be 1950 as severe air pollution in Japan started from mid-1950s. Figure 1 shows the inventory domain of REASv3.1 which includes East, Southeast, and South Asia. Central Asia and the Asian part of Russia, which were target areas of REASv2.1 are not included in REASv3 because of the difficulty in collecting necessary data for estimating long historical emissions in these areas. China, India, and Japan have been divided into 33, 17, and 6 regions, respectively to reduce the uncertainties in the spatial distribution as that of the previous versions. All target countries and sub-regions are listed in Table S1 in the Supplementary material. Spatial resolution is 0.25° × 0.25°, except in the case of large power plants, which are treated as point sources. Temporal resolution is monthly. These spatial and temporal resolution settings are the same as those of REASv2.1, except that more information for grid allocation and temporal distribution have been collected in this version (see Section 2.5).

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The source categories considered in REASv3.1 are the same as those in REASv2.1. Major sources include fuel combustion in power plants, industry, transport, and domestic sectors. Non-combustion sources include industrial process, evaporation (NMVOC), and agricultural activities (NH_3). NO_x emissions from soil as well as from international and domestic aviation and navigation, including fishing ships are exceptions and were not included in REASv3.1. For domestic and fishing ships, emissions were roughly estimated but their results were only used for comparison with other inventories (see Section 3.3).

In the case of Japan, the Republic of Korea, and Taiwan, REASv2.1 relied on results from other projects or officially opened data. However, in order to develop long historical inventories in a consistent way, their emissions were originally estimated in REASv3.1. One exception to this is NMVOC emission from evaporative sources, which was obtained from the Ministry of the Environment of Japan (MOEJ, 2017) and the National Air Pollutants Emission Service of the National Institute of Environmental Research (http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do) for Japan and the Republic of Korea, respectively. Another is NH_3 emissions from agricultural activities. Data of base year (2000 and 2005 for Japan and 2000 for others) were obtained from other research works (see Sect. 2.4).

2.2 Stationary sources

2.2.1 Basic methodology

Emissions from stationary fuel combustion and industrial processes are traditionally calculated using activity data and emission factors, including the effect of control technologies. In order to increase the accuracy of estimation and to analyze the effects of abatement measures, emissions should be calculated using information on technologies related to emission sources as much as possible. In REASv3.1, emissions from stationary combustion and industrial processes are estimated based on the following equation:

$$E = \sum_i \sum_j \sum_{k,l} \{A_{i,j} \times F_{i,j,k,l} \times EF_{i,j,k} \times (1 - R_{i,j,l})\} \quad (1)$$

where, E represents emission, i is the type of activity data, j is the type of sector category, k is the type of technology related to emission factor, l stands for the control technology after emission, A is amount of activity data, EF is the emission factor of each technology, R is the removal efficiency of each technology, and F is the fraction rate of activity data for combination of i, j, k, and l. When SO_2 emissions from combustion sources are estimated using sulfur contents of fuels, $EF_{i,j,k}$ in eq. (1) is calculated, as follows:

$$EF_{i,j,k} = NCV_{i,j} \times S_{i,j} \times (1 - SR_{i,j,k}) \times 2 \quad (2)$$

where, NCV is the net calorific value of fuel, S is the sulfur content of fuel, and SR is the sulfur retention in ash for combination of i, j, and k. 2 is a factor to convert the value of S to SO_2.

Unfortunately, in the case of Asia, information available on emission factors and removal efficiencies is limited. Even though there is information on the introduction rates of technologies both for emission factors and removal efficiencies, they are available independently. Therefore, for most cases, an average of the removal efficiencies is calculated using the values

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of each abatement equipment and its penetration rate. Then, the average removal efficiencies are commonly used to calculate the emission factors of each technology.

2.2.2 Activity data

Fuel consumption is the core activity data of the emission inventory of air pollutants and greenhouse gases. For most countries, the amount of energy consumption for each fuel type and sector was primarily obtained from the International Energy Agency (IEA) World Energy Balances (IEA, 2017). For China, province-level tables in the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017) were used. For countries and regions whose energy data are not included in IEA (2017), fuel consumption data were taken from the United Nations (UN) Energy Statistics Database (UN, 2016) and the UN data, which is a web-based data service of the UN (http://data.un.org/).

One major obstacle in this study was collecting activity data for the entire target period of REASv3.1, that is from 1950-2015. IEA (2017) includes data from Japan during 1960-2015 and those from other countries during 1971-2015; however, for many countries, fuel types and sector categories, the oldest years when data exist are more later than 1971. Furthermore, past data for sectors do not contain as many categories. For example, coal consumption data in detailed sub-categories of the industrial sector existed in Indonesia only after 2000, but corresponding data are only available for industry total before 1999. In this case, relative ratios of fuel consumption in detailed sub-categories to total industry in 2000 were used to distribute the total industry data to each sub-category for the years before 1999. This procedure is performed for similar cases for all sectors and sometimes for total final consumption. In cases where data did not exist beyond a certain year, fuel consumption data were extrapolated using trends of related data for each sub-category. For example, power generation, amount of industrial products, and population were used to observe trends of fuel consumption in power plant, each industry's sub-category, and residential sectors, respectively. Data for long historical trends were obtained from a variety of sources. For example, power generation data were obtained from Mitchell (1998) and population data were taken from UN (2018). Sources for industrial production are described toward the end of this section. For China, data of CESY for each province were available from 1985 to 2015. During 1950-1984, first, total energy data in China were developed based on IEA (2017) and then, fuel consumption in each province was extrapolated using the total data of China in each fuel type and sector category. For countries which used Energy Statistics Database, fuel consumption of each fuel and sector was taken from the UN data (available at http://data.un.org/Default.aspx) for the period between 1990-2015 and was extrapolated using the trend of total consumption of each fuel type obtained from the UN Energy Statistics Database.

As described in Section 2.1, India and Japan have 17 and 6 sub-regions, respectively. Therefore, for them, country total data of IEA (2017) need to be divided for each sub-region. For Japan, energy consumption statistics of each prefecture that were obtained from the Agency for Natural Resources and Energy (available at https://www.enecho.meti.go.jp/statistics/energy_consumption/ec002/results.html) were used as default weighting factors to allocate country total data to the six regions. Similarly, for India, default weighting factors for regional allocation were estimated from TERI (The Energy and Resources Institute) Energy & Environment Data Diary and Yearbook (TERI, 2013,

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2018), Annual Survey of Industries (Ministry of Statistics & Programme Implementation, available at http://www.csoisw.gov.in/cms/en/1023-annual-survey-of-industries.aspx), and Yevich and Logan (2003), among others. In general, details of these weighting factors are less than those of the country's total fuel consumption. In addition, these data are not available for all the years during 1950-2015. Therefore, regional allocation factors for some sectors were developed independently if corresponding proxy data were available. For the power plant sector, generation capacities of each region and year were calculated as proxy data using the World Electric Power Plants Database (WEPP) (Platts, 2018). For India, traffic volumes (see Section 2.3.1) and amount of industrial production in each region (see the last paragraph of this section) were used as proxy data.

Similar to REASv2.1, large power plants are treated as point sources in REASv3.1 and are updated based on REASv2.1 database. Before 2007, power plants that were classified as point sources were the same as those in REASv2.1 and their information, such as generating capacities, and start and retire years were updated using WEPP. During 2000 to 2007, fuel consumption data were the same as that in REASv2.1. In REASv3.1, power plants whose start years were after 2007 and generation capacities were larger than 300 MW were added as new point sources. Fuel consumption of new power plants were estimated based on relations between fuel consumption amounts and generation capacities of the point data in REASv2.1. If the (A) total fuel consumption of each power plant in a country is larger than (B) the corresponding data in power plant sector, values of each power plant were adjusted by ratios of (B) per (A). If (B) was larger than (A), differences between (B) and (A) were treated as data of area sources.

For emissions from industrial processes, activity data included amount of industrial products. Corresponding data were mainly obtained from related international statistics and national statistics. For example, iron and steel production data were taken from Steel Statistical Yearbook (World Steel Association, 1978-2016) and data for non-ferrous metals and non-metallic minerals were obtained from the United States Geological Survey (USGS) Minerals Yearbook (USGS, 1994-2015). Brick production data were obtained from a variety of sources, such as Zhang (1997), Maithel (2013), Klimont et al. (2017), and the UN data. For China and India, the authors also used internet database services, namely China Data Online (https://www.china-data-online.com) and Indiastat (https://www.indiastat.com/), respectively, which provided both national and regional statistics. The USGS Minerals Yearbook (USGS, 1994-2015) also provided information on plants in each sub-region of China, India, and Japan. Data in the aforementioned statistics were not available for the early years of the target period of REASv3.1. In such cases, data of Mitchell (1998) were used as factors to extrapolate the activity data until 1950.

2.2.3 Emission factors

Setting up of emission factors and removal efficiencies for stationary combustion and industrial processes are difficult procedures, especially for a long historical emission inventory. In this study, emission factors without effects of abatement measures were set, which were used for the entire target period of REASv3.1. Then, effects of control measures were set considering their temporal variations, both for abatement measures before emissions such as using low sulfur fuels and low NO_x burners and those after emissions such as flue gas desulfurization (FGD) and electrostatic precipitator (ESP). These

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settings were done for each country and region based on country and region-specific information. However, such information is still limited, especially in the Asian region. Therefore, default values of unabated emission factors were selected and default removal efficiencies were set to zero. Then, these default values were updated in case information and literature on each country and region were available. For default emission factors, a majority of settings was continuously used from REASv2.1, but some of them, including effects to control measures (net emission factors) were changed to unabated emission factors. Default emission factors were mainly obtained from Kato and Akimoto (1992) for SO_2 and NO_x; Bond et al. (2004), Kupiainen and Klimont (2004), and Klimont et al. (2002, 2017) for PM species; the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006) for CO_2; and the AP-42 (US EPA, 1995), the Global Atmospheric Pollution Forum Air Pollutant Emission Inventory Manual (SEI, 2012), Shrestha et al. (2013), the EMEP/EEA emission inventory guidebook 2009 (EEA, 2016), and other literatures for others.

For country and region-specific settings, in addition to literatures used in REASv2.1 (see Kurokawa et al., 2013), new information, especially for technologies related to settings of emission factors and removal efficiencies was surveyed. Although such information is still limited in Asia, the volume of accessible information on China is relatively large. General information on China in recent years was mainly obtained from Li et al. (2017b) and Zheng (2018). Introduction rates of technologies were obtained from Hua et al. (2016) for cement, Wu et al. (2017) for iron and steel, Huo et al. (2012a) for coke ovens, and Zhao et al. (2013, 2014, and 2015) for a variety of sources. For India, information for technology settings was mainly taken from Sadavarte and Venkataraman (2014), Pandey et al. (2014), Guttikunda and Jawahar (2014), and Reddy and Venkataraman (2002a). For power plants, WEPP database has elements for installed equipment to control SO_2, NO_x, and PM which were used for settings of emission factors and removal efficiencies of power plants treated as point sources. However, these data are not available for most power plants, especially in Asia. Therefore, in the case of South and Southeast Asia, a variety of literatures, such as Sloss (2012) and UN Environment (2017, 2018) were referred to, to set emission factors and removal efficiencies. For Japan, introduction of control technologies for air pollutants were initiated earlier than other countries in Asia. A lot of domestic reports for air pollution and control technologies in power and industry plants published in Japanese, such as MRI (2015), Simoda (2016), Suzuki (1990), and Goto (1981) were referred to, to determine emission factors, removal efficiencies, and their temporal variations.

2.3 Road transport

2.3.1 Basic methodology

Methodology for road transport sector is the same as that of REASv2.1. Equations to estimate hot and cold start emissions (except for SO_2 and CO_2) are, as follows:

$$E_{HOT} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi}\} \quad (3)$$

where, E_{HOT} is the hot emission, i is the vehicle type, NV is the number of vehicles in operation, ADT is the annual distance traveled, and EF_{HOT} is the emission factor. SO_2 emissions are calculated using sulfur contents in gasoline and diesel

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consumed in road transport sector, assuming sulfur retention in ash is zero. CO_2 emissions are estimated by calculating the consumption amounts of fuels (gasoline, diesel, liquefied petroleum gas, and natural gas) and the corresponding emission factors (IPCC, 2006).

Cold start emissions (E_{COLD}) are estimated for NO_x, CO, PM_{10}, $PM_{2.5}$, BC, OC, and NMVOC using the following equation:

$$E_{COLD} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi} \times \beta_i(T) \times F_i(T)\} \quad (4)$$

where, β is the fraction of distance traveled driven with a cold engine or with the catalyst operating below the light-off temperature, and F is the correction factor of EF_{HOT} for cold start emission. β and F are functions of temperature T and are estimated based on EEA (2016). For Japan, the ratio of cold start and hot emissions for each vehicle type was estimated from the JEI-DB. Furthermore, cold start emissions were calculated by hot emissions and the ratios for each vehicle type.

For evaporation from gasoline vehicles, emissions (E_{EVP}) were estimated using the following equation of Tier 1 of EEA (2016):

$$E_{EVP} = \sum_i \{NV_i \times EF_{EVPi}(T)\} \quad (5)$$

where, EF_{EVP} is the emission factor as a function of temperature. For Japan, evaporative emissions in 2000, 2005, and 2010 were obtained from the JEI-DB and those between 2000 (2005) and 2005 (2010) were interpolated. For emissions before 2000 and after 2010, emissions from running loss were extrapolated using trends of traffic volume and those from hot soak loss and diurnal breaking loss were extrapolated by trends of vehicle numbers.

2.3.2 Activity data

Basic activity data of road transport sector include number of vehicles in operation for each type. Data on the registered number of vehicles are available in the national statistics of each country and the World Road Statistics (IRF, 1990-2018). If these statistics did not contain data until 1950, the numbers were extrapolated using trends of data for aggregated vehicle categories in Mitchell (1998). For China, data for each sub-region were obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986-2016) and the China Data Online. Those for India were taken from Road Transport Yearbook (Morth, 2003-2017) and the Indiastat. A problem that was encountered was that registered vehicles were not always in operation. For India, the number of vehicles obtained as registered vehicles were corrected based on Baidya and Borken-Kleefeld (2009), Pandey and Venkataraman (2014), and Prakash and Habib (2018). For other countries, the number of registered vehicles were considered as those in operation due to lack of information. In addition, to estimate emissions, these numbers must be further divided into vehicles based on each fuel type. However, such information is not easily available in national statistics. In this study, settings of REASv2.1 were used as default and were updated if new information was available, such as Pandey and Venkataraman (2014), Sahu et al. (2014) and Mishra and Goyal (2014). If the number of LPG and CNG vehicles were available only for recent years, data were extrapolated using amounts of fuel consumption in road transport sector in IEA (2017).

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Emission factors of road transport sector used in this study were given as emission amounts per traffic volumes. Therefore, annual vehicles kilometer traveled (VKT) per each vehicle type need to be set for each country. We used data of Clean Air Asia (2012) for many countries. Clean Air Asia (2012) includes data for China and India, but data of China were estimated based on Huo et al. (2012b) and those of India were set after Prakash and Habib (2018) and Pandey and Venkataraman (2014). For Japan, the total annual VKT for detailed vehicle types were obtained from reports of Pollutants Release and Transfer Register published by the Ministry of Economy Trade and Industry until 2001 (METI, 2003-2017), which was originally estimated from Road Transport Census of Japan developed by the Ministry of Land, Infrastructure, Transport and Tourism. Before 2001, the total annual VKT was extrapolated using data of more aggregated vehicle categories in the Annual Report of Road Statistics (MLIT, 1961-2016) until 1960 and from the Historical Statistics of Japan (Japan Statistical Association, 2006) until 1950.

2.3.3 Emission factors

For most countries, road transport is one of major causes of air pollution. In Asian countries, vehicle emission standards were introduced after the late 1990s and were strengthened in phases (Clean Air Asia, 2014). Therefore, for road vehicles, year-to-year variation of emission factors must be taken into considered for a long historical emission inventory. In this study, trends in emission factors were estimated considering the timings of road vehicle regulations in each country and the ratios of vehicle production years. Moreover, emission factors of each vehicle type in selected base years were estimated. Finally, emission factors for the entire target period of REASv3.1 were calculated. For year-to-year variations of emission factors, ratios of vehicles based on annual production for each country were estimated using data on the age of vehicles from literatures, such as Zhang et al. (2016), Pandey and Venkataraman (2014), and Huo et al. (2012b). Moreover, emission factors for each production year were selected from the values of Europe and the United States standards and were based on regulation schedules of the target country. Then, tentative emission factors of the target country for each year were estimated during the target period of REASv3.1 using ratios of vehicles' production years as weighting factors. Finally, trends in emission factors from base years were calculated using the calculated tentative emission factors. For most countries, the years just before the regulations for road vehicles began were set as base years and no-controlled emission factors that were used in REASv2.1 were adopted for emission factors of the base years. Countries for which information on regulations were not obtained, the no-controlled emission factors were used for the entire target period of REASv3.1. For China and India, emission factors in 2010 were estimated to be base year's data using setting of REASv2.1 and recently published papers, such as Huo et al. (2012b), Xia et al. (2016), Mishra and Goyal (2014), and Sahu et al. (2014). For the Republic of Korea and Taiwan whose emissions were not originally estimated in REASv2.1, tentative emission factors calculated by the above procedures were used. For Japan, emission factors for each emission standard are available for several vehicle speeds (JPEC, 2012a). Combining these data with information for annual VKT of each vehicle speed, ratios of vehicle ages, and time series of regulation standards, emission of road transport in Japan were calculated.

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2.4 Agricultural sources

REASv3 includes NH_3 emissions from manure management and fertilizer application in agricultural sources. Approaches similar to REASv2.1 were adopted to estimate historical emissions and develop monthly gridded data. First, annual emissions of each country and sub-region except for Japan and their gridded data for the year 2000 were selected from REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) as base data. For Japan, corresponding base data were obtained from REASv2.1 (Kurokawa et al., 2013: JPEC 2012a, b, c; 2014) for the year 2000 and 2005. Second, trends of emissions during 1950-2015 were estimated for each country and sub-region. Third, annual emissions for the period were calculated using the trends and base data. Fourth, changes in spatial distribution from base years to target years and monthly variations in each country and sub-region were estimated. Finally, monthly gridded data of emissions were developed for 1950-2015. For Japan, emission data during 2001-2004 were interpolated between those in 2000 and 2005. Details for manure management and fertilizer application are described in Sections 2.4.1 and 2.4.2, respectively.

2.4.1 Manure management

Trends in NH_3 emissions from manure management of livestock, except for its application as fertilizer, were estimated based on the Tier 1 method of EEA (2016). In this method, emissions are calculated based on the numbers of livestock and the corresponding emission factors. Statistics on the number of animals, such as broilers, dairy cow, and swine are mainly obtained from FAOSTAT (available at http://www.fao.org/faostat/en) of the Food and Agriculture Organization (FAO) of the United Nations from the period between 1961 to 2015. For the years before 1960, data were obtained from Mitchell (1998). National statistics were surveyed for data on provinces, states, and prefectures in China, India, and Japan, respectively to develop activity data for each sub-region. Emission factors are obtained from EEA (2016). For spatial distribution, changes in grid allocation for each country and sub-region from the year 2000 were estimated using EDGARv4.3.2 from 1970 to 2012. Grid allocation factors in 1970 and 2012 were used for the period before and after 1970 and 2012, respectively. For temporal variations, monthly allocation factors are estimated as a function of temperature by referring to the monthly variations of emissions in Japan based on the JEI-DB.

2.4.2 Fertilizer

In most countries, fertilizer application is the largest source of NH_3 emissions. Emission trends after the application of manure and synthetic N fertilizer were estimated using EEA (2016). Manure application is one of the processes of manure management whose emissions trend was calculated based on the number of animals and the corresponding emission factor. For synthetic N fertilizer, trends of total consumption of fertilizer were used in REASv2.1. However, this simple approach causes uncertainties because emission factors are different among types of fertilizer (EEA, 2016). Therefore, in REASv3.1, emissions from each N fertilizer, such as ammonium phosphate and urea were estimated separately and trends in total emissions were calculated. For special distribution, changes in grid allocation factors for each country and sub-region from

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the year 2000 were estimated using a historical global N fertilizer application map during 1961-2010, developed by Nishina et al. (2017). Data for 1961 and 2010 were used for the period before 1961 and after 2010, respectively. For seasonal variations, monthly factors of China and Japan were determined based on Kang et al. (2016) and the JEI-DB, respectively. For other countries, data from Nishina et al. (2017) have monthly application amounts in each grid. However, there are cases that some months have high factors, whereas the others have almost zero. Referring to Janssens-Maenhout et al. (2015), we adopted the conservative way, such that the highest monthly factor was set at 0.2 and the factors of all months were adjusted accordingly.

2.5 Other sources

NMVOC emissions from evaporative sources are increasing significantly in Asia along with economic growth. Major sources of NMVOC emissions include usage of solvents for dry cleaning, degreasing operations, and adhesive application as well as for paint use. Fugitive emissions related to fossil fuels, such as extraction and handling of oil and gas, oil refinery, and gasoline stations are also important. However, statistics on activity data and information of emission factors for these sources are often less available than those for fuel combustion and industrial processes. In this study, default activity data and emission factors were obtained from REASv2.1 and were updated if information was available in recently published papers (Wei et al. (2011) for China and Sharma et al. (2015) for India). In general, activity data of the past years are not available, and, in such cases, proxy data are prepared for trend factors. For example, population numbers were used for dry cleaning and production numbers of vehicles were used for paint application for automobile manufacturing. GDP was used for default trend factors. For emission calculation, the same equation for stationary combustion was adopted.

In addition to agricultural activities, latrines are an important source of NH_3, especially in rural areas. Activity data are population numbers in no sewage service areas estimated referring settings of REASv2.1 and emission factor were based on EEA (2016) and SEI (2012). Also, humans themselves are sources of NH_3 emissions through perspiration and respiration. For these sources, population numbers are activity data and emission factors are obtained from EEA (2016). Equation to estimate emission is also the same as that of stationary combustion.

In REASv3.1, aviation and ship emissions are not included, but emissions of fuel combustion in other transport sector (except for aviation, navigation, and road), such as railway and pipeline transport were estimated. Also, emissions from domestic shipping including fishing ships were roughly calculated for comparison with other inventories (see Section 3.3). Equation (1) is also used for estimating emissions of these sources.

2.6 Spatial and temporal distribution

Procedures for developing gridded emission data were the same as those of REASv2.1. Large power plants were treated as point sources, and longitude and latitude of each power plant were provided. Positions of power plants were surveyed based on detailed information, such as names of units, plants, and companies from WEPP (Platts, 2018). These were searched on internet sites, such as Industry About (available at https://www.industryabout.com/) and Global Energy Observatory

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(http://globalenergyobservatory.org/). Positions for newly added power plants in REASv3.1 as well as those in REASv2.1 were surveyed because some of these services were not available when REASv2.1 was developed. For cement, iron, and steel plants (and non-ferrous metal plants in Japan), REASv3.1 still did not treat them as point sources due to lack of activity data. However, positions, production capacities, start and retire years for large plants were surveyed similar to power plants and used for developing allocation factors for corresponding sub-sectors. For road transport sector, REASv2.1 used coarse grid allocation data of REASv1.1 with 0.5° × 0.5° resolution. Therefore, in REASv3.1, grid allocation factors for each country and sub-region, except Japan, were updated using gridded emission data of road transport sector of EDGARv4.3.2 during 1970-2012. Before 1970 (after 2012), data for 1970 (2012) were used. For Japan, gridded emission data of the JEI-DB in 2000, 2005, and 2010 were used to develop grid allocation factors. For the year between 2000 (2005) and 2005 (2010), the JEI-DB data were interpolated. For years before 2000 (after 2010), the JEI-DB data for 2000 (2010) were used. For residential sectors, rural, urban, and total population of HYDE 3.2.1 (Klein Goldewijk et al., 2017) with 5′ × 5′ were used to create allocation factors. Data of HYDE 3.2.1 were available for 1950, 1960, 1970, 1980, 1990, 2000, 2005, 2010, and 2015 and the years between them were interpolated. Spatial distributions of total population were used for grid allocation of all other sources.

Methodology to estimate monthly emission data in REASv3.1 was the same as that of REASv2.1. In general, monthly emissions were estimated by allocating annual emissions to each month using monthly proxy index. Monthly generated power and production amounts of industrial products were used as the monthly allocation factors for power plant sector and corresponding industry sub-sectors, respectively. Basically, monthly factors of REASv2.1 during 2000-2008 were also used in REASv3.1 and were extended if data existed before (after) 2000 (2008). For the years where surrogate data were unavailable, the data of oldest (newest) year were used before (after) the year. For brick production, monthly allocation factors for Southeast and South Asian countries were estimated using Maithel et al. (2012) and Maithel (2013). For the residential sector, monthly variations of emissions were estimated using surface temperature in each grid cell, similar to REASv2.1. Surface temperatures during 1950-2015 were taken from NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (available at https://www.esrl.noaa.gov/psd/). For Thailand and Japan, most monthly factors were set based on country specific information from Thao Pham et al. (2008) and JPEC (2014), respectively.

3 Results and discussion

3.1 Trends of Asian and national emissions

Trends in air pollutants emissions from Asia, China, India, Japan, and other countries are described in this section, mainly focusing on SO_2, NO_x, and BC emissions as they have important roles in both air pollution and climate change. SO_2 and NO_x are precursors of sulfate and nitrate aerosols, respectively, which are the major components of secondary $PM_{2.5}$. NO_x is also a precursor of ozone. Furthermore, BC is a major component of primary $PM_{2.5}$. $PM_{2.5}$ and ozone not only harm human health

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and ecosystems, but influence climate change. BC and ozone have a warming effect on climate change, whereas sulfate and nitrate aerosols have a cooling effect. Note that all the air pollutant emissions from each country between 1950 to 2015 categorized based on major sectors and fuel types, are provided in the Supplement material (Figs. S1-S12).

3.1.1 Asia

Table 2 summarizes the national emissions of each species in 2015 and the total emissions from Asia in 1950, 1960, 1970, 1980, 1990, 2000, and from 2010-2015. Figure 2 shows emissions of SO_2, NO_x, CO, NMVOC, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC in China, India, Japan, Southeast Asia, East Asia other than China and Japan, and South Asia other than India from 1950 to 2015. Average total emissions in Asia during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO_2: 3.15 Tg, 42.4 Tg (13.5); NO_x: 1.83 Tg, 47.6 Tg (26.0); CO: 62.2 Tg, 319 Tg (5.13); NMVOC: 9.14 Tg, 61.8 Tg (6.77); NH_3: 7.99 Tg, 31.3 Tg (3.92); CO_2: 1.12 Pg, 18.3 Pg (16.3); PM_{10}: 5.76 Tg, 28.4 Tg (4.92); $PM_{2.5}$: 4.52 Tg, 20.3 Tg (4.50); BC 0.751 Tg, 3.38 Tg (4.51); and OC 2.62 Tg, 6.92 Tg (2.64). Clearly, all the air pollutant emissions in Asia increased significantly during these six decades. However, this increase was different among the aforementioned species. Growth rates of emissions were relatively large for SO_2, NO_x, and CO_2 because the major sources of these species are power plants, industries, and road transport, for which fuel consumption increased significantly along with economic development in Asia. SO_2 increased before the other species because a majority of the emissions were obtained from the combustion of coal, which is easier to obtain than oil and gas. SO_2, NO_x, and CO_2 emissions increased keenly in the early 2000s, along with rapid growth of emissions of these species in China. For NO_x, combustion of oil fuels, especially by road vehicles, contributed to a large growth of emissions in the latter half of 1950-2015. Growth rates of NMVOC have also increased recently due to an increase in the emissions from road vehicles and evaporative sources, such as paint and solvent usage in accordance with economic growth of Asian countries. On the other hand, rates of growth of CO, PM_{10}, $PM_{2.5}$, BC, and OC are relatively small. One reason is that emissions of these species are mainly from incomplete combustion in low temperature and thus, emissions from power plants and large industry plants are relatively small. Another reason is that a major source of these species is the combustion of coal and biofuels in residential sector, which were relatively large even in past years in Asia. Recently, emissions of these species from industries, including combustion and non-combustion processes are increasing. In addition, gasoline and diesel vehicles have contributed recently to the growth of CO and BC emissions, respectively. Agricultural activities, such as manure management of livestock and fertilizer application, which are major sources of NH_3 are rising to support a growing population in Asia. Although the growth rate of NH_3 emissions is smaller than other species, it still shows an increasing trend.

Differences in the trends of emissions were also observed on the basis of countries and regions. SO_2 and NO_x, emissions from Japan were relatively large in Asia during 1950s-1970s. Emissions from Japan in 1965 are comparable with and are larger than those of China for SO_2 and NO_x, respectively. In 2015, emissions of SO_2 and NO_x in Japan decreased largely and contribute only about 1.5 and 4% of Asia's total emissions, respectively. Similar tendencies were also observed in the case of other species. In 2015, China was the largest contributor of emissions for all the species. Recently, emissions of most species

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in China have shown decreasing or stable trends. In the case of SO_2, China contributed about 72% of emissions in 2005, but about 48% in 2015. On the other hand, emissions and their relative ratios are increasing in the case of India. Actually, contribution rates of SO_2, NO_x, and BC emissions in India increased from 14%, 17%, and 24% in 2005 to 31%, 23%, and 28% in 2015, respectively. Li et al. (2017c) suggested that, in 2016, SO_2 emissions in India exceeded those in China. Recent increase in air pollutants emissions have also been observed in Southeast Asia and South Asia other than India. Furthermore, emissions from East Asia other than China and Japan started to increase slightly later than Japan and then, recently show decreasing trends mainly reflecting trends of emissions from Republic of Korea and Taiwan.

3.1.2 China

Air pollutants emissions in China averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are, as follows: SO_2 1.10 Tg, 24.6 Tg (22.4); NO_x 0.544 Tg, 26.6 Tg (48.8); CO 27.2 Tg, 188 Tg (6.90); NMVOC 2.73 Tg, 27.8 Tg (10.2); NH_3 2.94 Tg, 14.0 Tg (4.74); CO_2 0.414 Pg, 11.4 Pg (27.5); PM_{10} 2.46 Tg, 16.8 Tg (6.81); $PM_{2.5}$ 1.96 Tg, 12.0 Tg (6.10); BC 0.316 Tg, 1.76 Tg (5.55); and OC 1.10 Tg, 2.93 Tg (2.65). Emission trends in China for all the pollutants in each sector and for each fuel type during 1950-2015 were presented in Figs. S1 and S2, respectively. It was observed that emissions of all pollutants increased largely during these six decades, but most species reached their peaks up to 2015. Exceptions to this were NMVOC, NH_3, and CO_2; however, their growth rates are at least small or almost zero.

Figure 3 shows recent trends in actual emissions (solid colored areas) and reduced emissions by control measures (hatched areas) from each sector for SO_2, NO_x, and BC during 1990-2015 in China. The reduced emission by control measures was the difference between emissions calculated without effects of all control measures (such as FGD, ESP, using low sulfur fuels, regulated vehicles, etc.) and actual emissions. Note that reduced emissions here do not include effects of substitution of fuel types, such as from coal to natural gas. SO_2 emissions in China increased rapidly in early 2000s, but decreased after 2006 and showed a continuous decline until 2015. Drastic changes in 2000s were mainly caused by emissions from coal-fired power plants, which increased rapidly along with large economic growth and later decreased due to the introduction of FGD based on the 11th Five Year Plan of China. After 2011, control measures for large industry plants started to become effective and as a result, total emissions in 2015 became comparable with those in 1990. Similar to SO_2, NO_x emissions increased rapidly from the early 2000s, but continued to increase until 2011 and then, started to decline. In the 2000s, low NO_x burner to power plants and regulation of road vehicles were introduced, but their effects were limited. From 2011, introduction of denitrification technologies, such as selective catalytic reduction (SCR) to large power plants and regulations for road vehicles were strengthened based on the 12th Five Year Plan of China. As a result, in 2015, NO_x emissions were about 83% of their peak values in 2011. For BC, emissions also increased from early 2000s, but growth rates were smaller than SO_2 and NO_x due to the effects of control equipment in the industrial sector. Another reason could be that BC emission factors for coal-fired power plants are originally low. Recently, BC emissions from residential sector as well as industrial sector show decreasing trends. In this study, the reductions in BC emissions in residential sector were mainly caused by a

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decrease in emissions from biofuel combustion. During 2010 to 2015, consumptions of primary solid biofuels were reduced about 30%, whereas consumption of natural gas and liquefied petroleum gas increased about 60% in the residential sector.

For CO, most emissions in 1950s were from residential sectors, which gradually increased with increasing coal consumption in the industrial sector. CO emissions increased largely in 2000s due to coal combustion and iron and steel production processes. Recently, CO emissions have seen a decline. A major reason for this declining trend is the decrease in biofuel consumption in residential sector and the phasing out of shaft kiln with high CO emission factor in the cement industry. NMVOC emissions increased significantly from the early 2000s, similar to other species. However, their major sources were different from others. Recent increasing trends are not caused by stationary combustion sources, but by road transport and evaporative sources, such as paint and solvent use. Growth rates of NMVOC emissions tended to slow down around 2015, but emissions increased almost monotonically after the 2000s. NH_3 emissions were mostly from agricultural activities. In China, emissions from fertilizer application showed a significant increase from early 1970s to early 2000s. In recent years, NH_3 emissions are almost stable. For PM_{10} and $PM_{2.5}$, majority of the emissions are from the industrial sector, followed by residential sector and power plants. Emissions increased largely from the early 1990s mainly due to coal combustion and industrial processes, especially in cement plants. Compared to SO_2 and NO_x, growth rates of PM_{10} and $PM_{2.5}$ emissions during the early 2000s were small, and later decreased due to the effects of control equipment in industrial plants. OC emissions were mostly from biofuel combustion in the residential sector. Contributions from the industrial sector has been increasing recently, but total OC emissions have decreased due to reduced usage of biofuels. CO_2 emissions were mainly controlled by coal combustion and their trend were similar to those of NO_x until 2011. After 2011, CO_2 emissions in China were found to be almost stable.

3.1.3 India

Emissions of air pollutants in India averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO_2 0.541 Tg, 10.5 Tg (19.5); NO_x 0.484 Tg, 9.62 Tg (19.9); CO 16.1 Tg, 62.9 Tg (3.91); NMVOC 2.86 Tg, 14.3 Tg (4.99); NH_3 2.95 Tg, 9.26 Tg (3.14); CO_2 0.299 Pg, 2.64 Pg (8.82); PM_{10} 1.40 Tg, 6.32 Tg (4.50); $PM_{2.5}$ 1.17 Tg, 4.57 Tg (3.91); BC 0.203 Tg, 0.903 Tg (4.45); and OC 0.701 Tg, 1.98 Tg (2.83). Figures S3 and S4 provide trends of emissions in India from each sector and fuel type for all the pollutants, respectively, from 1950 to 2015. In general, all the air pollutants show monotonous increase from 1950 to 2015 and growth rates (especially of recent years) are larger for SO_2, NO_x, NMVOC, and CO_2, which is similar to the case of Asia.

Figure 4 shows trends in emission of SO_2, NO_x, and BC from each fuel type as well as sector during 1950-2015 in India. Clear differences were seen in the structure of emissions in these species. For SO_2, large parts of emissions were from coal combustion in power plants and industry sector. Recently, a rapid increase in SO_2 emission was mainly from coal-fired power plants. For NO_x, emissions trends were close to those of SO_2 and contributions from coal-fired power plants were also large. However, contribution and growth rates from road transport especially diesel vehicles were almost comparable with those of power plants for NO_x. For BC, contributions from the residential sector and biofuel combustion were large,

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especially in 1950s-1960s. Contribution rates of residential sector were 76% in 1950 and 47% in 2015, and those of biofuel combustion, which were mainly used in residential sector and some parts are used in industry sector were 87% in 1950 and 53% in 2015. However, recent increasing trends of BC emissions were caused by growth of emissions from diesel vehicles and industry sector. For recent trends, relative ratios of SO_2 emissions from power plants were increased from 43% to 60% during 1990-2015. For NO_x, contribution rates from both power plants and road transport were increased and accounted for about 80% of the total emissions in 2015. Even in 2015, about half of the BC emissions were from the residential sector. However, as previously described, recent emission growths were mainly caused by the industrial sector and road transport. These tendencies were similar to Japan and China during their rapid emission growth periods.

Trends and structure of CO emissions were similar to those of BC but contribution rates of the residential sector were larger and those from road transport (mainly from gasoline vehicle) were smaller, as compared to BC. This tendency was also found in OC; however, relative ratios of emissions from residential sector were much larger (about 75% in 2015) and those of road transport were much smaller. For PM_{10} and $PM_{2.5}$, a majority of the emissions was from residential and industrial sectors. Both amounts were almost comparable in PM_{10} and those from residential sectors were larger in $PM_{2.5}$. Different from BC and OC, contributions from power plants exist in PM_{10} and $PM_{2.5}$ whose contribution rates in 2015 are about 22% and 17%, respectively. For NMVOC, most emissions were from biofuel combustion before 1980s. Later, emissions from variety of sources, such as road transport, extraction and handling of fossil fuels, usage of paint and solvents are increasing and are controlling recent trends. Most NH_3 emissions are from agricultural activities. Contributions from manure management and fertilizer use were comparable before 1980s. However, emissions from fertilizer application have increased largely which are now determining recent trends. For CO_2, trends during 1950-2015 were similar to SO_2 and NO_x, which show rapid growth after the 2000s mainly due to increasing consumption of fossil fuels.

3.1.4 Japan

Air pollutants emissions in Japan averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO_2 1.23 Tg, 0.601 Tg (0.491); NO_x 0.483 Tg, 2.03 Tg (4.21); CO 4.27 Tg, 4.54 Tg (1.06); NMVOC 0.326 Tg, 0.977 Tg (3.00); NH_3 0.231 Tg, 0.365 Tg (1.58); CO_2 0.178 Pg, 1.30 Pg (7.31); PM_{10} 0.922 Tg, 0.142 Tg (0.154); $PM_{2.5}$ 0.489 Tg, 0.113 Tg (0.232); BC 0.0625 Tg, 0.0196 Tg (0.314); and OC 0.141 Tg, 0.0161 Tg (0.114). Trends in air pollutants emissions from each sector and fuel type during 1950-2015 in Japan were shown in Figs. S5 and S6. Compared to the rest of Asia, emissions of all species in Japan except CO_2 were reduced significantly after reaching peak values. In addition, peak years were mostly 40 years ago (about 1960 for PM_{10}, $PM_{2.5}$, and OC, 1970 for SO_2 and CO, 1980 for NO_x and NH_3, 1990 for BC, and 2000 for NMVOC).

Figure 5, similar to Fig. 3, shows trends of actual emissions (solid colored areas) and reduced emissions by control measures (hatched areas) from each sector for SO_2, NO_x, and BC during 1950-2015. SO_2 emissions, especially from power plants and industry sector increased significantly in the 1960s (reflecting the rapid economic growth) and caused severe air pollutions in Japan. In the 1950s, more than half the emissions were from coal combustion; moreover, contributions from heavy fuel oil

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increased rapidly in 1960s (about 50% around the peak year). In order to mitigate air pollution, first, regulation of sulfur contents, especially in heavy fuel oil, were strengthened. Then, desulfurization equipment was mainly introduced from the mid-1970s. As a result, SO_2 emissions decreased keenly in the 1970s. Although coal consumption in power plants increased in 1990s, SO_2 emissions almost did not change due to these measures. In 2015, about 90% of the SO_2 emissions were reduced by regulatory measures. NO_x emissions also increased rapidly from the 1960s mainly by steep increase of traffic volumes and fossil fuel combustion in power and large industry plants. The largest contribution to NO_x emissions during the peak periods was from road transport sector, that is greater than 50% of total emissions. Regulations for road vehicles became effective from the late 1970s but an increase in the number of vehicles partially cancelled the effects. For stationary sources, the number of introduced denitrification equipment increased largely in the 1990s. As a result, NO_x emissions peaked later; furthermore, reduction rates after the peak were smaller compared to that of SO_2. For BC, contributing sectors changed during 1950-2015. In the 1950s, most emissions were from industries and residential sectors and their amounts were almost comparable. After the 1960s, both types of emissions declined, but their reasons for decline were different. In the 1950s, coal and biofuels, which have large BC emission factors were mainly used in residential sectors. However, these fuels were substituted for cleaner ones, such as natural gas and liquefied petroleum gas which reduced BC emissions significantly. Emissions in industrial sectors decreased gradually after the 1960s due to the introduction of abatement equipment for PM. Instead, emissions from road transport sector increased from the late 1960s to around 1990. Then, regulations for road vehicles were strengthened and BC emissions were reduced largely from peak values.

For CO, NMVOC, and OC, most emissions in 1950s were from biofuel combustion in the residential sector. CO and NMVOC emissions in road transport increased largely in the 1960s and then decreased gradually, similar to the case of NO_x. Recently, a majority of NMVOC emissions were from evaporative sources, such as paint and solvent use. These started to increase from the 1980s and then decreased after 2000. Emissions of CO and OC from the industrial sector showed a similar increase before 1970, whereas OC emissions started to decrease due to control equipment and CO emissions were almost stable after 1970. Majority of NH_3 emissions in Japan were from agricultural activities, especially manure management; however, contributions from latrines were also large in the past years. Overall, NH_3 emissions increased from 1950 to 1970s but, showed slightly decreasing trends after the 1990s. PM_{10} and $PM_{2.5}$ emission trends were almost the same. Majority of the emissions were from the industrial sector, which grew during the 1950s but decreased largely in the 1970s due to the effects of abatement equipment for PM. Contributions from the residential sector were relatively large from 1950s to 1960s. Furthermore, contributions from road transport increased from the 1970s and started to decrease after 1990, similar to BC. For CO_2, emission increased rapidly in 1960s, similar to that of SO_2 and NO_x. After that, CO_2 emissions have generally continued to increase, but growth rates are much smaller than those in the 1960s, thereby reflecting the economic status of Japan.

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545 **3.1.5 Other regions**

Similar to India, air pollutant emissions in Southeast Asia and South Asia other than India (OSA) tended to increase. Emissions in Southeast Asia averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are, as follows: SO_2 0.161 Tg, 4.52 Tg (28.1); NO_x 0.215 Tg, 6.10 Tg (28.3); CO 10.8 Tg, 43.2 Tg (4.00); NMVOC 2.36 Tg, 13.3 Tg (5.61); NH_3 0.784 Tg, 4.09 Tg (5.22); CO_2 0.161 Pg, 1.56 Pg (9.68); PM_{10} 0.680 Tg, 3.07 Tg (4.51); $PM_{2.5}$ 0.636 Tg, 2.28 Tg (3.59); BC
550 0.119 Tg, 0.422 Tg (3.55); and OC 0.486 Tg, 1.28 Tg (2.63); corresponding data in OSA are, as follows: SO_2 0.0602 Tg, 1.41 Tg (23.4); NO_x 0.0713 Tg, 1.26 Tg (17.7); CO 3.52 Tg, 14.5 Tg (4.11); NMVOC 0.783 Tg, 3.66 Tg (4.67); NH_3 0.907 Tg, 3.13 Tg (3.44); CO_2 0.0566 Pg, 0.435 Pg (7.69); PM_{10} 0.256 Tg, 1.69 Tg (6.62); $PM_{2.5}$ 0.240 Tg, 1.14 Tg (4.73); BC 0.0451 Tg, 0.222 Tg (4.93); and OC 0.180 Tg, 0.661 Tg (3.67). Figures S7 and S8 (S9 and S10) provide trends for all the air pollutant emissions in Southeast Asia (in OSA) for each sector and fuel type, respectively, from 1950 to 2015.

555 Figures 6 and 7 show emission trends of SO_2, NO_x, and BC for each sector category and contribution rates of each country from 1950-2015 in Southeast Asia and OSA. Contributing sources and their relative ratios in SO_2, NO_x and BC emissions are generally close between these regions. For both the regions, major sources of SO_2 emissions are power plants and industry sector. For fuel types, contribution from heavy fuel oil was large in the case of SO_2 emissions from OSA and was almost comparable to those of coal from Southeast Asia in the 1990s. After 2010, emissions from coal-fired power plants in
560 Southeast Asia increased rapidly. For NO_x, majority of the emissions were from road transport, mainly diesel vehicles. This controlled the recent trends in both regions. Contributions from gasoline vehicles were small in OSA, but relatively large in Southeast Asia. Recently, NO_x emissions from coal-fired power plants have been increasing, especially in Southeast Asia. Although trends are almost stable, emissions from biofuel combustion in the residential sector are relatively large in OSA. BC emissions are mostly from biofuel combustion in the residential sector, especially in OSA. Recent increasing trends are
565 caused by emissions from industry and road transport sectors. In the case of country-wise emissions, currently, the largest contributing countries are Indonesia and Pakistan in Southeast Asia and OSA, respectively. In 2015, the second and third highest contributing countries in Southeast Asia were Philippines and Vietnam for SO_2, Thailand and Philippines for NO_x, and Vietnam and Thailand for BC. Relative ratios of SO_2 emissions in Thailand were large in the early 1990s but decreased significantly due to the introduction of FGD in large coal-fired power plants. For OSA, the second highest contributing
570 country is Bangladesh; Sri Lanka is ranked third for SO_2 and NO_x and Nepal for BC.

For East Asia other than China and Japan (OEA), all the air pollutant emissions averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are, as follows: SO_2 0.0589 Tg, 0.753 Tg (12.8); NO_x 0.0339 Tg, 2.00 Tg (58.9); CO 0.349 Tg, 6.41 Tg (18.3); NMVOC 0.0828 Tg, 1.88 Tg (22.7); NH_3 0.169 Tg, 0.467 Tg (2.76); CO_2 0.0123 Pg, 0.981 Pg (79.7); PM_{10} 0.0412 Tg, 0.375 Tg (9.11); $PM_{2.5}$ 0.0244 Tg, 0.263 Tg (10.8); BC 0.00529 Tg, 0.0604 Tg (11.4); and OC 0.0109 Tg,
575 0.0592 Tg (5.46). In Figs. S11 and S12, emission trends in OEA from each sector during 1950-2015 were presented for all the air pollutants. Emission trends in the Republic of Korea and Taiwan were similar to those of Japan. SO_2 emissions increased rapidly in the 1970s and reduced largely from their peak values due to the introduction of low sulfur fuels and

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FGD. NO_x emissions started to increase steeply from the 1980s due to the road transport sector, in addition to power and industry plants. Then, NO_x emissions decrease after 2000 due to regulations related to road vehicles and the introduction of control equipment to power plants. However, their rate of decrease was lower than that of SO_2. BC emission trends were similar to those of NO_x until around the year 2000, but the ratio of decrease after 2000 is much larger than that of NO_x. These features are mainly determined by emissions from road vehicles. For Democratic People's Republic of Korea, emissions of SO_2, NO_x, CO_2 and PM decreased and those of CO, NMVOC, and NH_3 were almost stable recently. For Mongolia, emissions of all the air pollutants, except PM, show increasing trends recently. BC and OC emissions also increased after the 2000s but decreased after 2013. Emissions of PM_{10} and $PM_{2.5}$ almost stabilized after 2000. Note that information of these two countries are limited and therefore uncertainties are large.

3.2 Spatial distribution and monthly variation

Figure 8 presents the emission map of SO_2, NO_x, CO, NMVOC, NH_3, $PM_{2.5}$, BC, and OC in 1965 and 2015 at 0.25° × 0.25° resolution. Emission maps of CO_2 and PM_{10} are presented in Fig. S13. In 1965, high emission grids appeared in industrial areas of Japan, especially for NO_x, SO_2, and CO_2. On the other hand, high emission grids are seen in wide areas in China and India, in addition to Japan for CO and PM species, especially OC. This is because emissions of these species are mainly from the residential sector and small industrial plants. In 2015, high emission areas for all species clearly appear in China and India, especially in the northeastern area, around the Sichuan province, and Pearl River Delta for China and Indo-Gangetic Plain, around Gujarat, and southern area for India. High emission areas of SO_2 and PM species in Japan disappeared or shrinked in 2015 compared to 1965, but still remained in the case of NO_x, CO, NMVOC, and CO_2 maps. In Southeast Asia, high emission areas are seen in the Java island of Indonesia and around large cities, such as Bangkok (Thailand) and Hanoi (Vietnam). NH_3 and OC emissions, whose major sources are agriculture and residential sector, respectively are in relatively large areas of China, India, and Southeast Asia.

As described in Section 2.5, seasonality of emissions is taken into considered for sectors where proxy data for monthly profiles are available or can be estimated. Monthly variations of total emissions of SO_2, NO_x, BC, and NH_3 are shown for China, India, Japan, Southeast Asia, OEA, and OSA for the year 2015 in Fig. 9. For SO_2 and NO_x, monthly variations were generally small. In China, emissions were slightly larger in the second half of the year. Monthly factors of SO_2 emissions in OSA were high from December to May and low during July and September due to the timings of brick production. For BC, emissions in winter season were relatively large, especially in China and OEA. This seasonality is determined by fuel consumption in residential sector for the purposes of heating. Therefore, monthly variations of BC emissions are smaller in Southeast Asia. For NH_3, seasonality of emissions is controlled by the seasonality of emissions from fertilizer application and manure management. In China, Japan, and OEA, peaks of emissions appear during summertime. Monthly variations of emissions in the whole of Southeast Asia are small, but seasonality is different from each country. Finally, it must be noted that monthly variations of emissions in each grid are different to each other because they are determined by monthly profiles of major emission sources in each grid cell.

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3.3 Comparison with other inventories

In this section, estimated emissions of REASv3.1 were compared with those of other global, regional, and national inventories. As mentioned in Section 2.1, values of REASv3.1 include emissions from domestic and fishing ships, which are roughly estimated using fuel consumption and default emission factors. This procedure is done because most other inventories include ship emissions and it is difficult to remove their contribution for comparison with REASv3.1. Note that domestic and fishing ships are not officially included in target sources of REASv3.1, as described in Sect. 2.1. Figures 10 and 11 compare the results of REASv3.1 with other studies for SO_2, NO_x, and BC emissions in China and India, respectively. For other species, results based on comparison with China are presented in Fig. S14 and those with India are shown in Fig. S15. Furthermore, Figures S16-S19 provide the comparisons of emissions from Japan, Southeast Asia, East Asia other than China and Japan, and South Asia other than India, respectively.

For long historical trends of SO_2 emissions in China, values of REASv3.1 were similar to those of EDGARv4.3.2 and CEDS, until 1995. After this, EDGARv4.3.2 and CEDS showed a marginal decline, although the values of REASv3.1 were almost constant. SO_2 emissions in most results increased rapidly from the early 2000s and a majority of them decreased from the late 2000s, although the decreasing rates are different. Recent rapid decreasing tendency in REASv3.1 is similar to that of Zheng et al. (2018). Values of REASv3.1 were slightly larger than those of REASv2.1 during 2000-2005, but then reduced due to a larger decreasing rate of REASv3.1. Compared to SO_2, variability of NO_x emissions among estimations plotted in Fig. 10 is small, although values of CEDS are slightly larger than others. NO_x emissions in all the results increased largely in the 2000s and then decreased or stabilized. For BC, there were large discrepancies observed among plotted results in Fig. 10. Trends observed in REASv3.1 and CEDS were similar until early 2000s, but then CEDS showed a large and constant increase. Differences in REASv3.1 and REASv2.1 are small. BC emissions of both REASv3.1 and Zheng et al. (2018) decreased from 2012, but the ratio of decrease was much larger in Zheng et al. (2018). Values of BC emissions of REASv3.1 were larger than those of REASv2.1, especially in the early 2000s, but the difference in 2008 was small. For trends and emission amounts of PM_{10} and $PM_{2.5}$, tendencies of relationships among each result were similar. Growth rates of REASv3.1 were larger than EDGARv4.3.1 until mid-1990s, following which, similar temporal variations were shown in both results until the early 2000s. After the early 2000s, REASv3.1, MEICv1.2, Zhao et al. (2013), and Zheng et al. (2018) show decreasing trends, although REASv2.1, EDGARv4.3.2, and Klimont et al. (2017) continued to increase. For OC, most results decreased from 1995 to 2000 and then increase from the early 2000s. Then, peaks of REASv3.1, REASv2.1, and MEICv1.2 appeared around 2005, whereas EDGARv4.3.2 and CEDS show continuous increasing trends. CO emissions trends were relatively similar among most studies, although values of REASv3.1 were smaller (larger) before the mid-1990s (after middle of 2000s), as compared to other inventories. Emissions and their growth rates of EDGARv4.3.2 were smaller than other results. For NMVOC, emission amounts of REASv3.1 were smaller, but growth rates were larger than CEDS and EDGARv4.3.2 until the early 2000s. After this period, most studies showed significant increasing trends, but emission amounts of REASv3.1 in 2015 were larger than MEICv1.2, Zheng et al. (2018), and Li et al. (2019). Differences among

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studies in terms of NH_3 emissions were large not only in emission amounts, but also in temporal variations. REAS inventories, CEDS, and EDGARv4.3.2 generally showed increasing trends. On the other hand, trends of MEICv1.2 and Zheng et al. (2018) were almost stable after 2000 and the results of Kang et al. (2016) showed decreasing trends after mid-2000s. Emissions of REASv3.1 were also almost stable after 2010.

For SO_2 emissions in India, REASv3.1, EDGARv4.3.2, and Smith et al. (2011) showed similar long historical trends, until 2000. Then, values of REASv3.1 became slightly larger than these, but were close to those of Klimont et al. (2013). CEDS, Streets et al. (2000), and REASv2.1 values were similar but were larger than REASv3.1. On the other hand, emissions of REASv3.1 were larger than the recent national studies by Sadavarte and Venkataraman (2014) and Pandey et al. (2014). For long historical trends of NO_x, emissions of REASv3.1 are much smaller than CEDS and REASv2.1 as well as smaller than those of EDGARv4.3.2 and REASv1.1 until 2000. However, values of REASv3.1 are close to Garg et al. (2001) and those of EDGARv4.3.2 after 2000. Similar to SO_2, NO_x emissions of REASv3.1 are smaller than Sadavarte and Venkataraman (2014) and Pandey et al. (2014). For BC, trends of REASv3.1 and CEDS agree relatively well for all periods. However, as in the case of China, discrepancies among studies plotted in Fig. S15 are large. Trends of REASv1.1 and REASv3.1 showed similar trends but values of REASv3.1 were smaller. On the other hand, emissions of REASv3.1 were larger than EDGARv4.3.2 and REASv2.1 but growth rates of EDGARv4.3.2 after 2008 and REASv2.1 were larger than those of REASv3.1. OC emissions of REASv3.1 were smaller than CEDS and Lu et al. (2011) but growth rates were similar. On the contrary, growth rates of REASv3.1 were larger than EDGARv4.3.2, Pandey and Venkataraman (2014), Sadavarte and Venkataraman (2014), and Klimont et al. (2017). For PM_{10} and $PM_{2.5}$, temporal variations of REASv3.1 and EDGARv4.3.2 were similar until the late 2000s. Growth rates of EDGARv4.3.2 after late 2000s and REASv2.1 were larger than REASv3.1. $PM_{2.5}$ emissions of Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014) were larger than REASv3.1 but growth rates were alike. On the other hand, Klimont et al. (2017) showed different temporal variations. Emissions of PM_{10} and $PM_{2.5}$ decreased from 2000 to 2005 and differences in the values between 2005 and 2010 were marginal. Amounts and trends of CO emissions compared in Fig. S15 generally agree well except for REASv1.1. Values of REASv3.1 are between those of CEDS and EDGARv4.3.2. For NMVOC, emission amounts of REASv3.1 and EDGARv4.3.2 were almost the same in the early 1990s but growth rates after that were higher for REASv3.1 than for EDGARv4.3.2. Values of REASv3.1 were close to Streets et al. (2003a), Zhang et al. (2009), and Venkataraman et al. (2018). Moreover, values of EDGARv4.3.2 agree with Pandey and Venkataraman (2014), Sadavarte and Venkataraman (2014) and Sharma et al. (2015). Emissions of CEDS and REASv2.1 were more than that of other studies. For NH_3, a comparison of the emissions in Fig. S15 show similar trends. Differences in emission amounts are also relatively small, except for EDGARv4.3.2.

Results of SO_2 emissions in Japan compared in Fig. S16 generally agree well except for EDGARv4.3.2 and Streets et al. (2000), whose values were lager and smaller, respectively. For NO_x, emissions, amounts of REASv3.1 were larger than those of most studies, except for CEDS; although temporal variations were found to be similar. Emission amounts and trends of BC were generally alike among the results shown in Fig. S16, except for CEDS. Emissions of BC and OC of CEDS increased almost monotonically until their peak around 1990. These tendencies were much different from REASv3.1. In

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general, most studies show decreasing trends of emissions of PM species from 1990s. However, decreasing ratios of REASv3.1 before 1990s are larger than other studies. For CO emissions, significant increase was observed from the early 1960s in both REASv3.1 and CEDS, but emissions of REASv3.1 reached its peak in the early 1970s while those of CEDS continued to increase until 1980. Emissions of EDGARv4.3.2 decreased largely during the early 1970s. For NMVOC, results of REASv3.1 and CEDS agree well except from the late 1970s to early 1990s and after 2000s. Emission amounts of REASv3.1 and CEDS are smaller than EDGARv4.3.2 and Zhang et al. (2009). Trends of NH_3 emissions shown in Fig. S16 are similar except for EDGARv4.3.2 before the mid-1990s. Emission amounts of REASv3.1 are smaller than Kannari et al. (2001), REASv2.1, and CEDS.

For SO_2 emissions in Southeast Asia, values decreased keenly in CEDS from the late 1990s. A similar feature was seen in REASv3.1 but its rate of decrease was much smaller. After 2000, emissions of REASv3.1 showed increasing trends while those of Klimont et al. (2013) decreased almost monotonically. For NO_x, all the results shown in Fig. S17 agreed well until late 1990s, but showed different trends later. Emissions of REASv3.1 increased almost monotonically from the 1990s, but those of CEDS and EDGARv4.3.2 were stable until 2000 and then increased again. Growth rates of CEDS and EDGARv4.3.2 after 2000 were larger and smaller than REASv3.1, respectively. Trends of emissions in REASv3.1 and REASv2.1 were similar but amounts of REASv3.1 were larger. For BC, all the results in Fig. S17 increased gradually with similar increasing rates. However, emissions of EDGAR became almost stable after 2000 and those of REASv3.1 decreased during the late 2000s and then increased again.

Most results of SO_2 emissions in East Asia other than China and Japan show increasing and decreasing trends from late 1960s and early 1990s, respectively, although amounts in CEDS from 1970 and 2000 were much smaller. On the other hand, emission peaks in EDGAR were reached around 2000 and the amounts were much larger than those observed other studies after the late 1990s. For NO_x emissions, amounts and trends shown in Fig. S18 generally agreed until the late 1990s, but the results differed after the 2000s. Decreasing rates of REASv3.1 were similar to those of EDGARv4.3.2, but smaller than those of CEDS and REASv2.1. BC emissions of REASv3.1 and CEDS show similar increasing trends until the late 1980s. Then, emissions of REASv3.1 decreased almost monotonically, but those of CEDS were almost stable. Both emission amounts were larger than REASv1.1., REASv2.1, and EDGARv4.3.2. For South Asia other than India, trends and amounts of SO_2 emissions were similar. Emission values of Streets et al. (2003a) and Zhang et al. (2009) were larger than the results plotted in Fig. S18. For NO_x, values of REASv3.1, Kato and Akimoto (1992), Streets et al. (2003a) and Zhang et al. (2009) were almost the same and trends of REASv3.1 were similar to those of REASv1.1, REASv2.1, and EDGARv4.3.2. Emission amounts of CEDS were much higher than other results. For BC emissions, trends of REASv3.1 were generally similar with the results shown in Fig. S18. Emissions of REASv3.1 were close to those obtained by Streets et al. (2003a), Zhang et al. (2009), CEDS, and REASv2.1, but were larger than EDGARv4.3.2, and much smaller than Klimont et al. (2017).

Figure 12 compares trends of relative ratios of SO_2, NO_x, and BC emissions from each country and region among REASv3.1, CEDS, and EDGARv4.3.2. Comparisons of other species are presented in Fig. S19. For SO_2, REASv3.1 and CEDS values were similar until the mid-2000s. Contribution from Japan were relatively large from 1950 until around 1970 and then,

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decreased keenly. This was also found in EDGARv4.3.2, but the rate of decrease was smaller than that of REASv3.1 and CEDS. After the mid-2000s, relative ratios of SO_2 emissions in China were stable in CEDS and EDGARv4.3.2, but those in REASv3.1 decreased significantly. Recently, increasing trends of relative ratios of SO_2 emissions in India are a common feature in REASv3.1, EDGARv4.3.2, and CEDS. For NO_x, REASv3.1 and CEDS generally showed similar temporal variations of the contributing rates, although relative ratios of South Asia other than India were larger in CEDS. Relative ratios of Japan were large around 1970 and then gradually decreased. Instead, contribution rates from China increased almost monotonically until 2010. Similar to the case of SO_2, relative ratios of China decreased recently in REASv3.1, but they were almost stable in CEDS and EDGARv4.3.2. In all the results, contribution rates from India showed gradual increasing trends. Compared to SO_2 and NO_x, temporal variations of relative ratios of BC emissions from each country and regions were small for REASv3.1, CEDS, and EDGARv4.3.2. In REASv3.1, contribution rates of Japan were large before 1970 and then decreased afterwards. On the other hand, in CEDS, contribution rates of Japan after 1970 were larger than those before 1970. After 2000, contribution rates of China in REASv3.1 were almost stable and showed a marginal decrease after 2011. In CEDS and EDGARv4.3.2, contribution rates of China increased during the first half of 2000s and then became almost stable. Similar tendencies were seen in OC. Compared to BC, relative ratios of China started to decrease earlier only in REASv3.1. CO trends were similar to those of BC. But contribution rates of China in REASv3.1 increased gradually until the mid-2000s and then decreased, while those in CEDS and EDGARv4.3.2 were almost stable. For NMVOC, contribution rates of China in REASv3.1 increased largely after the early 2000s. Similar increasing trends were seen in EDGARv4.3.2 but growth rates of REASv3.1 were much larger. On the other hand, both temporal variations and values of contribution rates of China were relatively small in CEDS. For NH_3, contribution rates of China in REASv3.1 increased gradually until the mid-2000s and then became almost stable, whereas contribution rates from China in CEDS and EDGARv4.3.2 show slightly decreasing and increasing trends, respectively. In 2015, contribution rates of NH_3 emissions from China in REASv3.1 were between those of EDGARv4.3.2 and CEDS. Compared to EDGARv4.3.2 and CEDS, contribution rates of NH_3 emissions from Southeast Asian region were relatively small in REASv3.1.

3.4 Uncertainty

In REASv3.1, uncertainties in emissions were estimated for each country and region as well as each year using the same methodology as that of REASv2.1 (Kurokawa et al., 2013). First, uncertainties in all the parameters used to calculate emissions, such as activity data, emission factors, removal efficiencies, and sulfur contents of fuels were estimated in the range of 5-200%. Then, uncertainties in emissions from power plants, industries, road transport, other transport, domestic and other sectors, as well as uncertainties in total emissions were calculated for all the species by combining the estimated uncertainties of used parameters. Similar to REASv2.1, uncertainties of emissions that were not originally developed in REASv3.1 (NH_3 emissions from manure management and fertilizer application, and NMVOC evaporative emissions from Japan and the Republic of Korea) were not evaluated in this study.

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Table 3 summarizes the estimated uncertainties in total emissions of each species for China, India, Japan, Southeast Asia, East Asia other than China and Japan, and South Asia other than India in 2015, 1985, and 1955. Uncertainties in emissions from each sector were provided in Table S2 for SO_2, NO_x, CO, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC, in Table S3 for NMVOC, and in Table S4 for NH_3. For most regions and years, the smallest uncertainties are found for SO_2 emissions; uncertainties for NO_x and CO_2 are smaller than other species. Emissions of SO_2 are mainly determined by the sulfur contents of fuels, with contributions from power plants and industries sector being large. As compared to other sources, the accuracy of data of sulfur contents of fuels are assumed to be high because the information available on sulfur contents is relatively large, even for earlier years in the period of REASv3.1. Another reason for a relatively high accuracy of SO_2 emission data is that uncertainties of emission factors of combustion at high temperature, such as for large power and industry plants are considered to be small. This is also a major reason that uncertainties of NO_x and CO_2 emissions are lesser than others. On the other hand, uncertainties of PM species are large compared to other species for most regions and years. For most countries in Asia, a majority of their emissions was from combustion at relatively low temperatures in small industries and residential sectors. Accuracies of activity data and emission factors for these sources are assumed to be low, especially for biofuel combustion. Therefore, uncertainties of OC emissions mainly from biofuel combustion in Asia are the largest for most regions and years. For CO and NMVOC, in general, uncertainties of emission factors are assumed to be greater than SO_2, NO_x, and CO_2 and smaller than PM species. Therefore, uncertainties of CO and NMVOC emissions are generally middle of all species. Exceptions to these include uncertainties in CO emissions in India and Southeast Asia, which are comparable to PM species as their relative contribution from biofuel combustion is large.

Uncertainties in emissions from Japan are lesser than those of other countries and regions. This is mainly due to the accuracy of activity data. Accessibility to detailed information in Japan is relatively high compared to other countries in REASv3.1. For China and India, accuracies of emissions are generally improved for most species compared to REASv2.1 using information from recently published literatures of emission inventory of these countries. However, the improvement is not significant due to the lack of country specific information. This situation is almost the same for other countries and regions. Although studies of national emission inventories in Asia are being published, as described in Section 1, information on technologies related to emissions and their introduction rates is not as easily available. Therefore, continuous efforts to update emission inventories by collecting information of each country and region are essential. For all countries, uncertainties of emissions in 1955 were much larger than those in 2015. This is because most activity data were not obtained directly from statistics, especially in the early half of the target period of REASv3.1. In this study, activity data, which were not available in statistics were extrapolated or assumed using proxy data as described in Section 2. In order to reduce uncertainties of emissions in long past years, these procedures need to be considered based on detailed information of each country and region during the period.

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4 Summary and remarks

A long historical emission inventory of major air and climate pollutants in Asia during 1950-2015 was developed as Regional Emission inventory in ASia version 3.1 (REASv3.1). Target species were SO_2, NO_x, CO, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, BC, OC, and CO_2 and the domain areas included East, Southeast, and South Asia. Emissions from fuel combustion in power plants, industries, transport, and domestic sectors and those from industrial processes were estimated for all the species. In addition, emissions from agricultural activities and human physiological phenomenon were considered for NH_3 and those from evaporative sources were included in NMVOC. REASv3.1 provides gridded data as well as emissions from each country and sub-region. Spatial resolution is mainly 0.25° × 0.25° and large power plants are treated as point sources. Temporal resolution is monthly. Emissions were estimated based on information of technologies related to emission factors and removal efficiencies, although available data and literatures are limited in the case of Asia. Activity data for recent years were collected from international and national statistics and those of past years, when detailed information was not available, were extrapolated using proxy data for the target period of REASv3.1.

Total emissions in Asia averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are: SO_2: 3.15 Tg, 42.4 Tg (13.5); NO_x: 1.83 Tg, 47.6 Tg (26.0); CO: 62.2 Tg, 319 Tg (5.13); NMVOC: 9.14 Tg, 61.8 Tg (6.77); NH_3: 7.99 Tg, 31.3 Tg (3.92); CO_2: 1.12 Pg, 18.3 Pg (16.3); PM_{10}: 5.76 Tg, 28.4 Tg (4.92); $PM_{2.5}$: 4.52 Tg, 20.3 Tg (4.50); BC 0.751 Tg, 3.38 Tg (4.51); and OC 2.62 Tg, 6.92 Tg (2.64). Clearly, all the air pollutant emissions in Asia increased significantly during these six decades. However, situations were different among countries and regions. In recent years, the relative contribution of air pollutant emissions from China was the largest along with rapid increase in economic growth, but most species have reached their peaks and the growth rates of other species have become at least small or almost zero. For SO_2 and NO_x, introduction of abatement equipment, especially for coal-fired power plants, such as FGD and SCR were considered to be effective in reducing emissions. For PM species, in addition to control equipment in industrial plants, emissions decreased recently due to reduced usage of biofuels. On the other hand, air pollutant emissions from India showed an almost continuous increase. Growth rates were larger for SO_2 and NO_x, but their structures of emissions were different. Large parts of SO_2 emissions were obtained from coal combustion in power plants and industrial sector, and the recent rapid increase of SO_2 emission was mainly from coal-fired power plants. For NO_x, contribution and growth rates from road transport especially diesel vehicles were almost comparable with those of power plants. For PM species, a majority of emissions was from the residential sector in 1950s-1960s; its contribution is still considered to be large. Recent increasing trends were mainly caused by emissions from power and industrial plants and road vehicles. Trends in Japan were much different than those of the whole of Asia. Emissions increased rapidly along with economic growth during the 1950s-1970s, but those of most species were reduced largely from peak values. In addition, peak years were mostly 40 years ago, reflecting the time series of introduction of regulations and laws to mitigate air pollution. Similar features were found in the Republic of Korea and Taiwan. For other countries in Asia, emissions of air pollutants generally showed increasing trends along with economic

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situation and motorization. As described above, trends and spatial distribution of air pollutants in Asia are not simple and are becoming complicated.

Mitigation of air and climate pollutant emissions is an urgent issue in most Asian countries, but the situation is different country-wise. Therefore, continuous efforts to develop and update emission inventories in Asia that are based on country specific information are essential. However, there are inevitable uncertainties in parameters required to develop emission inventories, such as activity data and emission factors. In addition, it is fundamentally impossible to develop a real-time emission inventory because there is a time lag in the publication of basic statistics essential to estimate emissions. Recently, satellite observation data of air pollutants are becoming available at a finer scale for many species, such as NO_x, SO_2 and NH_3. Evaluations and improvements of REASv3.1 based on these data as well as results of modeling studies, such as inverse modeling are important next steps. Also, addition of target species, especially CH_4, which is one of the key species to mitigate both air pollution and global warming is another important task for future studies.

Data availability:

Monthly gridded emission data sets at 0.25° × 0.25° resolution for major sectors from 1950 to 2015 are available from a data download site of REAS. The URL of the site is http://www.nies.go.jp/REAS/. Country and regional emission table data for major sectors during 1950-2015 and those for major fuel types are also provided at the site.

Author contribution:

JK and TO conducted the study design. JK contributed to actual works for development of REASv3.1 such as collecting data and information, settings of parameters, calculating emissions and creating final data sets. JK and TO analyzed and discussed the estimated emissions in REASv3.1. JK prepared the manuscript with contributions from TO.

Competing interest:

The authors declare that they have no conflict of interest.

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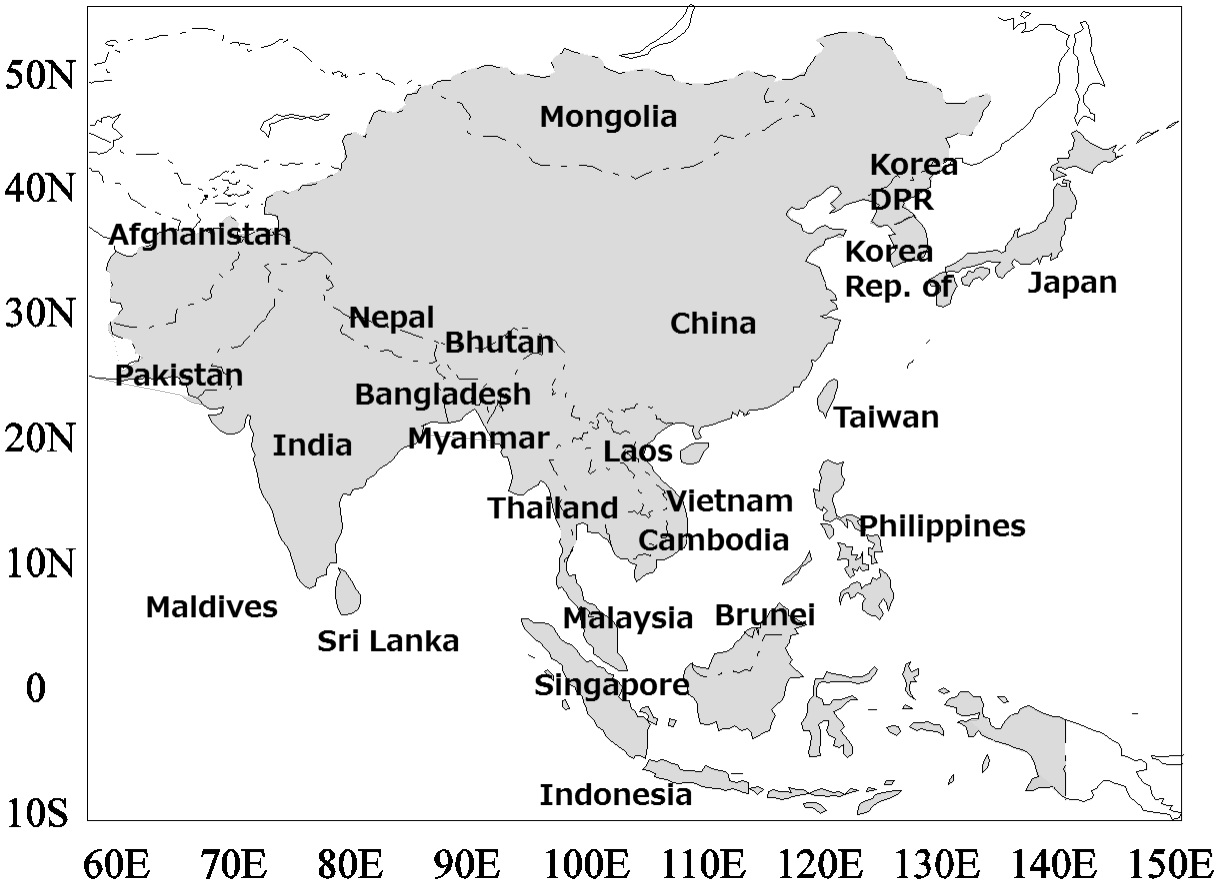

Figure 1: Domain and target countries of REASv3.1.

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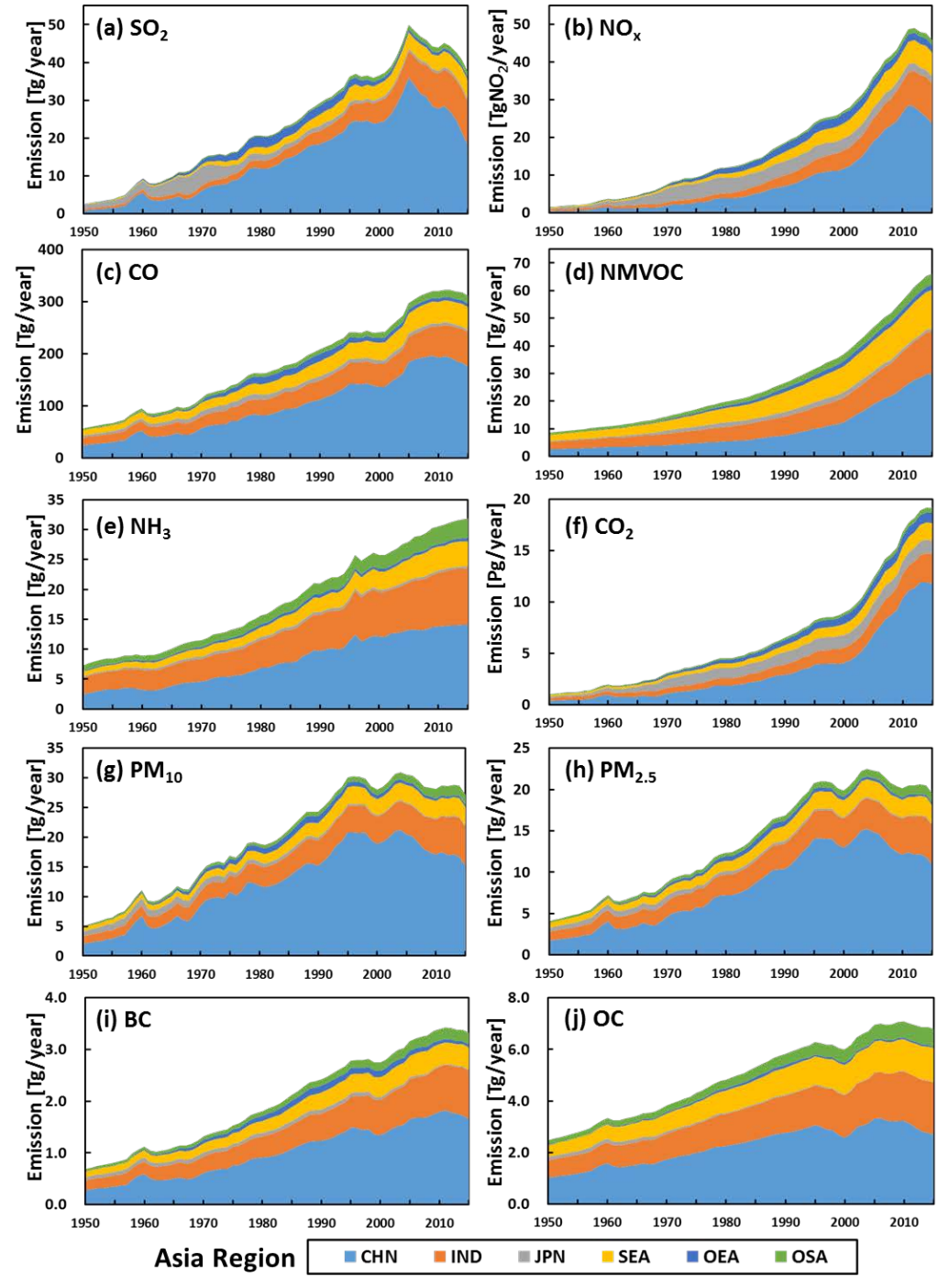

Figure 2: Trends of (a) SO_2, (b) NO_x, (c) CO, (d) NMVOC, (e) NH_3, (f) CO_2, (g) PM_{10}, (h) $PM_{2.5}$, (i) BC, and (j) OC emissions in Asia during 1950-2015 for each region. Regions: CHN = China, IND = India, JPN = Japan, SEA = Southeast Asia, OEA = East Asia other than China and Japan, and OSA = South Asia other than India. See Table S1 for countries included in SEA, OEA, and OSA.

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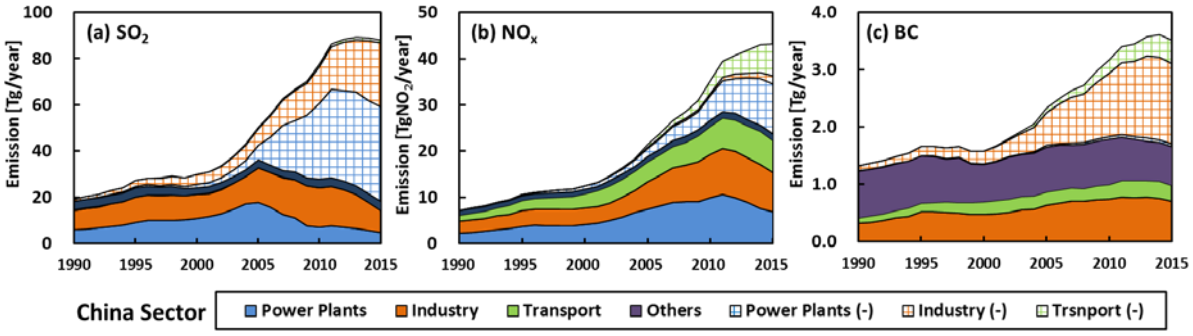

Figure 3: Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in China during 1950-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures.

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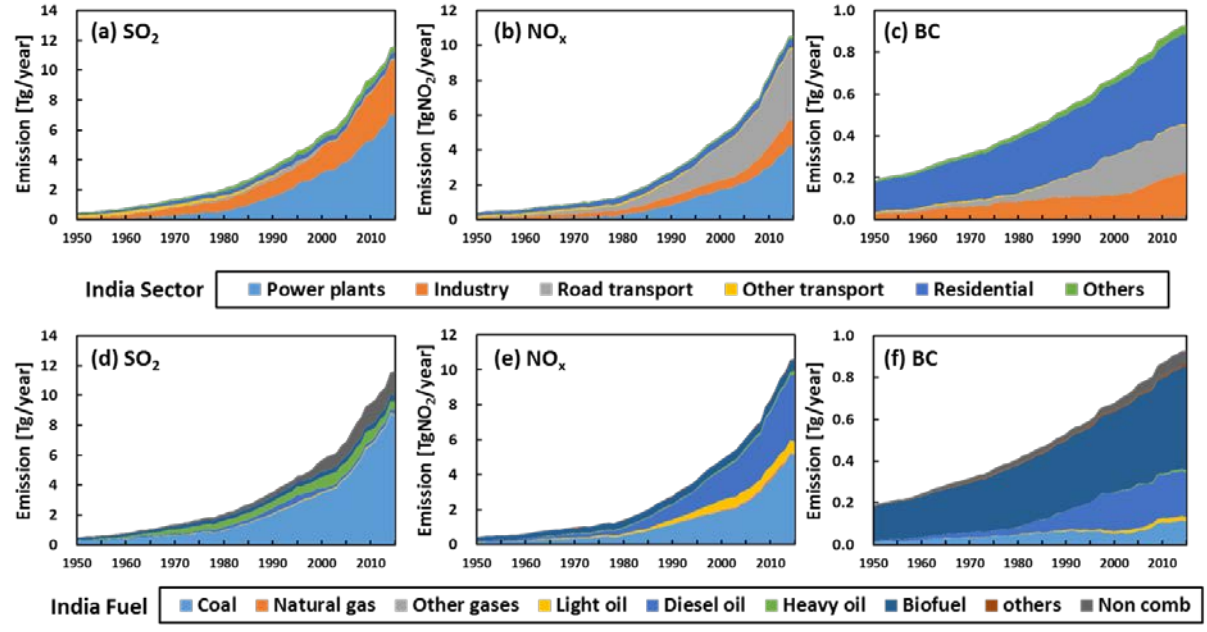

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Figure 4: Emissions of (a, d) SO_2, (b, e) NO_x, and (c, f) BC from each major sector category (upper panels) and fuel type (lower panels) in India from 1950 to 2015. (Non comb = Non combustion sources)

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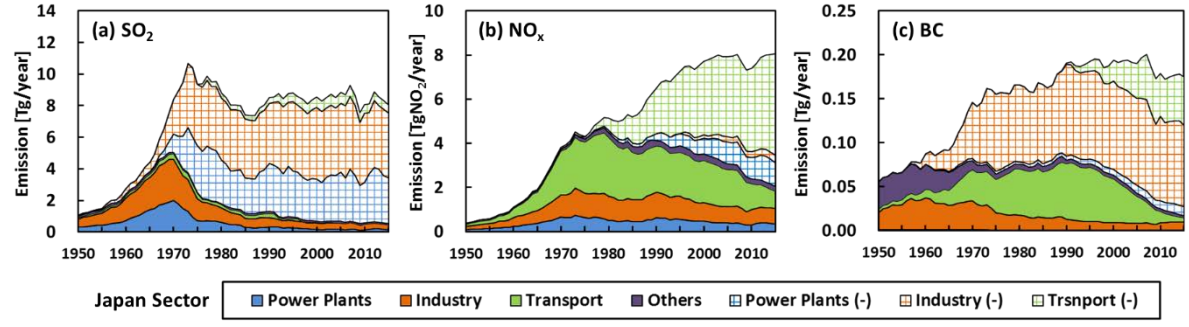

Figure 5: Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in Japan during 1950-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures.

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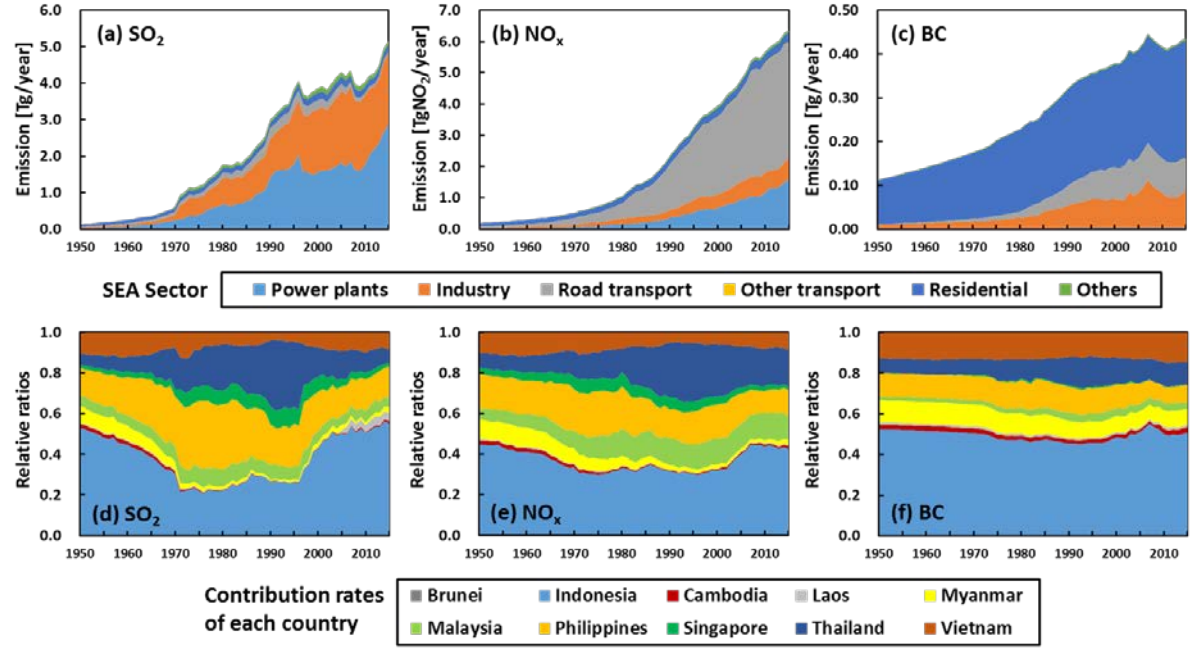

Figure 6: Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in total Southeast Asia (SEA) (upper panels) and (d, e, f) relative ratios of emissions from each country in SEA (lower panels) during 1950-2015.

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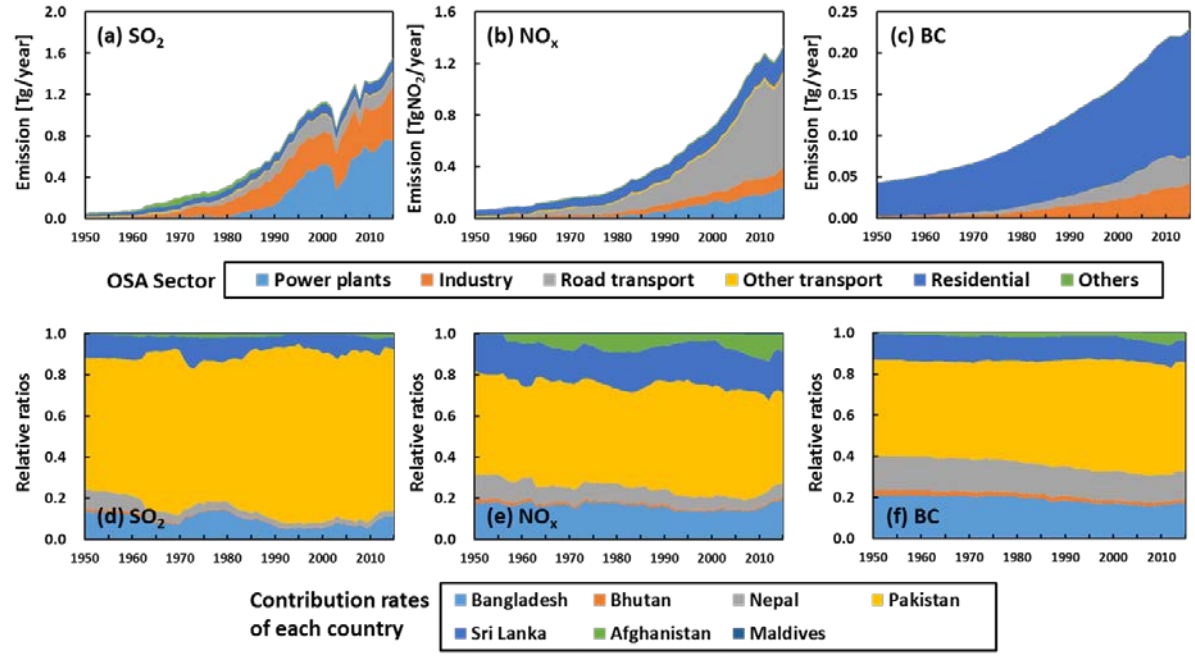

Figure 7: Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in total South Asia other than India (OSA) (upper panels) and (d, e, f) relative ratios of emissions from each country in OSA (lower panels) from 1950 to 2015.

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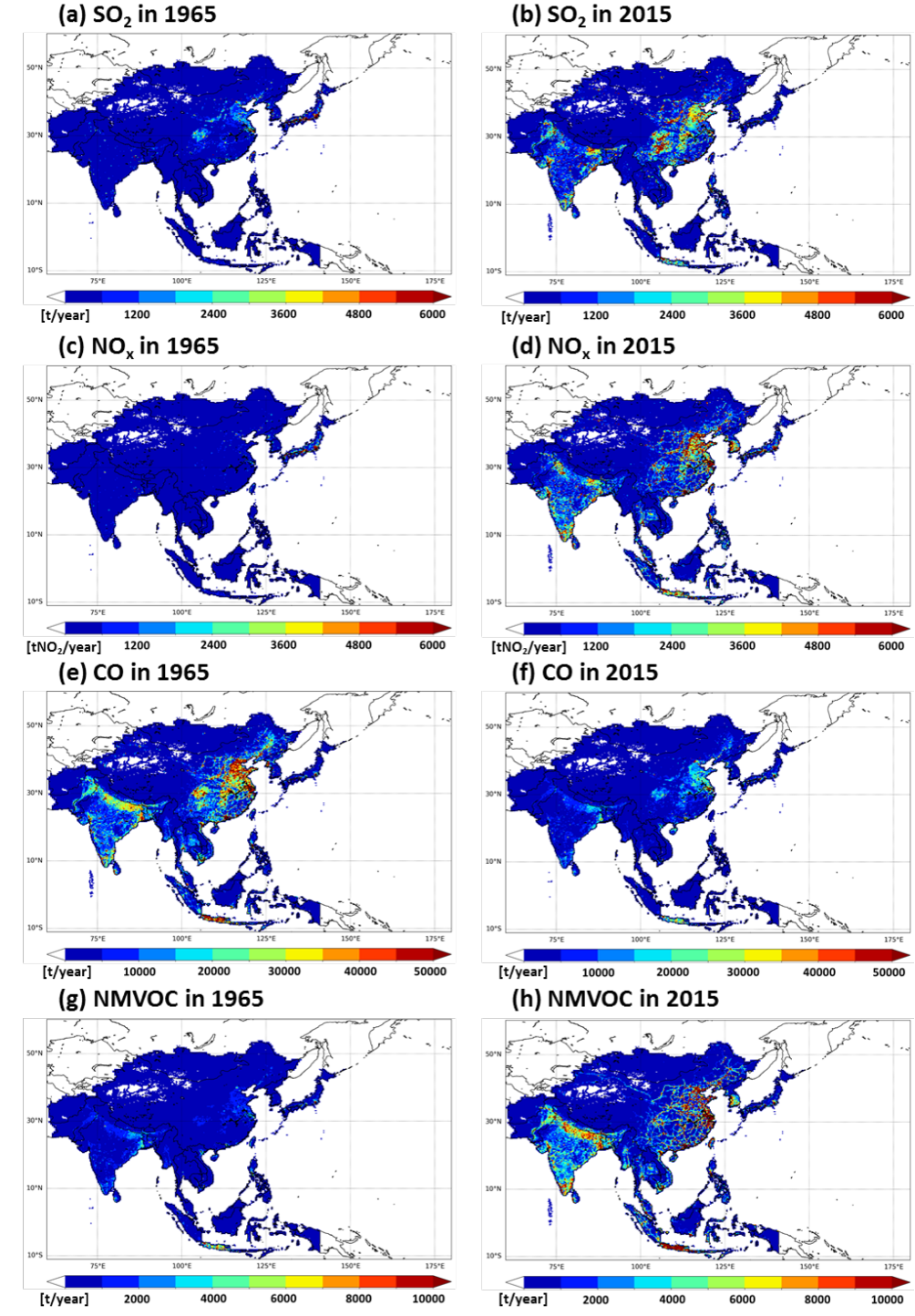

Figure 8: Grid maps of annual emissions of (a, b) SO_2, (c, d) NO_x, (e, f) CO, (g, h) NMVOC, (i, j) NH_3, (k, l) $PM_{2.5}$, (m, n) BC, and (o, p) OC in 1965 (left panels) and 2015 (right panels).

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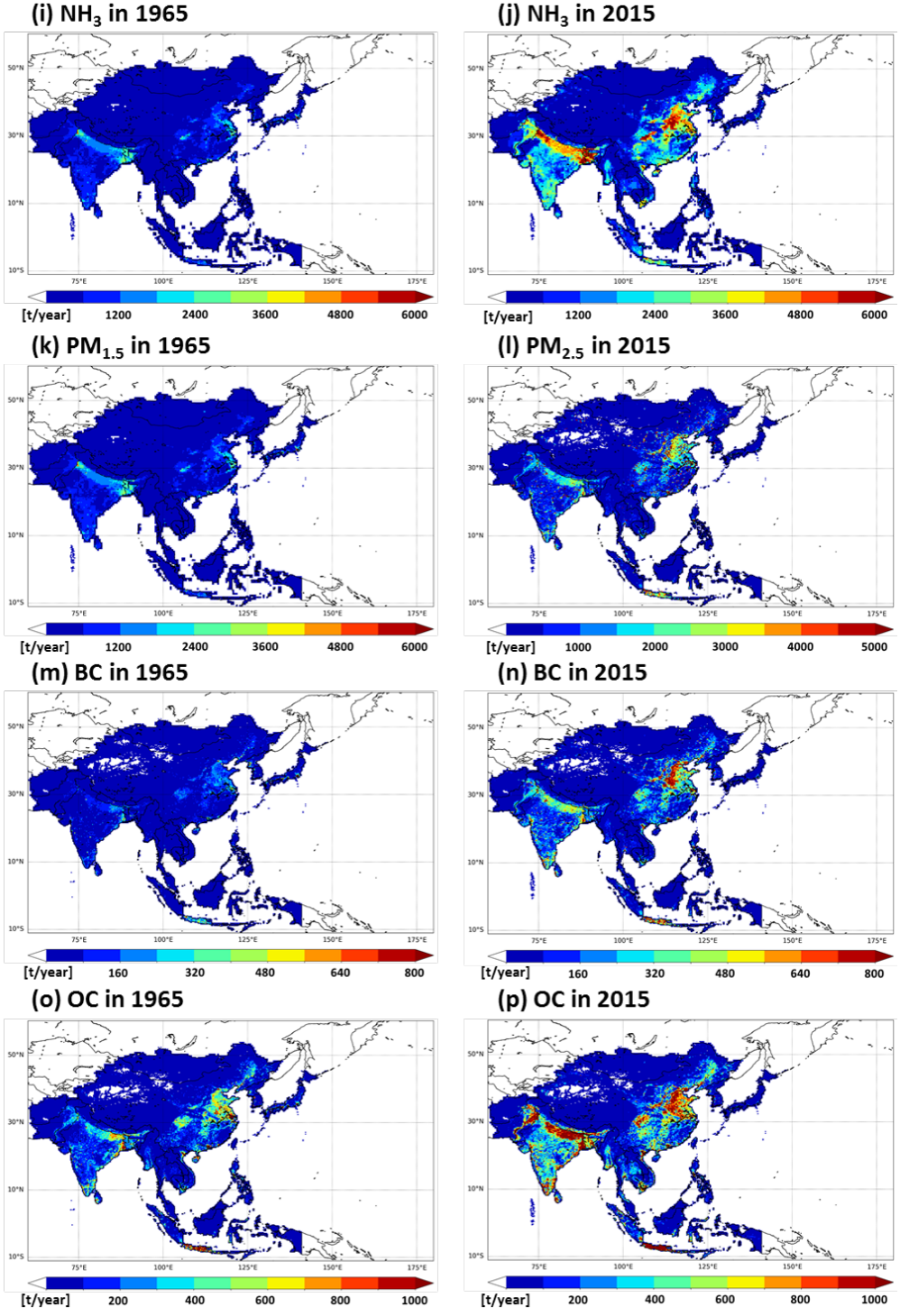

1160 **Figure 8: Continued.**

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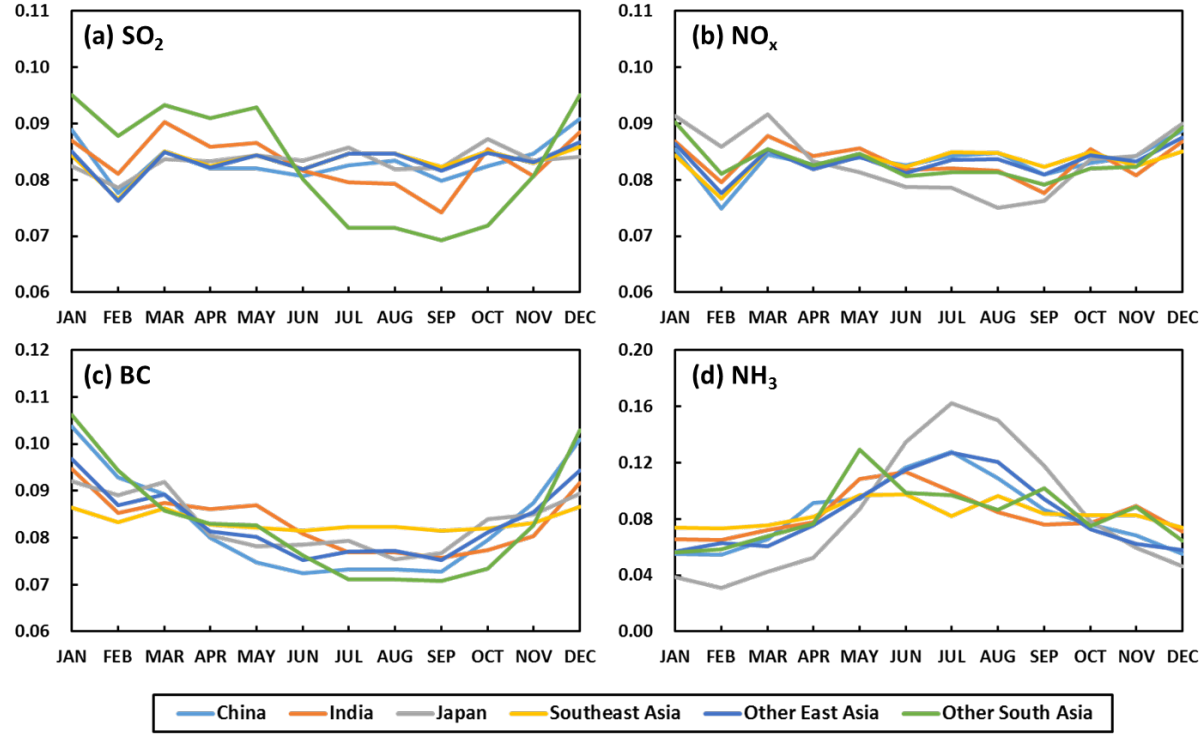

Figure 9: Monthly variations of (a) SO_2, (b) NO_x, (c) BC, and (d) NH_3 emissions for each region of Asia in 2015.

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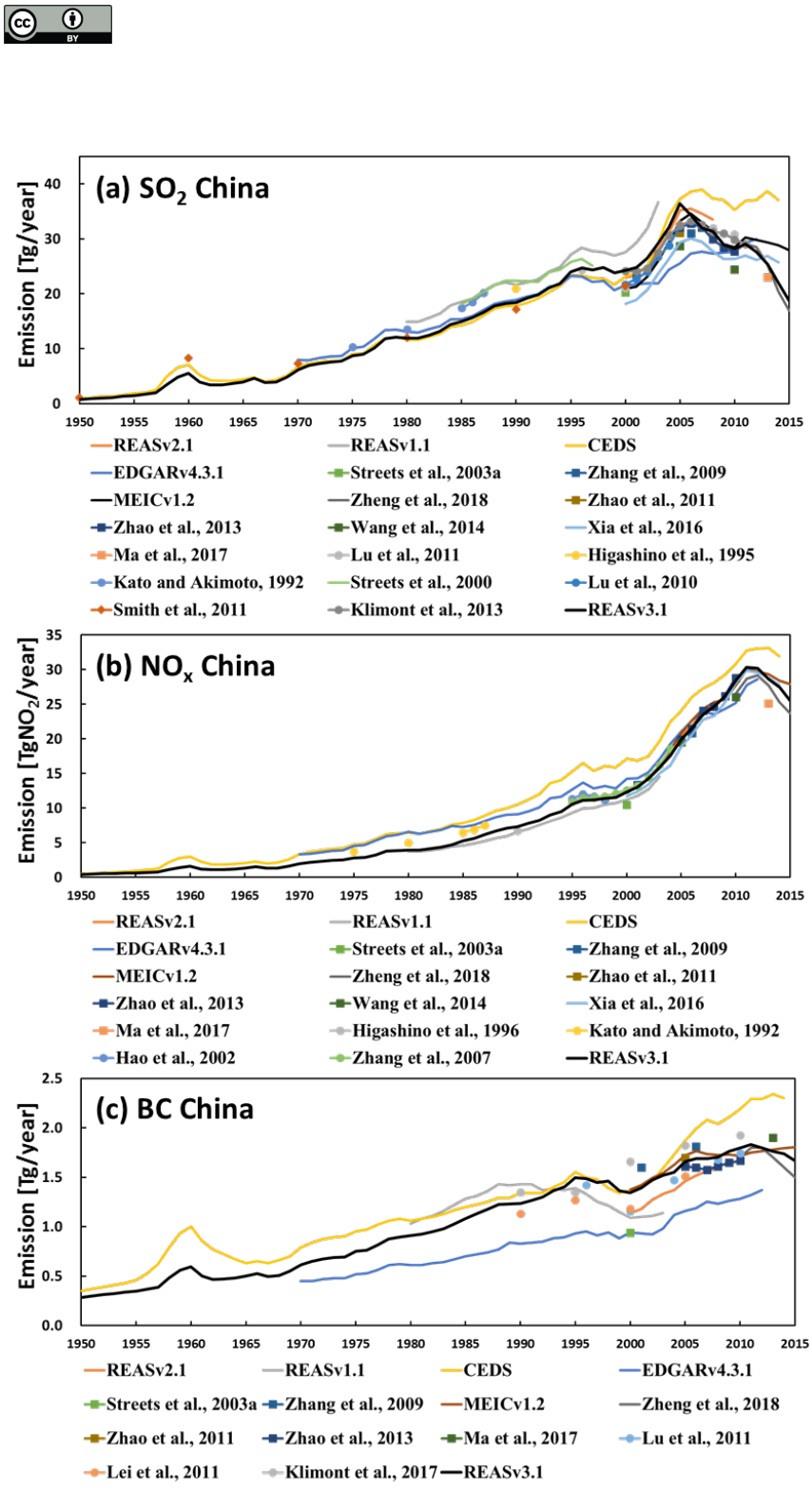

1165 **Figure 10: Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in China between REASv3.1 and other studies.**

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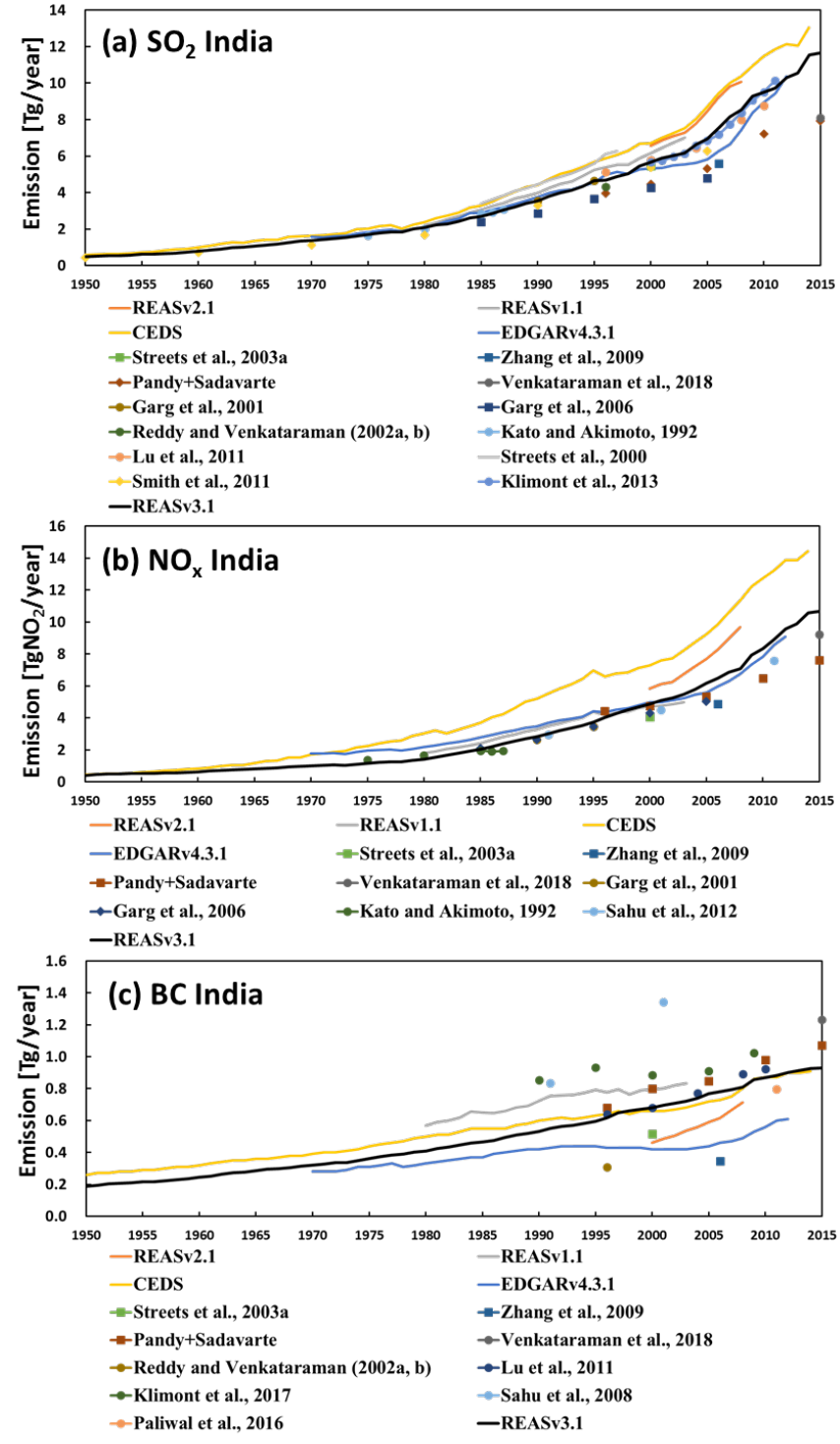

Figure 11: Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in India between REASv3.1 and other studies. Note that values of "Pandy+Sadavarte" are calculated from Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014).a あ

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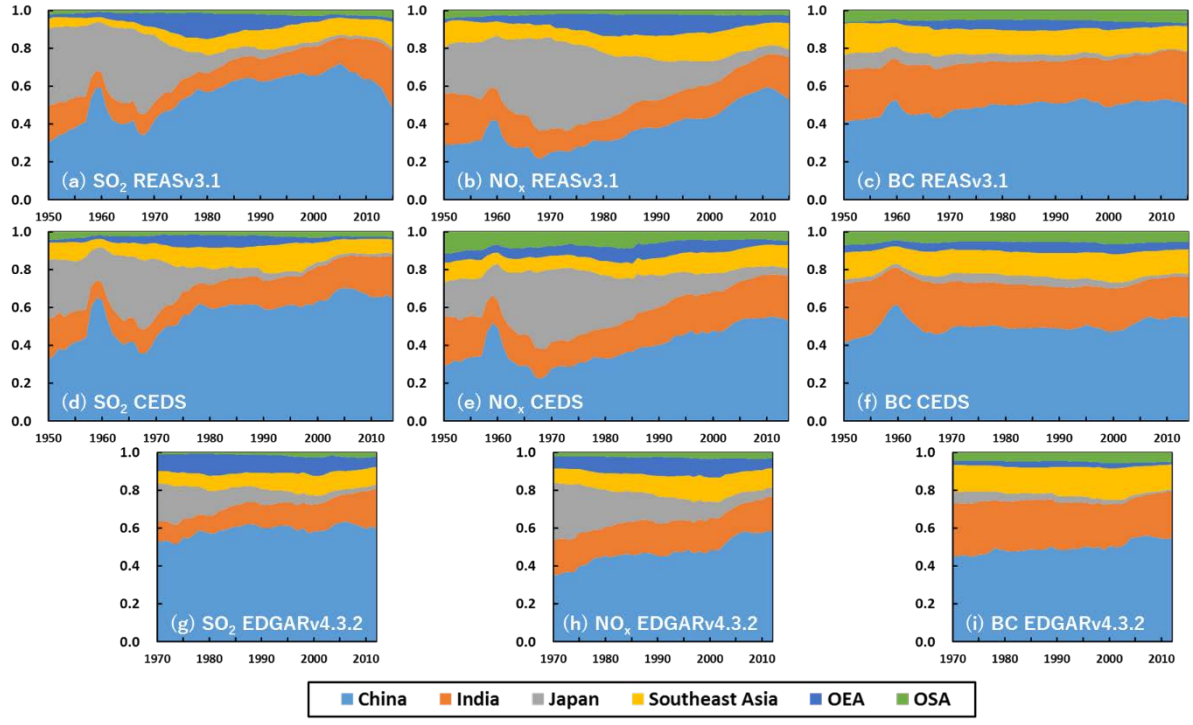

Figure 12: Comparison of trends of relative ratios of emissions from China, India, Japan, Southeast Asia, East Asia other than China and Japan (OEA), and South Asia other than India (OSA) for (a, d, g) SO_2, (b, e, h) NO_x, and (c, f, i) BC among REASv3.1 (upper panels), CEDS (middle panels), and EDGARv4.3.2 (lower panels). Note that periods of CEDS and EDGARv4.3.2 shown here are during 1950-2014 and 1970-2012, respectively.

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Table 1: General information on REAS 3.1.

Item	Description
Species	SO_2, NO_x, CO, NMVOC, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC
Years	1950–2015
Areas	East, Southeast, and South Asia
Emission sources	Fuel combustion in power plans, industry, transport, and domestic sectors; Industrial processes; Agricultural activities (fertilizer application and livestock); and Others (fugitive emissions, solvent use, human, etc.)
Spatial resolution	0.25 degree by 0.25 degree
Temporal resolution	Monthly
Data distribution	http://www.nies.go.jp/REAS/

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1180 **Table 2: Summary of national emissions in 2015 for each species and total annual emissions in Asia in 1950, 1960, 1970, 1980, 1990, 2000, and 2010-2015 (Gg yr^{-1}).**

Country	SO_2	NO_x[a]	CO	NMVOC	NH_3	CO_2[b]	PM_{10}	$PM_{2.5}$	BC	OC
China	18144	23647	175656	29923	14053	11768	15166	10993	1660	2687
Japan	551	1825	3976	900	354	1277	135	106	16	16
Korea, Rep of	361	1203	1713	985	171	663	142	125	27	35
Korea, DPR	116	205	2809	143	92	29	99	54	15	18
Mongolia	107	126	1059	51	139	18	43	24	2.7	3.2
Taiwan	135	425	660	704	90	278	40	35	4.4	5.9
Brunei	4.0	16	29	45	3.8	6.1	4.8	1.5	0.2	0.0
Cambodia	57	78	1500	290	77	22	117	82	12	44
Indonesia	2856	2741	21006	6814	1589	643	1512	1173	220	690
Laos	201	43	387	94	67	11	43	28	4.6	14
Malaysia	238	715	1480	1012	162	226	149	83	13	15
Myanmar	153	158	2933	1093	620	58	187	171	35	128
Philippines	806	874	3544	1103	387	133	252	174	37	76
Singapore	92	104	74	299	8.4	46	16	11	1.0	1.1
Thailand	332	1140	7150	1598	540	316	409	302	47	158
Vietnam	435	568	6620	1641	746	243	532	349	64	185
Bangladesh	170	263	2465	765	879	108	402	224	40	112
Bhutan	3.3	11	283	64	9.2	4.7	28	21	4.0	14
India	11633	10632	65963	15481	9462	2919	6526	4709	929	2042
Nepal	40	89	2295	573	318	39	194	158	32	111
Pakistan	1230	598	8440	2150	1760	272	1032	670	121	381
Sri Lanka	92	263	1355	325	102	37	132	104	24	59
Afghanistan	24	114	360	71	251	9.4	24	20	10	8.5
Maldives	3.1	6.1	6.9	2.7	0.4	0.8	0.4	0.3	0.2	0.1
Asia[c] 1950	2485	1532	57246	8597	7280	1002	4966	4074	688	2465
Asia[c] 1960	9324	3802	94786	10892	8932	1976	11045	7262	1125	3330
Asia[c] 1970	14384	7567	113836	14398	11538	3064	14088	8779	1317	3824
Asia[c] 1980	20660	12112	161936	19861	15582	4490	18925	12366	1806	4823
Asia[c] 1990	29186	18625	208143	26603	20975	6514	24249	16846	2420	5831
Asia[c] 2000	36592	27126	241096	36916	25711	8940	28057	19904	2751	6004
Asia[c] 2010	43884	46174	320578	56423	30550	16764	28110	20166	3392	7095
Asia[c] 2011	45111	48741	323203	58893	30809	17742	28750	20517	3421	6998
Asia[c] 2012	44407	49059	322774	61230	31214	18193	28591	20502	3408	6939
Asia[c] 2013	42669	47935	319557	63100	31491	18894	28818	20614	3383	6858
Asia[c] 2014	40797	47653	318009	65179	31700	19154	28683	20513	3383	6857
Asia[c] 2015	37785	45845	311766	66126	31882	19125	27185	19617	3319	6801

[a]Gg-NO_2 yr^{-1}.
[b]Tg yr^{-1}.
[c]Asia in this table include all target countries and sub-regions in REASv3.1.

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Table 3: Uncertainties [%] of emissions in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in 2015, 1985, and 1955.

	SO_2	NO_x	CO	NMVOC	NH_3	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
2015										
China	±30	±35	±81	±75	±80	±28	±94	±102	±125	±220
India	±34	±39	±137	±122	±99	±40	±128	±157	±156	±240
Japan	±29	±25	±35	±31	±92	±23	±61	±61	±54	±74
SEA	±39	±44	±121	±102	±103	±39	±141	±173	±184	±242
OEA	±37	±68	±84	±78	±93	±33	±72	±84	±88	±204
OSA	±31	±34	±91	±79	±87	±28	±105	±119	±140	±233
1985										
China	±35	±61	±146	±184	±123	±59	±110	±141	±206	±295
India	±33	±61	±202	±231	±116	±89	±168	±208	±214	±275
Japan	±25	±30	±44	±35	±80	±23	±65	±60	±55	±58
SEA	±37	±59	±176	±181	±122	±87	±184	±223	±240	±277
OEA	±43	±73	±79	±90	±96	±41	±74	±72	±91	±110
OSA	±34	±47	±138	±164	±119	±51	±117	±150	±196	±285
1955										
China	±78	±161	±282	±300	±150	±180	±272	±341	±379	±420
India	±72	±118	±259	±306	±138	±156	±262	±296	±296	±326
Japan	±63	±60	±141	±203	±325	±58	±85	±105	±153	±251
SEA	±88	±141	±234	±281	±147	±169	±301	±312	±331	±326
OEA	±80	±94	±126	±186	±123	±71	±111	±145	±140	±284
OSA	±66	±110	±258	±287	±149	±147	±233	±297	±338	±378