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Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2

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Abstract

We have updated the Regional Emission inventory in ASia (REAS) as version 2.1. REAS 2.1 includes most major air pollutants and greenhouse gases from each year during 2000 and 2008 and following areas of Asia: East, Southeast, South, and Central Asia and the Asian part of Russia. Emissions are estimated for each country and region using updated activity data and parameters. Monthly gridded data with a 0.25° × 0.25° resolution are also provided. Asian emissions for each species in 2008 are as follows (with their growth rate from 2000 to 2008): 56.9 Tg (+34 %) for SO_2, 53.9 Tg (+54 %) for NO_x, 359.5 Tg (+34 %) for CO, 68.5 Tg (+46 %) for non-methane volatile organic compounds, 32.8 Tg (+17 %) for NH_3, 36.4 Tg (+45 %) for PM_{10}, 24.7 Tg (+42 %) for $PM_{2.5}$, 3.03 Tg (+35 %) for black carbon, 7.72 Tg (+21 %) for organic carbon, 182.2 Tg (+32 %) for CH_4, 5.80 Tg (+18 %) for N_2O, and 16.7 Pg (+59 %) for CO_2. By country, China and India were respectively the largest and second largest contributors to Asian emissions. Both countries also had higher growth rates in emissions than others because of their continuous increases in energy consumption, industrial activities, and infrastructure development. In China, emission mitigation measures have been implemented gradually. Emissions of SO_2 in China increased from 2000 to 2006 and then began to decrease as flue-gas desulfurization was installed to large power plants. On the other hand, emissions of air pollutants in total East Asia except for China decreased from 2000 to 2008 owing to lower economic growth rates and more effective emission regulations in Japan, South Korea, and Taiwan. Emissions from other regions generally increased from 2000 to 2008, although their relative shares of total Asian emissions are smaller than those of China and India. Tables of annual emissions by country and region broken down by sub-sector and fuel type, and monthly gridded emission data with a resolution of 0.25 × 0.25° for the major sectors are available from the following url: http://www.nies.go.jp/REAS/.

1 Introduction

Estimating anthropogenic emissions of air pollutants and greenhouse gases in Asia, where dramatic spatial and temporal variations of emissions have occurred in the last three decades, is a very important task for understanding and controlling the regional and global atmospheric environment. The earliest Asian emission inventory developed by Kato and Akimoto (1992), estimated SO_2 and NO_x emissions of East Asian, Southeast Asian, and South Asian countries in 1975, 1980, and 1985–1987. Akimoto and Narita (1994) provided gridded data sets with a resolution of 1 × 1°. Streets et al. (2003a, b) developed detailed emission inventories in Asia for the year 2000 for modeling study of TRACE-P (Transport and Chemical Evolution over the Pacific) field campaigns (Jacob et al., 2003). The TRACE-P project studied Asian outflows of gaseous and aerosol species and their chemical evolution over the western Pacific during the spring of 2001. For TRACE-P's successor mission Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) performed in 2006 (Singh et al., 2009), Zhang et al. (2009a) developed a new emission inventory in Asia for the year 2006. For this effort, Zhang et al. (2009a) improved the methodology for estimating emissions from China by using a detailed technology-based approach that took into consideration recent rapid technology renewal in China. The TRACE-P and INTEX-B data sets have been used not only for their original purpose (e.g. Adhikary et al., 2010) but also for many other atmospheric chemistry modeling studies in Asia (e.g. Liu et al., 2010).

For analyses of long-term trends of the Asian atmospheric environment, Ohara et al. (2007) developed the first inventory of historical and future projected emissions in Asia on the basis of a consistent methodology, the Regional Emission inventory in ASia version 1.1 (REAS 1.1). The target years were from 1980–2003 for historical emissions and 2010–2020 for future projections. REAS 1.1 includes emissions of following species: SO_2, NO_x, CO, non-methane volatile organic compounds (NMVOC), black carbon (BC), organic carbon (OC), CO_2, NH_3, CH_4, and N_2O. The inventory domain includes East, Southeast, and South Asia. Both country and sub-regional emissions and

gridded data sets with $0.5 \times 0.5^{\circ}$ resolution were provided in annual amounts. REAS 1.1 data have been used for many atmospheric chemistry modeling studies in Asia (e.g. Nagashima et al., 2010). However, energy consumption in the Asian region has grown continuously since 2003, the last year of REAS 1.1 (IEA, 2011). In addition, REAS 1.1 generally did not incorporate known temporal variations in emission factors and removal efficiencies. Zhang et al. (2009a) noted that emission factors in China have changed recently because of implementation of emission control measures especially for coal-fired power plants and new vehicles. Therefore, activity data and parameters of REAS 1.1 have become outdated since 2000. Furthermore, improvements in computational power and atmospheric chemistry models allow modeling studies to be conducted on, expanded target areas and species, at higher spatial and temporal resolutions.

In response to these developments, we have updated the REAS inventory and issued it as version 2.1. This paper provides a description of methodology, results and discussions of REAS 2.1. In Sect. 2, we describe the revisions to REAS 1.1, and give an overview of the basic methodology of data processing, and updated activity data and emission factors. We also describe the new emissions data from Japan, South Korea, and Taiwan that are incorporated in REAS 2.1. Section 3.1 presents the basic results of Asian and national emissions of each species, and their spatial and temporal variations are presented in Sects. 3.2 and 3.3, respectively. We compare the results of REAS 2.1 with REAS 1.1 in Sect. 3.4 and with other inventories in Sect. 3.5. Section 3.6 briefly discusses the uncertainties in REAS 2.1. Information about data distribution is given in Sect. 3.7, and Sect. 4 presents a summary including future plans for REAS.

2 Methodology

2.1 Revisions from REAS version 1

We developed REAS 2.1 by updating REAS 1.1 (Ohara et al., 2007). Major revisions were as follows (Table 1):

- Two categories of particulate matter (PM_{10} and $PM_{2.5}$) were added to the target species.
- Target years were changed to 2000–2008, and two new regions, Central Asia and the Asian part of Russia (Asian Russia) were added to the target area.
- Basic activity data, parameters, and methodologies were revised in light of recent studies of the Asian emission inventory.
- Spatial resolution of gridded data was improved to $0.25 \times 0.25^{\circ}$, and temporal resolution was increased to monthly.

REAS 2.1 includes most major air pollutants and greenhouse gases. Among primary aerosols, REAS 1.1 considered BC and OC, which have climate impacts, but did not include particulate matter (PM). However, epidemiological studies have shown that high PM concentrations have negative health impacts, including asthma, heart attacks, and premature mortality. In addition, assessment of long-range transport of PM has become important in the Asian region. For these reasons, REAS 2.1 includes emissions of PM_{10} and $PM_{2.5}$. Whereas REAS 1.1 extrapolated the NMVOC emissions developed by Klimont et al. (2002a) and Streets et al. (2003a), in REAS 2.1 emissions were calculated from activity data and emission factors for each target year.

REAS 2.1 focuses on emissions after the year 2000. One reason is that Asian emissions increased rapidly after around 2000 (Ohara et al., 2007). Another reason is that penetration of new technologies, abatement equipment, and regulated vehicles have led to gradual changes in Asian emission factors particularly in China, after 2000 (Zhang et al., 2009a; Lu et al., 2010).

Figure 1 shows the inventory domain of REAS 2.1. In addition to the area covered in REAS 1.1, the new inventory includes Asian Russia (Urals, Western and Eastern Siberia, and Far East) lying east of the Ural Mountains (east of 60° E) and Central Asia (Kazakhstan, Kyrgyzstan, Tajikistan, Turkmenistan, and Uzbekistan). We also divided China into 33 sub-regions and India into 17 sub-regions to reduce uncertainty in the spatial distribution of emissions. Definitions of the sub-regions are slightly different from those in REAS 1.1, a reflection of changes of districts in each country during the decade after the release of REAS 1.1 (see Table S1 in the supplement for the list of all target countries and sub-regions). With respect to Japan, South Korea, and Taiwan, we decided to use inventories from recent studies based on new detailed basic data and parameters (see Sect. 2.5).

Source categories considered in REAS 2.1 are basically the same as in REAS 1.1. For most species, major emission sources are combustion of fossil fuel and biofuel in power plants, industry, road transport, other transport and domestic sectors. Sources other than combustion in the industry sector include production of cement, non-ferrous metals, chemical products, etc. NMVOCs have specific emission sources including solvent use, paint use, and evaporation from road vehicles. Fugitive emissions from coal mining and oil and gas production are considered for CH_4, and those from extraction, handling and transport of coal, petroleum and gas are estimated for CH_4 and NMVOC.

Basic methodologies for estimating emissions are almost the same as those of REAS 1.1. However, we collected more country-specific and region-specific information from recent studies of emission inventories for Asian countries (see Sects. 2.3 and 2.4). With regard to road transport emissions, we calculated traffic volumes on the basis of the number of vehicles and annual distance traveled for this study rather than energy consumption and fuel economy as in REAS 1.1. In addition, cold start emissions which were ignored in REAS 1.1 were calculated by a simple methodology in REAS 2.1 (see Sect. 2.2.2). REAS 1.1 estimated emissions related to agricultural activities such as fertilizer application and manure management of livestock for NH_3, CH_4, N_2O, and NO_x up to the year 2000. For REAS 2.1, we used trends of fertilizer use, number of

livestock, and corresponding emission factors to extrapolate the agricultural emission data of REAS 1.1 beyond 2000 (see Sect. 2.2.3). We also considered following emission sources of NH_3 for REAS 2.1: latrines and human perspiration and respiration. Emissions from natural sources such as vegetation and volcanoes and open biomass burning are not considered in either REAS inventory.

The major role of the REAS inventory is to provide emission input data for atmospheric chemistry models. In addition to improving the spatial resolution of gridded data from 0.5 × 0.5° to 0.25 × 0.25°, information about power plants as point sources was totally updated (see Sect. 2.3). As for temporal resolution, REAS 1.1 used annual totals for everything except for soil NO_x emissions, which were monthly totals. In REAS 2.1, we include monthly variations whenever monthly activity statistics or surrogate data were available. Data on weekly and diurnal variations will be considered in future version of REAS.

2.2 Basic methodology

2.2.1 Stationary combustion and industrial processes

Figure 2a shows the basic procedure used in REAS 2.1 to estimate emissions from stationary combustions and industrial processes. Emissions of SO_2 from fuel combustions were calculated from the following equation:

$$E = \sum_{i,j} \left\{ A_{i,j} \times S_{i,j} \times \left(1 - \mathrm{SR}_{i,j}\right) \times \left(1 - R_{i,j}\right) \right\} \quad (1)$$

where E is emissions from each country and sub-region, i and j are, respectively, fuel and sector types, A is fuel consumption, S is sulfur content of fuel, SR is sulfur retention in ash, and R is removal efficiency. Emissions of other combustion species and all species from industrial processes were estimated from the following equation:

$$E = \sum_{i,j} \left\{ A_{i,j} \times \mathrm{EF}_{i,j} \times \left(1 - R_{i,j}\right) \right\} \quad (2)$$

where E is emissions, i is fuel type or sub-category of industry, j is sector type, A is fuel consumption or amount of industrial product, EF is the unabated emission factor, and R is removal efficiency. For fuel combustion sources, the matrix of fuel consumption was prepared based on the energy statistics for each country and sub-region. For power plants, the fuel consumption matrix was created individually if position and fuel consumption data were available, and the power plants were treated as point sources. For industrial process emissions, the amounts of industrial production were collected for each country and sub-region. Large industrial plants have not been surveyed separately, but we plan to treat them as point sources in the next version of REAS. With respect to parameters, region-specific emission factors, sulfur contents of fuel, sulfur retention in ash, and removal efficiencies were prepared for each activity category from a literature survey of Asian emission inventories. See Sects. 2.3 and 2.4 for details about activity data and emission factors, respectively.

2.2.2 Road transport

Figure 2a shows the flow of diagram for estimating emissions from road transport. REAS 2.1 considered not only hot emissions (when the engine is at normal operating temperature), but also separate emission during cold starts. On-road vehicles were classified as passenger cars, buses, light and heavy trucks, and motorcycles, each of which includes gasoline and diesel vehicles, rural vehicles, and LPG vehicles. In REAS 1.1, emissions were calculated from fuel consumption by vehicle type which was distributed by using fuel economies and corresponding emission factors (Ohara et al., 2007). Because fuel economy data were very limited, the same values were used for many countries. However, no new information of this kind was obtained for use in improving REAS 2.1. Although data of annual distance traveled for Asian countries are also limited, Borken et al. (2008) provided new data for China for 2000. For these reasons, REAS 2.1 calculated hot emissions based on vehicle numbers, annual distance

traveled, and emission factors for each vehicle type with the following equation:

$$E_{\mathrm{HOT}} = \sum_i \{\mathrm{NV}_i \times \mathrm{ADT}_i \times \mathrm{EF}_{\mathrm{HOT}i}\} \quad (3)$$

where E_{HOT} is hot emissions, i is vehicle types, NV is number of registered vehicles, ADT is annual distance traveled, and $\mathrm{EF}_{\mathrm{HOT}}$ is emission factor. Exceptions are SO_2, CO_2, and LPG vehicle emissions. SO_2 emissions are estimated based on sulfur contents in gasoline and diesel consumed in road transport sector for each country and sub-region assuming sulfur retention in ash is zero. CO_2 and LPG vehicle emissions are also calculated by amounts of fuel consumption due to the unit of emission factors selected in this study.

REAS 2.1 includes the cold start emissions for NO_x, CO, PM_{10}, $PM_{2.5}$, BC, OC, and NMVOC estimated by the following equation:

$$E_{\mathrm{COLD}} = \sum_i \{\mathrm{NV}_i \times \mathrm{ADT}_i \times \mathrm{EF}_{\mathrm{HOT}i} \times \beta_i \times F_i\} \quad (4)$$

where E_{COLD} is cold emissions, 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 $\mathrm{EF}_{\mathrm{HOT}}$ for cold start emissions. The parameter β and F are functions of average monthly temperature. Equations for β and F and related parameters were taken from the EMEP/EEA emission inventory guidebook 2009 (EEA, 2009). Monthly average surface temperatures for each country and sub-region were calculated from the Japanese 25 yr ReAnalysis (JRA-25) gridded meteorological dataset ($0.125 \times 0.125^\circ$ resolution) developed by the Japan Meteorological Agency (JMA) and the Central Research Institute of Electricity Power Industry (CRIEPI) (Onogai et al., 2007).

2.2.3 Agricultural activities

Agricultural activities related to emissions of air pollutants and greenhouse gases include fertilizer application (NH_3, N_2O, and NO_x), manure management (NH_3, N_2O,

CH_4, and NO_x), enteric fermentation (CH_4) of livestock, and rice cultivation (CH_4). REAS 1.1 developed emission data sets for all these sources over East, Southeast, and South Asia (Yamaji et al., 2003, 2004; Yan et al., 2003a,b,c, 2005). In REAS 2.1, emissions from agricultural activities during 2001 and 2008 were extrapolated from the gridded emission data of REAS 1.1 for 2000 (Fig. 2b). First, activity data such as numbers of livestock and amounts of fertilizer applied were collected from international, national, and regional statistics. Second, the ratio of emissions in the target year to those in 2000 was calculated for each country and sub-region using the activity data and corresponding emission factors from EMEP/EEA emission inventory guidebook 2009 (EEA, 2009). Third, REAS 1.1 gridded emissions for each country and sub-region in 2000 were multiplied by their respective ratios to produce the data for the target years over the REAS 1.1 domain. For the new sub-regions in REAS 2.1 (Asian Russia and Central Asia), we used the Emission Database for Global Atmospheric Research (EDGAR) 4.2 (EC-JRC/PBL, 2011) from 2000 to 2008. Finally, all these sources were used to prepare the agricultural emission datasets of REAS 2.1 between 2000 and 2008.

2.2.4 Other sources

Applications of solvents and paint are major sources of NMVOC emissions, especially in relatively developed countries. Activity data include paint use for architectural and domestic purposes, ink used for publication, and production of many types of solvents. Emissions for these sources were calculated by multiplying corresponding emission factors and activity data.

Some sources of NH_3 emissions are directly related to human life. Activity data for human perspiration and respiration were derived from population numbers. For emissions from latrines (storage tanks of human excreta), ratios of population in areas with and without sewage service were required.

Fugitive emissions from production, processing, and distribution of fossil fuels are the major sources of CH_4. In REAS 2.1, fugitive CH_4 emissions were estimated using

the Tier 1 methodology of the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006). Activity data of fugitive emissions were collected from international, national, and regional energy statistics; these were also used to estimate NMVOC emissions from extraction and processing of fossil fuels. Emissions of CH_4 from solid and water waste were also calculated with Tier 1 of the 2006 IPCC Guidelines.

For emissions from aviation (both domestic and international at altitudes less than 1 km) and international ship navigation, we used gridded data from EDGAR 4.2 for SO_2, NO_x, CO, NMVOC, PM_{10}, BC, OC, CH_4, N_2O, and CO_2 between 2000 and 2008. Emissions of $PM_{2.5}$ were estimated from PM_{10} emissions using emission factors of $PM_{2.5}$ and PM_{10} for ship emissions.

2.2.5 Seasonal variation and grid allocation

To estimate monthly emissions, activity data, emission factors, and removal efficiencies are required on a monthly basis. Because such monthly data are very limited, we prepared monthly proxy indexes when appropriate information was available. Otherwise, emissions were treated as constant fluxes. Interannual variability of monthly variation was also considered, when possible.

Monthly generated power was used as a surrogate for combustion emissions from power plants. Monthly production of industrial products and fossil fuels were used not only for activity data but also for proxy indexes of monthly fuel consumption in each industrial sub-category. For the residential sector, monthly variations in fuel consumption for heating were estimated in each grid based on monthly surface temperature in JRA-25 after Streets et al. (2003a). Monthly variations of emission factors and removal efficiencies were ignored except for cold start emissions which depend on ambient temperature as described in Sect. 2.2.2. Emission factors for NMVOC evaporation have temperature dependencies, which will be considered in future work. In REAS 2.1, the monthly variation in agricultural emissions was not considered except for NO_x from soil in Asia and NH_3 in Japan (see Sect. 2.5). Yan et al. (2003c, 2005) developed global

soil NO_x emissions for 2001 with monthly variability and we used the same seasonality for all years between 2000 and 2008. Seasonal variation of agricultural emissions will be also considered in the next version of REAS.

With respect to grid allocation, we used the same methodology of REAS 1.1. As described in Sect. 2.2.1, emissions from power plants for which we had location information were allocated to the appropriate grid cell. We used the spatial distribution of rural, urban, and total populations and road network to allocate country- and sub-region-based emissions from area sources to grid cells. In REAS 2.1, population data were updated with the Global Rural-Urban Mapping Project version 1: Urban/Rural Extents with $30 \times 30''$ grid cells (GRUMPv1) and Gridded Population on the World, version 3 with $2.5 \times 2.5'$ grid cells (GPWv3) (CIESIN et al., 2005, 2011). Spatial distribution of total population for 2000, 2005, and 2010 were obtained from GPWv3 and interpolated for each year between 2000 and 2008. Using GRUMPv1 data for 2000, total population data were divided and aggregated to $0.25 \times 0.25^\circ$ urban and rural population data. We used the updated total population data to divide the road network data with $0.5 \times 0.5^\circ$, into $0.25 \times 0.25^\circ$ grid cells. In other words, the procedure for deriving surrogate data to allocate road transport emissions was not fundamentally changed. Considering the rapid motorization in Asian regions during recent years, the road network data should be updated in a future version of REAS.

2.3 Activity data

For most countries, energy consumption data for each fuel type including biofuels and sector categories were taken from the International Energy Agency (IEA) Energy Balances database (IEA, 2011). Total energy consumptions in India from IEA data were distributed to 17 sub-regions by using regional consumption ratios from the Greenhouse Gas and Air Pollution Interaction and Synergies (GAINS) INDIA database (IIASA, 2012). Asian Russia was treated similarly by using national energy consumption for Russia in IEA data and sub-regional data from Mastepanov (2001).

For countries whose energy consumptions were not presented in IEA, we used the United Nations (UN) Energy Statistics Database (UN, 2011).

As for China, after Akimoto et al. (2006) and Zhang et al. (2007), we selected province-level energy tables in the China Energy Statistical Yearbook (CESY; National Bureau of Statistics, 2004–2009) with the following modifications. Provincial diesel consumption values were adjusted by factors which are ratios of China's national consumption in IEA to the sum of provincial data in the CESY for each year. Motor gasoline consumption, which was listed in various categories in CESY, was assumed to be consumed in road transport sector except for consumption by agriculture/forestry. Fuel consumption in the industrial sector was distributed to sub-categories based on the statistical yearbook of each province. Data on production of primary and secondary fuels, which were used for estimating industrial process emissions and fugitive emissions, were obtained from the same sources as energy consumption. Note that REAS 1.1 used CESY only for coal, whereas other fossil fuel consumption data were obtained from IEA. Also, whereas biofuel consumption data for most countries were extrapolated from RAINS-Asia (IIASA, 2001) for the year 1995 and usage of crop residue as biofuel in China was estimated from Yan et al. (2006) in REAS 1.1. These changes may account for part of the difference in emissions between REAS 1.1 and 2.1 (see Sect. 2.5).

Both REAS 1.1 and 2.1 treat power plants as point sources. Although both versions overlap in their coverage from 2000 to 2003, information for point sources was fully updated for REAS 2.1. Figure 3 is a schematic diagram of the development of a new database of point sources that included their locations and fuel consumption of each type. First, CO_2 emissions in 2000 and 2007 and locations of power plants were collected from the Carbon Monitoring for Action (CARMA) Database (available at: http://www.carma.org) (Wheeler and Ummel, 2008). Second, information on generation capacity, fuel type, and start and retire years were extracted from the UDI World Electric Power Plants Database (WEPP) (Platts, 2009). Fuel consumption in 2000 and 2007 for each power plant was then estimated by using CO_2 emissions from CARMA

and emission factors for each fuel type from REAS (see Sect. 2.4). Finally, fuel consumption from 2000 to 2008 was estimated by using the trend of total fuel consumption in the power plant sector for each country and sub-region in which the power plant was located.

Activity data for sources other than energy came from a variety of international, national, and regional statistics and studies. For example, monthly production of iron and steel was obtained from the Steel Statistical Yearbook (World Steel Association, 2010). Annual production of non-ferrous metals and non-metallic minerals came from the US Geological Survey Minerals Yearbook (USGS, 2004–2008) and monthly data were available for several countries from statistics of each country. Vehicle numbers for cars, buses, trucks, and motor cycles were from the World Road Statistics (IRF, 2006–2010) and then subdivided into vehicle types by using the national and sub-regional statistics and database of the GAINS model. For China, numbers of vehicles of each type in 2000 were taken from Borken et al. (2008), and numbers from 2001 to 2008 were extrapolated by using trends from the China Statistical Yearbook (National Bureau of Statistics, 2001–2009). Agricultural activity data, such as numbers of livestock and amounts of applied fertilizers, were collected from the Food and Agriculture Organization Corporate Statistics Database (FAOSTAT) (FAO, 2011) and national and sub-regional statistics. The China Data Center of the University of Michigan, in its China Data Online resource, provided monthly statistics for each province such as production of steel, and cement, and power generation. We also used the basic data from the GAINS model when they were the only available information. If no activity data were found, we assumed that there were no emissions related to the activities, although this might instead be due to a lack of records.

2.4 Emission factors

REAS 1.1 obtained parameters such as emission factors and removal efficiencies from 1980 to 2003 from many sources, including Asian emission inventories although the number of inventories of Asian countries was limited. In REAS 2.1, we continued to

use the parameters of REAS 1.1 as default settings for 2000. Some parameters that were not part of REAS 1.1 needed to be determined for REAS 2.1. For Asian Russia and Central Asia, parameters from REAS 1.1 that were not country- or region-specific were used for default values. Default emission factors for PM_{10} and $PM_{2.5}$ were based on Klimont et al. (2002b) and AP-42 (US EPA, 1995) and for consistency, those for BC and OC were taken from Kupiainen and Klimont (2004). Default emission factors for total NMVOC and speciation factors for each NMVOC species were taken from Klimont et al. (2002a) and Streets et al. (2003a). In Asia, the influence of emission abatement equipment increased after 2000, especially for SO_2 and aerosols. In REAS 2.1, settings of the GAINS model for SO_2, PM_{10}, and $PM_{2.5}$ for 2000, 2005, and 2010 were adopted through linear interpolation as default settings. These were then updated for target countries and sub-regions after a survey of more recent studies when appropriate.

Many recently published emission inventories for China have been incorporated in REAS 2.1 as follows: Sulfur content in fuels, sulfur retention in ash, and penetration of flue gas desulfurization (FGD) from 2000 to 2008 were from Zhao et al. (2008; 2010) and Lu et al. (2010). Emission factors for NO_x CO, NMVOC, and primary aerosols (PM_{10}, $PM_{2.5}$, BC, and OC) except for road transport and cement production, were from Zhang et al. (2007), Streets et al. (2006), Wei et al. (2008) and Lei et al. (2011a), respectively. Emission factors for NO_x CO, and aerosols from cement production were based on Lei et al. (2011b). For road transport, temporal variations of emission factors due to control strategies and policies were estimated from 2000 to 2008 based on Borken et al. (2008) and Wu et al. (2011).

Emission inventories for countries other than China are still limited. Emission factors for India were updated as follows: NO_x and CO emission factors for power plants were from Chakraborty et al. (2008); SO_2 and NO_x emission factors for biofuel combustions were from Gadi et al. (2003) and Gurjar et al. (2004); emission factors for aerosols from fossil fuel combustion were from Reddy and Venkataraman (2002a); and those from biofuel combustions were from Reddy and Venkataraman (2002b) and Venkataraman et al. (2005). These updated emission factors for India were also used for other South

Asian countries. For Asian Russia and Central Asian countries, NO_x emission factors were taken from Ryaboshapko et al. (1996). We could not find other county- or region-specific emission factor.

2.5 Japan, South Korea, and Taiwan

As described in Sect. 2.1, REAS 2.1 used improved emission inventories for Japan, South Korea, and Taiwan from recent studies works with detailed information about activity data and parameters.

Except for the maritime sector, emissions in Japan were from the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) developed by JPEC (2012a, b, c). JEI-DB includes vehicle emissions in 2000, 2005, and 2010 and non-vehicle emissions in 2000 and 2005 for SO_2, NO_x, CO, NMVOC, PM, and NH_3, with monthly variations and spatial resolution of 1 km. Emissions between 2001 and 2004 and between 2006 and 2008 were derived by using the interannual variations of activity data and effects of emission regulations in Japan. Note that JEI-DB includes CO_2 emissions only from vehicles. Therefore, non-vehicle CO_2 emissions were estimated using the same activity data for other species and emission factors used for REAS 2.1. For NMVOC evaporative emissions from stationary sources, we used data developed by Ministry of the Environment of Japan (MOEJ, 2009). For the maritime sector, we used the database developed by the Ocean Policy Research Foundation (OPRF, 2012), which includes gridded emissions for 2005 from inland navigation, fishing fleets, and bunker fuel consumption by seagoing ships in Japanese national waters for SO_2, NO_x, CO, NMVOC, PM_{10}, CH_4, N_2O, and CO_2. For other years between 2000 and 2008, emissions were extrapolated based on reported energy consumptions.

For emissions from South Korea, we relied on the Clean Air Policy Support System (CAPSS) developed by Lee et al. (2011). City-level and province-level emissions for each sector and fuel type are available from the National Air Pollutants Emission website maintained by the National Institute of Environmental Research-Korea (http://airemiss.nier.go.kr). For REAS 2.1 we obtained emissions for SO_2, NO_x, CO,

NMVOC, and PM_{10} from 2000 to 2008 and allocated them to grids based on the population distribution within each city and province.

For Taiwan, we obtained data on SO_2, NO_x, CO, NMVOC, PM_{10} and $PM_{2.5}$ emissions developed by the Environmental Protection Administration of Taiwan at its website (http://ivy2.epa.gov.tw/air-ei). We obtained detailed emissions for each sector and fuel type in 2000, 2003 and 2007 as well as historical and projected total emissions between 1987 and 2021. We estimated emissions between 2000 and 2008 based on the trends of total emissions and then used the updated allocation factors described in Sect. 2.2.5 to distribute them into grids.

There are several data categories that were not included in these national inventories. Emissions of BC and OC (and $PM_{2.5}$ for Japan and South Korea) were converted from those of $PM_{2.5}$ (PM_{10} for Japan and South Korea) using known relations among emission factors for BC, OC, $PM_{2.5}$, and PM_{10}. We used the same methodologies described previously for other countries to develop emission datasets for NH_3, CH_4, and N_2O.

3 Results and discussion

3.1 Asian and national emissions for each species

Table 2 summarizes national emissions for each species in 2008, total annual Asian emissions from 2000 to 2008, and ratios of total Asian emissions between 2008 and 2000. Figures 4, 5, and 6 show the annual emissions from 2000 to 2008, divided by country/sub-region and by sector, for gaseous pollutant species, primary aerosols, and greenhouse gases, respectively. (In the supplement, Table S2 presents national emissions for 2000–2008, and Table S3 presents emissions for each sector and fuel type in 2000, 2004, and 2008 in each nation/sub-region). Table 3 lists the relative contribution of national/sub-regional emissions to total Asian emissions, and Table 4 does the same for the nations of Southeast Asia relative to the Southeast Asian sub-regional total.

3.1.1 SO_2

Total SO_2 emissions in 2008 (growth rate between 2000 and 2008) in REAS 2.1 are 56.9 Tg (+34 %) for Asia, 33.5 Tg (+46 %) for China, 10.1 Tg (+53 %) for India, 1.6 Tg (−12 %) for East Asia outside China (OEA), 4.2 Tg (+13 %) for Southeast Asia (SEA), 1.4 Tg (+18 %) for South Asia outside India (OSA), and 6.2 Tg (+2 %) for Asian Russia and Central Asia (RCA). The majority of Asian SO_2 emissions come from China (54–62 % during 2000–2008), followed by India (15–18 % during 2000–2008). The trends of total Asian emissions for this and most other species are dominated by China.

The fact that Asian SO_2 emissions increased monotonically from 2000 to 2006 and then began to decrease reflects trends of emissions in China and the power plant sector; growing number of coal-fired power plants have been equipped with FGD in China (Fig. 4a, b). REAS 2.1 used average FGD penetration rates in Chinese coal-fired power plants from Lu et al. (2010), which showed increase from 4 % in 2004 to 54 % in 2008. Whereas SO_2 and CO_2 emissions from power plants in China increased by 71 and 92 %, respectively from 2000 to 2005, sulfur emissions subsequently fell, while CO_2 emissions continued to climb. Thus in 2008, SO_2 emissions were 20 % greater than in 2000, while CO_2 emissions were 153 % greater. These tendencies were most prominent in Inner Mongolia, where many new power plants were constructed (Zhang et al., 2009b). Inner Mongolia CO_2 emissions increased by a factor of 4.7 from 2000 to 2008, whereas its SO_2 emissions between 2000 and 2008 increased less than 30 %.

Emissions from the industry sector in Asia increased almost monotonically between 2000 and 2008, and growth rates became larger after 2005. Industrial emissions from China and India showed similar trends. In India, emissions from power plants increased steadily from 2000 to 2008, thus, the growth rate of total SO_2 emissions from 2000 to 2008 was slightly larger for India than for China, although the absolute amounts were much larger for China. In SEA, SO_2 emissions have increased recently, although its contribution to the Asian total has been small compared to China and India. The largest contributor of SO_2 emissions in SEA was Indonesia (43 % in 2008) followed by Thailand

(16 % in 2008). In OSA the majority of emissions were from Pakistan (80 % in 2008). The contribution from RCA to the Asian total was relatively large for SO_2 emissions because of the output from large plants producing non-ferrous metals, such as copper and zinc, in Ural and Eastern Siberia (especially Norilsk). For Japan, South Korea, and Taiwan, total SO_2 emissions decreased from 2000 to 2008, by 12, 15, and 35 %, respectively. In Japan, emissions from the industrial sector increased slightly from 2000 to 2004 and decreased rapidly after 2005. Emissions from power plants increased but those from other sectors decreased slightly between 2000 and 2008. In South Korea, emissions from power plants and road transport decreased rapidly late in the target period, but emissions from the industrial sector increased slightly. Ship emissions in South Korea increased gradually. In Taiwan, emissions from power plants and industry decreased almost monotonically, whereas from other sources were almost constant from 2000 to 2008.

3.1.2 NO_x

Total NO_x emissions as NO_2 in 2008 (growth rate between 2000 and 2008) in REAS 2.1 are 53.9 Tg (+54 %) for Asia, 27.0 Tg (+89 %) for China, 11.1 Tg (+56 %) for India, 4.1 Tg (−14 %) for OEA, 5.5 Tg (+56 %) for SEA, 2.1 Tg (+30 %) for OSA, and 4.1 Tg (+14 %) for RCA. The largest contributors were China (41–50 % during 2000–2008) and India (about 20 % during 2000–2008). China's share of the Asian total was smaller for NO_x than for SO_2, but the ratios increased monotonically from 2000 to 2008. India's share of the Asian total was almost constant, but its emissions increased by 56 % from 2000 to 2008.

Year-to-year growth rates of Chinese NO_x emissions increased rapidly after 2002 but decreased slightly after 2005. One reason was the installation of low-NO_x boilers especially in new large power plants (Zhang et al., 2007). Thus, the growth rates of NO_x emissions from 2000 to 2008 (130 %) in the power plant sector were smaller than those of CO_2 (153 %). Again, this effect was pronounced in Inner Mongolia, although the effects of low-NO_x boilers were much smaller than the effects of FGD on SO_2.

Another reason is the implementation of new vehicle emission standards (Zhang et al., 2009a). Industrial emissions of NO_x in China increased rapidly rising to double the emissions in the road transport sector by 2008, although the two sectors were roughly equal in 2000.

In India, road transport emissions were the largest contributor to NO_x emissions (32 % in 2000 and 42 % in 2008) and doubled from 2000 to 2008. The contribution of power plants was almost constant (about 28 %). In SEA, rapid increases after 2005 were mainly due to road transport emissions in Indonesia, a reflection of the increased number of cars, buses, and trucks. In 2008, about half of Southeast Asian NO_x emissions were from Indonesia, 15 % from Thailand, and 11 % from Malaysia. Emissions in Pakistan increased until 2005, then remained almost constant from 2006 to 2008, the total growth rate being about 50 % from 2000 to 2008. Contributions to NO_x emissions in RCA were 66–70 % from Asian Russia and 14–19 % from Kazakhstan during the period. Total emissions increased slightly due to growth in the road transport sector. Total NO_x emissions in Japan, South Korea, and Taiwan decreased from 2000 to 2008, as did SO_2 emissions. In Japan, NO_x emissions were almost constant from 2000 to 2004 and decreased after 2005 as road transport emissions fell. Taiwan showed a similar pattern, although there were year-to-year variations between 2000 and 2003. Emissions in South Korea increased from 2000 to 2004 but then decreased due to reduced emissions from power plants.

In addition to fuel combustions, soil is an important source of NO_x emissions in Asia. The proportion of emissions from soil in the annual total during the period was 10–14 % for Asia, 7–11 % for China, 12–18 % for India, 3–4 % for OEA, 10–16 % for SEA, 25–31 % for OSA, and 23–26 % for RCA. Soil NO_x emissions have large monthly variations, with a peak in summer. Therefore, their relative importance depends on regions and seasons.

3.1.3 CO

Total CO emissions in 2008 (growth rate between 2000 and 2008) in REAS 2.1 were 359.5 Tg (+34 %) for Asia, 202.0 Tg (+42 %) for China, 61.8 Tg (+33 %) for India, 12.3 Tg (−15 %) for OEA, 48.3 Tg (+34 %) for SEA, 15.2 Tg (+26 %) for OSA, and 19.9 Tg (+20 %) for RCA. The largest contributors were China (53–57 % during 2000–2008) and India (16–18 % during 2000–2008) as was the case for SO_2 and NO_x. The contribution from SEA was also relatively large (13–14 % during 2000–2008). In 2000, Asian emissions from the domestic sector were the largest (42 %) followed by the industrial and road transport sectors (both 28 %). The power plants sector was not an important source for CO. Industrial emissions grew faster in Asia than from other sectors and becoming equal to the domestic sector at about 37 % after 2007.

These changes reflect the variation of CO emissions in China between 2000 and 2008. Industrial emissions in China more than doubled during this period, the increase being related to the production of steel, coke, cement, bricks, and similar commodities. Emissions increased from 2000 to 2006 at a slower rate in the domestic sector than in the industrial sector, but then began to decrease after 2007 as consumption of coal and biofuel for residential use decreased in China. This is one reason why growth rates of emissions in China decreased slightly in the latter part of the period. Other reasons are include the effects of regulations on road vehicle emissions and the substitution of shaft kilns for rotary kilns in cement production plants.

In India, CO emissions from the domestic, industrial, and road transport sectors increased monotonically between 2000 and 2008. The largest sector was domestic (53–59 % during 2000–2008), but contribution rates are decreasing. Growth rates of emissions in road transport sector were increasing and reached nearly 20 % of total emissions in 2008. In SEA, the majority of emissions were from biofuel combustion in the domestic sector, which showed an increasing trend. Furthermore, the growth rate of road transport emissions was much higher, and emission values more than doubled from 2000 to 2008. Indonesia was the largest contributing country (47 %) in

2008, followed by Vietnam and Thailand (16–17 %). In OSA, emissions generally increased with 56 % of the total coming from Pakistan, 16 % from Bangladesh, and 14 % from Nepal in 2008. The majority of CO emissions in OSA were from biofuel combustion. Emissions in RCA were almost constant from 2000 to 2005 and increased after 2006 due to emissions from the road transport sector. Contributions were dominated by Asian Russia (77–82 % during 2000–2008) and Kazakhstan (9–14 % during 2000–2008). CO emissions from Japan, South Korea, and Taiwan decreased from 2000 to 2008 by 28 %, 23 %, and 34 %, respectively. For these countries, emissions were mostly from the road transport sector and their values decreased monotonically from 2000 to 2008.

3.1.4 NMVOC

Total NMVOC emissions in 2008 (growth rate between 2000 and 2008) were 68.5 Tg (+46 %) for Asia, 27.1 Tg (+71 %) for China, 15.9 Tg (+37 %) for India, 3.1 Tg (−17 %) for OEA, 15.0 Tg (+47 %) for SEA, 3.7 Tg (+29 %) for OSA, and 3.7 Tg (+45 %) for RCA. During 2000–2008, China, India, and SEA contributed 34–40 %, 23–25 %, and 21–22 % of the Asian total, respectively. In 2000, emissions from the domestic sector were comparable to those from the transport sector (including evaporative emissions) and larger than those from solvent use. However, the growth rates for the road transport and solvent use sectors exceeded those for the domestic sector. By 2008, road transport emissions were the largest source (34 %), and solvent and paint use (24 %) was almost the same as domestic emissions in the Asia total.

In China, the road transport sector including evaporation was the largest NMVOC source (45 %) in 2000, but growth rates were moderate especially in later years, because of regulations on vehicles. Emissions from solvent use (including paint use) increased rapidly to 38 % in 2008, exceeding those from road transport (35 %). Emissions in Jiangsu, Guangdong, and Zhejiang provinces were disproportionate to NO_x emissions because of solvent and paint use. Domestic emissions in China, basically

from biofuel combustion increased slightly from 2000 to 2005 but then decreased after 2006.

In India, NMVOC emissions from biofuel combustion increased monotonically and were the largest contributor, but road transport emissions grew faster than other sectors. Thus the percentages contributed by the domestic and road transport sectors were 55 and 22 % in 2000, but became 43 and 35 %, respectively in 2008. Contributions from solvent and paint use were small in India. Similarly, in SEA both domestic and road transport emissions increased, but road transport grew much faster. The relative importance of road transport emissions increased from 30 % in 2000 to 42 % in 2008, while of the percentage contributed by the domestic sector decreased from 35 to 26 %. Nearly half of SEA emissions were from Indonesia, whereas 14 % were from Thailand, and 11 % each were from Malaysia and Vietnam. NMVOC emissions in OSA were mostly from biofuel combustion during 2000 and 2008 (60–66 %) and generally increased. In RCA, emissions from extraction and handling of oil and gas made up a much larger share of the total in Siberia (36–40 %) than in other regions (about 4 %). Because of increases in this sector, road transport, and solvent use, the growth rate in NMVOC between 2000 and 2008 was larger than for other species in RCA. In Japan, South Korea, and Taiwan, NMVOC emissions showed different trends. Emissions in Japan decreased almost constantly from 2000 to 2008 by 35 % because of reduced emissions from road transport, usage of paint and solvents. Emissions in South Korea increased slightly, particularly in paint and solvent use. Trends in Taiwan were small overall but mixed across sectors. Road transport emissions increased from 2000 to 2003 but started to decrease after 2004. Emissions from solvent use decreased from 2000 to 2008, and emissions from paint use increased rapidly after 2003.

3.1.5 NH_3

Total NH_3 emissions in 2008 (growth rate between 2000 and 2008) in REAS 2.1 were 32.8 Tg (+17 %) for Asia, 14.8 Tg (+18 %) for China, 9.4 Tg (+17 %) for India, 1.1 Tg (−2 %) for OEA, 4.2 Tg (+19 %) for SEA, 3.1 Tg (+21 %) for OSA, and 0.2 Tg (+6 %) for

RCA. As shown in Fig. 4j, the majority of NH_3 emissions in Asia come from agricultural activities (55–58 % from application of fertilizer and about 20 % from manure management). Like other species, Asian NH_3 emissions are dominated by China (about 45 %) and India (about 28 %). Emissions from South Asia (including India) account for about 38 % of the Asian total. This proportion is larger than it is for other species because of relatively larger contributions from Pakistan and Bangladesh (about 5 and 3 % in 2008, respectively).

Asian emissions show increasing trends during 2000 and 2008, although the increased amount was relatively small compared to other species. Year-to-year variations were basically controlled by emissions from fertilizer applications. In China, fertilizer-related emissions increased from 2000 to 2002, and decreased in 2003 and 2004, then increased again after 2005. In SEA, emissions related to fertilizer increased rapidly from 2001 to 2004, but varied little in other years.

As described in Sect. 2.2.3, NH_3 emissions from agricultural activities were extrapolated from the 2000 data of REAS 1.1 based on the amount of fertilizer usage and numbers of livestock. Therefore, interannual variability of emissions directly reflects the trends in statistics for fertilizer application and livestock. Emissions from latrines made considerable contributions to total emissions (about 13 %). They were the largest contributor (about 46 %) in RCA because emissions related to agricultural activities were smaller (about 29 %) than in other regions. Contributions from human perspiration and respiration were small for all regions (about 3 % in Asia). Emissions in Japan, South Korea, and Taiwan were also predominantly from agricultural activities, as in other Asian countries. Emissions in Japan decreased from 2000 to 2008 by 10 %. Emissions in Taiwan increased almost monotonically, growing by 18 % from 2000 to 2008. Emissions of South Korea in 2000 and 2008 were almost the same although there were relatively large year-to-year variations. Trends of NH_3 emissions in these countries mainly reflected fertilizer applications.

3.1.6 Primary aerosol emissions

This sub-section presents the results for PM_{10}, $PM_{2.5}$, BC, and OC. PM_{10} and $PM_{2.5}$ showed similar trends and contributions from each region and sector. Total emissions of PM_{10}/$PM_{2.5}$ in 2008 (growth rates between 2000 and 2008) in REAS 2.1 were 36.4/24.7 Tg (+45/+42 %) for Asia, 21.6/14.5 Tg (+54/+53 %) for China, 6.7/4.9 Tg (+41/+38 %) for India, 0.7/0.4 Tg (−3/−9 %) for OEA, 3.1/2.3 Tg (+11/+7 %) for SEA, 1.2/1.0 Tg (+39/+29 %) for OSA, and 3.1/1.7 Tg (+58/+48 %) for RCA. The major sources were the industry and domestic sectors. Both species had smaller emissions from the power plant sector. However, the relative contribution from each sector differed in each country.

In China, the industrial sector made the largest contributions to both PM_{10} and $PM_{2.5}$. (55–63 % and 46–57 % for PM_{10} and $PM_{2.5}$, respectively, for 2000–2008), and those contributions increased by 75 and 87 %, respectively. The proportion of emissions from cement production was large, but decreased from 2000 to 2008 although production of cement increased by a factor of 2.4. As was the case for CO, the shift to rotary kilns led to lower emission factors of PM_{10} and $PM_{2.5}$. Emissions from production of steel, coke, and non-ferrous metals increased. Emissions from the domestic sector increased from 2000 to 2005 and then decreased with the reduction in coal and biofuel consumption for residential use in China.

In India and OSA, domestic biofuel combustion accounted for the majority of emissions. In India, power plants ranked second with 27–30 % and 15–17 % for PM_{10} and $PM_{2.5}$, respectively. For OSA countries such as Pakistan, industrial emissions increased. For primary aerosol emissions in SEA, the largest contributing country was Indonesia, but Vietnam ranked second (18–21 % and 20–23 % of regional PM_{10} and $PM_{2.5}$ emissions, respectively), unlike the case of other species, because of its large consumption of biofuel. In Vietnam, about 60 % of PM_{10} and 70 % of $PM_{2.5}$ emissions were from domestic biofuel combustion in 2008. The majority of primary aerosol emissions in Indonesia and Malaysia were from the domestic and industry sectors,

respectively. In Thailand, emissions were mainly from the industrial sector, but contributions from power plants were relatively large. In RCA, more than 90 % of PM_{10} and $PM_{2.5}$ emissions were from production of iron, steel, and cement.

Mitigation of Short-Lived Climate Force (or Short-Lived Climate Pollutants) is considered to be important for air pollution reduction, climate protection, and sustainable development (Shindell, 2012). Black carbon is one of key species for this category. Black carbon absorbs visible light and contributes to warming of the atmosphere. It also causes health problems and premature death. Organic carbon is considered to have a cooling effect because it reflects incoming sunlight. In addition, BC and OC have many common emission sources. Asia is a large source of BC and OC emissions, and therefore, their accurate estimation is fundamentally important.

Total BC emissions in 2008 (growth rate between 2000 and 2008) were 3.03 Tg (+35 %) for Asia, 1.59 Tg (+40 %) for China, 0.71 Tg (+54 %) for India, 0.07 Tg (−26 %) for OEA, 0.37 Tg (+18 %) for SEA, 0.19 Tg (+21 %) for OSA, and 0.10 Tg (+25 %) for RCA. Compared to their PM_{10} and $PM_{2.5}$ emissions, China's contribution of BC was smaller and India's was slightly larger. Although the ratios of regional contributions of BC were similar to those for $PM_{2.5}$, the mix of contributions from different sectors was different. More than half of BC emissions were from the domestic sector, and the shares of domestic and road transport emissions were larger for BC than for $PM_{2.5}$. All countries showed similar tendencies.

In China, more than half of total BC emissions were from the domestic sector in 2000, but industrial and domestic emissions reached parity in 2008 (about 45 %). The road transport sector accounted for a larger share of the emissions of BC than of $PM_{2.5}$ but a smaller share than the domestic and industrial sectors (about 11 %). In India, the majority of emissions were from domestic biofuel combustion, but road transport emissions grew rapidly, increasing their share from 23 % in 2000 to 34 % in 2008. Pakistan was similar to India although its domestic sector was larger, and the growth rate of its road transport emissions was smaller than India's. Domestic biofuel combustion dominated emissions in Indonesia and Vietnam. In Thailand and Malaysia, more than half

of emissions were from the road transport sector. More than 60 % of emissions in RCA were from road transport, and the remainder was mostly from the industrial sector. In RCA, the domestic sector contributed less than 10 %, a relatively small share, because biofuel consumption was small.

Total OC emissions in 2008 (growth rate between 2000 and 2008) were 7.72 Tg (+21 %) for Asia, 3.08 Tg (+22 %) for China, 2.29 Tg (+30 %) for India, 0.05 Tg (−14 %) for OEA, 1.42 Tg (+9 %) for SEA, 0.67 Tg (+19 %) for OSA, and 0.21 Tg (+24 %) for RCA. OC emissions differed from other primary aerosols. Emissions in the domestic sector from biofuel combustion dominated the Asian total. Countries with large emissions such as China, India, Indonesia, and Vietnam showed similar features. In Thailand and Malaysia, which had smaller biofuel consumption, the majority of emissions were from industry (Thailand) or industry plus road transport (Malaysia). In RCA, the industrial sector dominated total emissions.

As described in Sect. 2.5, emissions of BC and OC from Taiwan were estimated from $PM_{2.5}$ and those of $PM_{2.5}$, BC, and OC from Japan and South Korea were estimated from PM_{10}. In Japan, primary aerosol emissions decreased almost constantly and by large amounts (29, 32, 45, and 39 % for PM_{10}, $PM_{2.5}$, BC, and OC, respectively, from 2000 to 2008) in response to trends in road transport emissions. The industrial and domestic sectors made considerable contributions to PM_{10} and $PM_{2.5}$ emissions, but their interannual variation was small. In South Korea, PM_{10} emissions were almost constant from 2000 to 2006 but then increased very rapidly because of industrial emissions; 2007 and 2008 totals were about 50 and 70 % greater than 2006, respectively. Trends for $PM_{2.5}$ were similar. Most BC and OC emissions were from the road transport sector, which increased from 2000 to 2004 but decreased rapidly after 2004. Total emissions were slightly smaller in 2008 than in 2000. In Taiwan, the majority of emissions of PM_{10} and $PM_{2.5}$ were from the industrial sector, which increased from 2000 to 2003 and then decreased rapidly. However, road transport emissions showed almost opposite tendencies, and as a result total emissions decreased slightly from 2000 to 2008. BC and OC

showed similar patterns, but the majority of emissions were from road transport and their trends were weaker than those of PM_{10} and $PM_{2.5}$.

3.1.7 Greenhouse gases (CH_4, N_2O, and CO_2)

Total CH_4 emissions in 2008 (growth rate between 2000 and 2008) in REAS 2.1 were 182.2 Tg (+32 %) for Asia, 76.0 Tg (+56 %) for China, 29.4 Tg (+8 %) for India, 4.7 Tg (+3 %) for OEA, 29.0 Tg (+18 %) for SEA, 10.6 Tg (+22 %) for OSA, and 32.5 Tg (+37 %) for RCA. China (35–42 %) and India (16–20 %) were the two largest contributors; however, contributions from SEA (16–18 %) and RCA (17–19 %) were also relatively large. Fugitive emissions from fossil fuels and agricultural emissions were the major sources for Asia as a whole. In China, emissions increased rapidly, and the largest contributions were from coal mining, mostly of which is underground mining where the CH_4 emission factor is large (IPCC, 2006). Mitigation of these emissions is considered to be an important measure to reduce Short-Lived Climate Forcers (Shindell, 2012). Emissions of CH_4 from coal mining in India were estimated to be small, although coal production is large. This is because we assumed that most coal production in India was by surface mining for which the emission factor for CH_4 is small (IPCC, 2006). More than 85 % of total emissions in RCA were from fugitive emissions from oil and gas activities especially in Western Siberia, Ural, and Kazakhstan. We estimated relatively large contributions (about 36 %) from fugitive emissions from fossil fuels in SEA. In SEA and South Asia (including India), the majority of emissions were from agricultural activities. In India, enteric fermentation and rice cultivation contributed 35–38 % and 21–25 %, respectively, emissions from 2000 to 2008. Tendencies in OSA were similar. Agricultural emissions in SEA were about 47 % of total emissions, a slightly smaller share than in South Asia due to the relative large contributions from fugitive emissions. In Japan, South Korea, and Taiwan, the majority of CH_4 emissions in 2008 were from solid waste disposal and waste water treatment: about 48 % for Japan, about 44 % for South Korea, and about 65 % for Taiwan. Interannual variation of total CH_4 emissions in these countries was relatively small. In other Asian countries,

the share of CH_4 emissions from waste treatments was about 13–15 % between 2000 and 2008 except for RCA where it was much smaller.

Total N_2O emissions in 2008 (growth rate between 2000 and 2008) in REAS 2.1 were 5.80 Tg (+18 %) for Asia, 2.66 Tg (+23 %) for China, 1.72 Tg (+14 %) for India, 0.25 Tg (−2 %) for OEA, 0.60 Tg (+13 %) for SEA, 0.52 Tg (+20 %) for OSA, and 0.05 Tg (+20 %) for RCA. Agricultural emissions, specifically direct and indirect soil and manure management sources, dominated N_2O emissions in Asia from 2000 to 2008: about 41 % from direct soil, about 33 % from indirect soil, and about 15 % from manure management. China made up about 45 % and India about 30 % of Asian emissions. India had a large share for this species than others. Japan and South Korea had larger proportions from industrial processes and road transport than other regions, although agricultural emissions were also large. In RCA, the contribution of agricultural emissions was exceptionally small (about 21 %), and emissions from fuel combustion sources were relatively large compared to other regions.

CO_2 emissions generally are a direct reflection of energy consumption, infrastructure buildup, and economic growth. REAS 2.1 includes emissions from combustion of fossil fuels and biofuels and emissions from industrial processes mainly cement production. Total CO_2 emissions in 2008 (growth rate between 2000 and 2008) in REAS 2.1 were 16.7 Pg (+59 %) for Asia, 9.5 Pg (+208 %) for China, 2.1 Pg (+36 %) for India, 2.1 Pg (+6 %) for OEA, 1.5 Pg (+33 %) for SEA, 0.4 Pg (+33 %) for OSA, and 1.2 Pg (+12 %) for RCA. The growth of CO_2 emissions in China was extremely high during 2000 and 2008, and its share of the Asia total increased from 43 % in 2000 to 57 % in 2008, although most other Asian countries also increased CO_2 emissions. More than 60 % of Chinese emissions were from coal combustion and its proportion increased from 2000 to 2008. Biofuel combustion went from about 12 % in 2000 to about 7 % in 2008, while industrial process emissions remained about 10 %. Other regions (except RCA) had larger relative contributions from biofuel combustion than China. Biofuel accounted for 27–33 % in India, 25–30 % in SEA, and 45–52 % in SA outside India from 2000 to 2008. The, largest contributor was coal combustion in India (39–46 %) and gas and oil

combustion in SEA (46–51 %). In RCA, emissions from gas and coal combustion were 39–43 % and 39–42 % of total CO_2 emissions, respectively. In Japan, total CO_2 emissions increased very little between 2000 and 2008, pattern consistent with its recent economic situation. Emissions in South Korea and Taiwan increased gradually about 19 % and 18 %, respectively, between 2000 and 2008. Emissions from power plants in these three countries increased almost monotonically except for Japan and Taiwan in 2008, probably because of the economic downturn precipitated by the Lehman Brothers bankruptcy.

3.2 Spatial distribution

Figure 7 shows the spatial distributions of annual emissions of SO_2, NO_x, BC, NMVOC, NH_3, and CH_4 in 2000 and 2008 at 0.25 × 0.25° resolution. The areas of highest emissions were in China and India for all species, especially in eastern China, Chongqing, Sichuan province, the Indo-Gangetic Plain, and southern India. These areas have large populations and significant economic and industrial activity. NH_3, CH_4, and N_2O, emissions were distributed over rural areas of intensive agricultural activity such as southern China and India. High CH_4 emissions also occurred over Ural, Western Siberia, and SEA because of the large fugitive emissions from gas- and oil-related activities. Spatial distributions of CO, PM_{10}, $PM_{2.5}$, OC, N_2O, and CO_2, emissions are presented in Fig. 1S in the supplementary materials.

3.3 Monthly variation

Figure 8 presents the distributions of monthly SO_2, NO_x and BC emissions in January and July 2008 for all of Asia. Monthly fractions for each sector and 2000–2008 trends for anthropogenic emissions of SO_2, NO_x (excluding soil emissions) and BC are shown in Fig. 9 for China and Fig. 10 for India.

For China, emissions from the power plant and industrial sectors generally increased throughout the year, in keeping with the general trend from 2000 to 2008 (Fig. 9). In

the power plant sector, small peaks during summer months probably reflect power generation for air conditioning. Small dips in February in both sectors appear to reflect the reduction of economic activity during the Chinese Spring Festival. The majority of SO_2 and NO_x emissions in China were from the power plant and industrial sectors, which exerted the predominant influence on monthly variation of total emissions. Note that emissions from power plants and industry decreased after the summer of 2008, probably because of the Lehman Brothers bankruptcy. Emissions from the domestic sector in China showed peaks during winter months because fuel consumption for residential heating was estimated on the basis of monthly surface temperature, as described in Sect. 2.2.5. These tendencies are especially evident in BC emission maps (Fig. 8e and f) in higher-latitude area. Winter peaks of smaller amplitude were also evident in NO_x and BC emissions in the road transport sector because of cold start emissions.

In India, emissions from the power plant and industrial sectors were lower during the summer and higher in winter, except for a dip in January (Fig. 10). These two sectors therefore govern the monthly variations of total SO_2 and NO_x emissions. As in China, monthly variations of total BC emissions reflected emissions from the domestic sector, emissions from which were larger in winter and lower in summer. In India, cold start emissions from road vehicles also added to the total BC emissions in winter. However, because winter temperatures are generally warmer in India than in China, the amplitude of these effects was much smaller.

Figure 11 presents the regional trends of monthly anthropogenic emissions of SO_2, NO_x, and BC between 2000 and 2008. As described in Sect. 2.2.5, monthly variations of emissions were determined only if monthly activity data or proxy data were available; otherwise, emissions were considered to be constant throughout the year. Therefore, these monthly variations often depend on residential and cold start emissions, whose monthly fractions are determined from surface temperature and can be determined for all countries and regions. That appears to be true in Fig. 11 for OSA and RCA, where monthly emissions of NO_x and BC are higher in winter and lower in summer. Emissions

in SEA were nearly constant within each year because seasonal variations of surface temperature are small and few data were available for estimating monthly fractions of other sectors. In OEA, monthly variations were basically governed by those in Japan, which were estimated from JEI-DB (Sect. 2.5), whereas very limited monthly data were available for other countries. Seasonal variations of SO_2 in Japan were generally small, whereas those of NO_x and BC were mainly controlled by emissions from the road transport sector and reflected cold start emissions in winter. Large gaps sometimes appear between December and January in Fig. 11 and will affect the results of simulations by chemical transport model. This issue should be improved by collection of continuous monthly data and use of inverse modeling.

In Fig. 8c and d, NO_x emissions were higher in July than in December, especially over rural and higher latitude areas. Most of these seasonal differences were caused by soil NO_x emissions, which respond to surface soil temperature and leaf area index (Yan et al., 2005). Although the contribution of soil sources to total NO_x emissions was about 10 % in China (see Sect. 3.1.2) the monthly variation was much greater than for other sources, as the average fraction ranged from about 0.01 in January and 0.21 in July. Seasonal variations of agricultural emissions, which were the majority of NH_3, CH_4, and N_2O emissions, are not considered in REAS 2.1 as described in Sect. 2.2.5 (except for NH_3 emissions in Japan). Better data on the seasonality of NH_3 emissions will improve the model reproducibility of aerosol concentrations, acidity of soils, and other environmental metrics.

3.4 Comparison with REAS version 1

Figure 12 compares the 2000 emissions of five Asian regions (China, India, OEA, SEA, and OSA) under REAS 1.1 and 2.1 (abbreviated as Rv1 and Rv2 in this section and Sect. 3.5). We selected the year 2000 because it is the base year of Rv1. There are several reasons for the differences between Rv1 and Rv2. For activity data such as energy consumption and industrial production, statistics were sometimes updated. As mentioned in Sect. 2.3, different sources were used for some of the energy data

in Rv1 and Rv2, such as consumption of fossil fuels other than coal in China and biofuels. Emission factors and removal efficiencies were updated (Sect. 2.4), and the methodology for estimating road transport emissions was changed (Sect. 2.2.2).

In China (Fig. 12a), SO_2 emissions were about 17 % smaller in Rv2 than in Rv1 because of the difference in industrial sectors. In Rv1, sulfur retention in coal ash after combustion in the industrial sector was at 15 %, but in Rv2 this value was changed to 25 % after Lu et al. (2010). For NO_x, differences in energy statistics, emission factors, and methodology for road transport have influenced the results for each sector, the results being increased emissions from the power plant, road transport and domestic sectors and reduced industrial emissions. The total NO_x emissions were about 10 % larger in Rv2 than in Rv1, but discrepancies between inventories in relative ratios of emissions from each sector were not large. Total emissions of CO, BC, and OC differed little between Rv1 and Rv2, but the relative sector contributions changed. Whereas the majority of CO emissions in the transport sector are from gasoline cars in both Rv1 and Rv2, the amount of emissions are much larger for Rv2. Emission factors for gasoline cars in China are almost the same in Rv1 and Rv2. Therefore traffic volumes in Rv2, as calculated by number of vehicles and annual distance traveled, are larger than in Rv1, as calculated by gasoline consumption and fuel economy. In addition, about 30 % of road transport emissions of CO for the year 2000 in Rv2 are from cold start emissions, which are not included in Rv1. For BC and OC, emissions from the industrial sector are larger and those from the domestic sector are smaller in Rv2 compared to Rv1. In the absence of emission factors, the latter did not include emissions from production of coke and bricks. However, these emissions are considered in Rv2 (after Lei et al., 2011a), where their contributions were about 60 % of the industrial sector. For the domestic sector, emission factors for BC from coal combustion and for OC from biofuel combustion were smaller in Rv2 than in Rv1. NMVOC emissions in Rv1 (Sect. 2.1) were developed by Klimont et al. (2002a) and Streets et al. (2003a), when activity data were mostly projected values. Therefore, updates of both emission factors and activity

data caused the difference in NMVOC emissions in each sector. This explanation can be applied not only to China but also to other countries and regions.

In India, SEA, and OSA (Fig. 12b, d, e, respectively), differences between Rv1 and Rv2 in SO_2 emissions from each sector were relatively small, although total emissions in Rv2 were slightly larger in India and smaller in OSA. For NO_x, road transport emissions in India and SEA were about 50 % larger and 30 % smaller, respectively, in Rv2 than in Rv1. Emission factors for diesel buses and trucks in India were about 60 % larger in Rv2 than in Rv1. Emission factors for SEA used in Rv1 and Rv2 were almost the same. Thus traffic volumes for SEA were smaller in Rv2 than in Rv1. For CO, BC, and OC, emissions from the domestic sector (mostly from biofuel combustion) were much smaller in Rv2 than in Rv1 for all three regions. Rv2 adopted smaller emission factors for biofuel combustions than Rv1 based on Gurjar et al. (2004) and Venkataraman et al. (2005). In addition, biofuel consumptions by India and OSA for Rv2, taken from IEA Energy Balances (IEA, 2011) were respectively about 30 % and 20 % smaller than those for Rv1. For NMVOC, emissions from stationary combustion sources in India and OSA were much higher for Rv2 than Rv1 because of high emission factors for biofuel, especially for dung cake (Gurjar et al., 2004).

Emission data for Japan, South Korea, and Taiwan were obtained from different source in Rv2 than in Rv1 (Sect. 2.5). Table S4 in the supplement shows the total emissions of SO_2, NO_x, CO, BC, OC, and NMVOC in these countries. For Japan, differences between Rv1 and Rv2 for SO_2 and NMVOC emissions were within 10 %, but there were large discrepancies for other species. NO_x emissions were about 35 % larger in Rv2 than in Rv1, mainly because of road transport emissions. CO emissions were much larger, by a factor of 2.6. The larger BC and OC emissions in Rv1 than Rv2 in Japan were mainly caused by sources other than the transport sector, although road transport emissions were also smaller in Rv2. Stationary emissions of BC and OC were respectively more than 3 and 60 % of total emissions in Rv1 and less than 20 and 30 %, respectively, in Rv2. For South Korea, emissions of all species were much larger in Rv1 than in Rv2. SO_2 emissions in the industry sector were larger in Rv1 than

in Rv2 by a factor of 3, and NO_x emissions in the road transport sector were twice as large in Rv1 as in Rv2. Emissions of CO, BC, and OC, from other than road transport were almost negligible in Rv2 but made relative contributions in Rv1 of about 70, 50, and 80 %, respectively. NMVOC emissions from solvent and paint use were almost unchanged between Rv1 and Rv2, but contributions from road transport and extraction and processing of fossil fuels were much larger in Rv1 than in Rv2. For Taiwan, emissions were larger in Rv1 than in Rv2 for all species except BC, for which road transport emissions were much smaller in Rv1 than in Rv2. Differences between Rv1 and Rv2 for SO_2 and CO were similar to those for South Korea. For OC, emissions from the industrial and transport sectors were almost the same in Rv1 and Rv2, whereas those from other sectors were larger in Rv1 than in Rv2. Differences of NO_x emissions were relatively small. NMVOC emissions in Rv2 were about 50 % larger than in Rv1 due to differences in fuel combustion and solvent use sources. In North Korea and Mongolia, emissions of BC and OC are much smaller in Rv2 than in Rv1, a response to decreased biofuel consumption in the activity data of Rv2 compared to Rv1.

3.5 Comparison with other inventories

Table 5 summarizes emissions of SO_2, NO_x, CO, NMVOC, BC, and OC in China estimated by Rv2 and several other published inventories. The year-to-year variation of SO_2 in Rv2 is very similar to that of Lu et al. (2010). This similarity is reasonable because Lu et al. (2010) was the source of the penetration ratios of FGD in power plants that strongly affected the trends of total SO_2 emissions. The amounts of SO_2 emissions are slightly larger in Rv2 than in Lu et al. (2010). EDGAR 4.2 (EC-JRC/PBL, 2011; hereafter EDGAR) does not appear to consider the effects of FGD penetration sufficiently fast. Emission factors of NO_x were mainly from Zhang et al. (2007). Therefore, emissions in Rv2 during 2000–2004 and 2006 were similar to those in Zhang et al. (2007 and 2009a), respectively. Compared to EDGAR, emissions of Rv2 were larger, especially after 2005 (nearly 30 % larger). For CO, amounts of emissions in 2001 were very similar in Streets et al. (2006), Ohara et al. (2007), Zhang et al. (2009a) and Rv2.

Emission factors in Rv2 were primarily from Streets et al. (2006). CO emissions for 2006 were about 15 % larger in Rv2 than in Zhang et al. (2009a). Emissions of CO in EDGAR were about half of the emissions in other inventories, but the growth rates between 2000 and 2008 for Rv2 and EDGAR were almost the same (about 42 %), indicating that the applied emission factor derived from common European techniques seems not valid for Asian technology. All NMVOC emissions in 2000 in Table 6 are within 15 % except for Bo et al. (2008), whose results are smaller than others results in 2000 and 2005. Compared to Zhang et al. (2009a), NMVOC emissions in Rv2 are smaller in 2001 but larger in 2006, the result being that estimated growth rates in Rv2 are larger than in Zhang et al. (2009a). The growth rate for NMVOC between 2000 and 2008 in Rv2 (about 70 %) was much larger than in EDGAR (about 30 %). With respect to aerosols, BC emissions in Rv2 were very similar to those of Qin and Xie (2012) as well as Lei et al. (2011a) and Lu et al. (2011). BC emissions for 2001 and 2006 in RV2 were both smaller than in Zhang et al. (2009a), but growth rates for that period were larger (about 30 % compared to about 15 %). All OC emissions for 2000 showed similar values except for Cao et al. (2006). BC and OC emissions in 2000 estimated by Cao et al. (2006) were about 25 and 50 % larger, respectively, than in Rv2. From 2001 to 2008, differences in OC emissions between Rv2 and other inventories were less than 15 %. However, trends of Rv2 were almost the same in 2004 and 2008, whereas those of Lu et al. (2011) increased by about 8 % from 2004 to 2008. OC emissions in Rv2 decreased after 2006 because of the reduction of biofuel consumption in China.

Table 6 lists the emissions in India estimated by Rv2 and other inventories. For SO_2, NO_x, and NMVOC, emissions in Rv2 were higher than other inventories all through the period. SO_2 emissions of Streets et al. (2003a), Lu et al. (2011) and EDGAR are relatively similar but about 15–25 % smaller than the Rv2 emissions. Growth rates between 2000 and 2008 were about 38, 48, and 53 % in Lu et al. (2011), EDGAR, and Rv2, respectively. SO_2 emissions of Garg et al. (2006) and Zhang et al. (2009a) are much smaller than the Rv2 emissions, perhaps because Rv2 uses larger net emission factors. The NO_x and NMVOC emissions have similar patterns for SO_2 across the

inventories. In addition, growth rates from 2000 to 2008 in Rv2 (about 66 and 37 % for NO_x and NMVOC, respectively) were much larger than those of EDGAR (about 40 and 11 %). The rate of increase of road transport emissions in Rv2 is much larger than in EDGAR. For CO, emissions in Rv2 and other inventories were all within 15 % in 2000, except for Ohara et al. (2007); however, the growth rate from 2000 to 2008 was larger in Rv2 (about 33 %) than in EDGAR (about 11 %). BC emissions were smaller in Rv2 than in other inventories except for Zhang et al. (2009a), whose emissions in 2006 are about 45 % smaller than the Rv2 emissions. BC emissions of Lu et al. (2011) are about 47, 38, and 25 % larger than Rv2 for 2000, 2004, and 2008, respectively. Therefore, growth rates from 2000 to 2008 in Rv2 (about 54 %) are larger than Lu et al. (2011) (about 31 %). For OC, emissions estimated by Rv2 were within 5 % of those in Lu et al. (2011) during 2000 and 2008. OC emissions of Zhang et al. (2009a) are much smaller, but those of Streets et al. (2003a) are larger than Rv2 for India.

Table 7 presents emissions from Rv2 and other inventories for OEA, SEA, and OSA. For OEA, the SO_2, CO, and NMVOC emissions of both Rv2 and EDGAR decrease from 2000 to 2008, the decrease in EDGAR is much greater than in Rv2. Net emission factors, including removal efficiencies, are thought to be much higher in EDGAR than in Rv2, whose emissions for Japan, South Korea, and Taiwan are obtained from detailed studies (see Sect. 2.5). Results of Rv2 for these species generally agree with Streets et al. (2003a) and Zhang et al. (2009a). Differences in NO_x emissions between Rv2 and other inventories are relatively small. Large differences in BC emissions between Rv2 and Zhang et al. (2009a) are due to estimations for Taiwan. OC emissions of Rv2 are much smaller than those in Streets et al. (2003a) and Zhang et al. (2009a) because Rv2 has lower estimated emissions for North Korea. For SEA, results in Rv2 generally agree with Streets et al. (2003a) and Zhang et al. (2009a) within 15 % except that the SO_2 and NO_x emissions for 2006 of Zhang et al. (2009a) are about 30 % larger than those in Rv2. Emissions of SO_2, NO_x, CO, and NMVOC in Rv2 agree with those in EDGAR within 10 % for 2000. SO_2 emissions between 2000 and 2008 show similar trends in EDGAR and Sv2, but growth rates for other species were much larger for Rv2.

With respect to OSA, emissions of Rv2 are larger than those of EDGAR for all species. In general, however, their trends are similar, especially for SO_2, although growth rates between 2000 and 2008 for NO_x, CO, and NMVOC in Rv2 are slightly larger than in EDGAR. Compared to Streets et al. (2003a) and Zhang et al. (2009a), emissions in Rv2 are much smaller for SO_2 but larger for NO_x and NMVOC. However, CO, BC, and OC emissions are generally consistent in all inventories.

Table 8 presents NH_3, CH_4, and N_2O emissions estimated by Rv2 and other inventories for China, India, and other parts of Asia (OA) which exclude RCA. The NH_3 and N_2O emissions in Rv2 are much larger than those in EDGAR, result arising from differences in agricultural emissions related to manure management and agricultural soil. Differences in NH_3 emissions between Rv2 and Streets et al. (2003a) are relatively small, about 6, 11, and 16 % for China, India, and OA, respectively. With respect to CH_4, Rv2 and EDGAR agree very well in their amounts of emissions and year-to-year variations. However, differences of CH_4 emissions between Rv2 and Streets et al. (2003a) are larger than those of NH_3. CH_4 emissions of Rv2 in China and OA are about 29 and 13 % larger than, respectively, than those in Streets et al. (2003a), and those in India are about 16 % smaller.

3.6 Uncertainty

In REAS 2.1, country and regional emissions were calculated using activity data, emission factors and removal efficiencies. For emission factors and removal efficiencies, we updated the parameters using recently published literatures for Asian emission inventory, especially for China as described in Sect. 2.4. Therefore, uncertainties are smaller than REAS 1.1. However, country- and region-specific information is not enough especially for Southeast and South Asian countries. Further investigation of literatures and cooperation with researchers in these countries are essential to reduce the uncertainties of Asian emission inventory. In general, uncertainties of emission factors for complete combustion sources are relatively small, whereas those for imperfect combustion and non-combustion sources such as evaporation are larger.

For activity data of stationary sources, we relied on international, national, and regional statistics. Uncertainties in fossil fuel consumption and production of major industrial products such as metal and cement are expected to be smaller because these are basic statistics for each country. Especially, as described in Sect. 2.3, we fully updated the database for power plants as point sources with detailed information and thus, their uncertainties are smaller than other sources. On the other hand, biofuel consumption data in IEA statistics are the sum of fuel wood, crop residue, and animal waste. We distributed the data to each fuel type referring limited information such as Streets et al. (1998) and database of the GAINS model. Therefore, uncertainties of biofuel consumption are larger than fossil fuels. For industrial production, basic information for brick production in Asia is not enough. UNEP (2011) indicated that brick kiln is one of major sources to reduce BC emissions for mitigating near-term climate change and improving air quality. Therefore further survey of brick production is required to improve the accuracy of BC emissions especially for the support of policy-making. For road transport sector, national statistics of most countries have data for number of registered vehicles and average annual distance traveled for China were updated by new information as described in Sect. 2.2.2. In addition, REAS 2.1 included cold start emissions. Therefore, uncertainties for road transport emissions in China are improved compared to REAS 1.1. However, annual distance traveled for other countries were still mostly taken from REAS 1.1. Situation of road traffic is changing and emissions from road transport became important not only in China but also in other Asian countries. Therefore, road transport sector is one of major causes of uncertainty especially in Southeast and South Asian countries.

REAS 2.1 improved both spatial and temporal resolution. For large power plants, we updated not only position data but also start and retired year. In addition, we obtained statistics for monthly generated power in China, India, and Vietnam. Accuracy of monthly gridded data for power plants sector was increased from REAS 1.1. We also updated population distribution data for finer resolution and monthly variation of emissions from heating stoves is considered using objective analysis meteorological

data. Therefore, uncertainty of monthly gridded emission data for residential sector was also decreased. For industrial sector, we used statistics for monthly production of major industrial products such as cement and metals to estimate monthly emissions for several countries. However, we could not obtained information for large industrial plants such as position, production amounts and fuel consumption. For transport sector, we could not update both proxy data for grid allocation and monthly variation of emissions. Improvements of accuracy for spatial distribution and temporal variation for these sectors are required especially for atmospheric chemistry model simulations and are major issues for the next version of REAS.

Uncertainties of emissions are different from each species, source, and country and region in Asia. Generally, uncertainties of SO_2, NO_x, and CO_2 emissions whose major sources are complete combustions are relatively small, whereas those of CO and primary aerosols basically from imperfect combustions are relatively large. For NMVOC, uncertainties for fossil fuel combustion sources are small, but those for biofuel combustion and evaporative emissions are large. For NH_3, CH_4, and N_2O, major emission sources are agricultural activities. As described in Sect. 2.2.3, agricultural emission data were extrapolated from the gridded data of REAS 1.1 for 2000. In addition, seasonal variations were also not considered in REAS 2.1. Therefore, uncertainties of emissions for these species are relatively large. Note that emissions from Japan, South Korea, and Taiwan were taken from recent research works with detailed information about activity data and parameters as described in Sect. 2.5. Uncertainties of emissions in these countries are expected to be small.

3.7 Data distribution

We have prepared a data download site for REAS 2.1 at the following URL: http://www.nies.go.jp/REAS/. Monthly gridded emission data sets at $0.25 \times 0.25^{\circ}$ resolution for each major sector can be downloaded for all species. As was true for REAS version 1, NMVOC emissions are speciated to 19 species (ethane, propane, butanes, pentanes, other alkanes, ethylene, propene, terminal alkenes, internal alkenes, acetylene,

benzene, toluene, xylenes, other aromatics, formaldehyde, other aldehyde, ketones, halocarbons, and others). We also provide country and regional emission tables that present annual emissions for detailed sub-sectors and fuel types.

4 Summary

We developed a Regional Emission inventory in ASia (REAS) 2.1 by updating REAS 1.1 of Ohara et al. (2007). The new inventory includes most major air pollutants and greenhouse gases: SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC, NH_3, CH_4, N_2O, and CO_2. The target years are from 2000 to 2008, and areas have been expanded from East, Southeast and South Asia to encompass Central Asia and Asian Russia (Ural, Western and Eastern Siberia, and Far East). Emissions are estimated for each country and region and are allocated to grids at a $0.25 \times 0.25^{\circ}$ resolution with monthly variation. Activity data such as energy consumption and industrial production were obtained in many cases from new statistics and the database of power plants as point sources was fully updated. We collected country- and region-specific parameters such as emission factors and removal efficiencies from recently published studies. For Japan, South Korea, and Taiwan, we used the newest available national inventories based on detailed activity data and information (JPEC, 2012a, b, c; OPRF, 2012; Lee et al., 2011; Environmental Protection Administration of Taiwan).

Emissions from all of Asia for each species in 2008 (growth rate from 2000 to 2008) were 56.9 Tg (+34 %) for SO_2, 53.9 Tg (+54 %) for NO_x, 359.5 Tg (+34 %) for CO, 68.5 Tg (+46 %) for NMVOC, 32.8 Tg (+17 %) for NH_3, 36.4 Tg (+45 %) for PM_{10}, 24.7 Tg (+42 %) for $PM_{2.5}$, 3.03 Tg (+35 %) for BC, 7.72 Tg (+21 %) for OC, 182.2 Tg (+32 %) for CH_4, 5.80 Tg (+18 %) for N_2O, and 16.7 Pg (+59 %) for CO_2. For all species, China was the largest contributor to Asian emissions, and its growth rates in emissions were also the largest because of the continuous increase in its energy consumption, economic activities, and infrastructural development. Emission mitigation measures were implemented gradually in China during the target years in REAS 2.1.

SO_2 emissions in China increased from 2000 to 2006 and then decreased because of increasing penetration of FGD to large power plants. New vehicle emission standards are also becoming effective against NO_x, CO, NMVOC, and primary aerosol emissions. However, emissions of these species in China still increased monotonically, and growth rates were large from 2000 to 2008. The second largest contributor to Asian emissions was India, and its emissions also grew rapidly from 2000 to 2008. This is because consumption of fossil fuels and biofuels, industrial production, and the number of vehicles were increasing rapidly while emission regulation measures in India were assumed to be limited. Emissions of air pollutants in East Asia outside China decreased from 2000 to 2008. This region's emissions were dominated by Japan, South Korea, and Taiwan, where economic growth rates are smaller than China and India and regulation of emissions has become more effective since 2000. Although the proportional contributions of other regions in Asia are small compared to China and India, emissions in Southeast Asia, South Asia outside India, Central Asia, and Asian Russia generally increased from 2000 to 2008. Indonesia and Pakistan were the largest contributing countries to emissions in Southeast Asia and South Asia outside India, respectively. Emissions from the road transport sector were increasing in Southeast Asia. The relative contribution of SO_2 emissions from Asian Russia was large because of non-ferrous metal production in Ural and Eastern Siberia regions, especially in Norilsk.

The areas of greatest emissions were in China and India, especially in eastern China and the Indo-Gangetic Plain, where there are large populations and vigorous economic and industrial activity. Emissions of NH_3, CH_4, and N_2O were distributed over rural areas dominated by agriculture. High CH_4 emissions occurred in Ural, Western Siberia, and Southeast Asia as a result of gas- and oil-related activities. Noteworthy seasonal variations included a winter peak in emissions, such as primary aerosols and CO, especially in northern locations with cold winters. Contributions to these emissions from residential stoves were large. There was also a large summer peak in soil NO_x emissions, the controlling factors being surface soil temperature and leaf area index.

To improve this bottom-up emission inventory, it is essential to get critical feedback from users of the data, especially researchers who use atmospheric chemistry models including inverse modeling and observation data. In addition, there is a requirement for continuous improvement of basic activity data, country- and region-specific emission factors, and information related to emission regulations. For our next steps, we plan to prepare projections of future emissions based on several scenarios, especially for air quality and climate change studies of the Asian region. We also plan to update historical data and extend the last year of the inventory.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/13/10049/2013/acpd-13-10049-2013-supplement.pdf.

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Table 1. General information on REAS 2.1.

Item	Description for targets
Species	SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC, NH_3, CH_4, N_2O, and CO_2
Years	2000–2008
Areas	East, Southeast, South, and Central Asia. Asian part of Russia (Far East, Eastern and Western Siberia, and Ural)
Emission sources	fuel combustions 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	$0.25^\circ \times 0.25^\circ$
Temporal resolution	monthly
Data distribution http://www.nies.go.jp/REAS/	

Table 2. Summary of national emissions in 2008 for each species ($Gg\,yr^{-1}$), total annual emissions in Asia from 2000 to 2008, and ratios of total Asian emissions between 2008 and 2000.

Country	SO_2	NO_x[a]	CO	NMVOC	PM_{10}	$PM_{2.5}$
China	33 457	26 969	201 967	27 098	21 606	14 514
Japan	761	2207	5029	1317	130	94
Korea, Rep of	417	1059	690	857	110	56
Korea, DPR	226	288	5137	158	291	128
Mongolia	73	136	661	46	78	33
Taiwan	128	442	740	687	86	54
Brunei	11	11	6	31	1	1
Cambodia	31	73	1007	207	58	55
Indonesia	1808	2817	22 499	7316	1327	997
Laos	140	61	386	82	24	22
Malaysia	357	619	3454	1680	210	132
Myanmar	54	196	2651	724	160	152
Philippines	436	349	2286	842	169	114
Singapore	177	114	156	310	7	5
Thailand	678	851	8208	2144	483	267
Vietnam	520	458	7671	1660	650	520
Bangladesh	126	434	2444	758	313	218
Bhutan	4	18	283	46	23	19
India	10 077	11 061	61 803	15 946	6651	4884
Nepal	30	104	2080	425	146	135
Pakistan	1133	1160	8596	1978	570	529
Sri Lanka	111	141	1313	372	138	106
Afghanistan	3	207	387	122	18	17
Maldives	3	10	146	8	0	0
Far East[b]	349	633	2599	298	228	122
East Siberia[b]	1600	671	2782	387	385	204
West Siberia[b]	639	965	5920	1284	479	274
Ural[b]	1492	432	4011	586	1088	618
Kazakhstan	1409	756	2885	522	439	220
Kyrgyzstan	34	50	300	41	69	31
Tajikistan	15	36	194	29	24	14
Turkmenistan	57	248	371	232	60	28
Uzbekistan	560	300	860	307	374	165
Asia[c] 2000	42 315	34 915	267 431	46 787	25 088	17 445
Asia[c] 2001	43 193	35 922	270 518	48 412	25 904	18 059
Asia[c] 2002	45 361	37 463	279 525	50 537	26 925	18 886
Asia[c] 2003	48 123	39 933	291 557	53 370	28 423	19 905
Asia[c] 2004	51 973	42 944	305 790	57 090	30 305	21 113
Asia[c] 2005	56 862	46 124	327 213	59 889	32 174	22 337
Asia[c] 2006	58 659	48 795	335 547	63 310	33 923	23 350
Asia[c] 2007	58 285	51 833	347 743	66 710	35 819	24 446
Asia[c] 2008	56 913	53 875	359 525	68 501	36 397	24 729
Asia[c] 2008/2000	1.34	1.54	1.34	1.46	1.45	1.42

Table 2. Continued.

Country	BC	OC	NH_3	CH_4	N_2O	CO_2[d]
China	1589	3081	14 844	75 975	2661	9478
Japan	26	10	483	1992	81	1192
Korea, Rep of	13	9	190	1171	82	532
Korea, DPR	15	18	106	664	19	78
Mongolia	1	2	120	465	36	12
Taiwan	11	6	160	385	29	271
Brunei	0	0	8	544	2	8
Cambodia	11	43	126	768	22	17
Indonesia	179	682	1743	11 398	219	573
Laos	4	16	84	353	12	6
Malaysia	15	34	238	2733	49	206
Myanmar	31	123	421	3146	65	48
Philippines	14	68	415	1708	66	103
Singapore	1	1	11	66	3	40
Thailand	33	142	580	4347	92	280
Vietnam	83	312	572	3909	68	206
Bangladesh	32	117	936	3957	111	76
Bhutan	3	13	43	141	11	4
India	713	2286	9421	29 431	1721	2103
Nepal	26	102	245	852	42	33
Pakistan	108	374	1595	4925	301	245
Sri Lanka	16	59	125	376	14	29
Afghanistan	7	9	137	368	45	2
Maldives	0	0	0	3	0	2
Far East[b]	19	23	19	627	4	109
East Siberia[b]	12	19	23	1441	5	170
West Siberia[b]	29	50	41	19 697	10	310
Ural[b]	18	73	22	3682	5	172
Kazakhstan	12	28	41	3980	8	208
Kyrgyzstan	2	3	13	77	1	7
Tajikistan	1	1	15	82	1	4
Turkmenistan	2	3	14	1470	5	48
Uzbekistan	3	11	55	1491	10	128
Asia[c] 2000	2240	6385	28 013	137 694	4928	10 523
Asia[c] 2001	2311	6571	28 246	140 022	4964	10 747
Asia[c] 2002	2433	6908	29 263	145 159	5074	11 299
Asia[c] 2003	2533	7075	29 455	152 874	5124	12 043
Asia[c] 2004	2618	7238	29 686	159 960	5207	13 019
Asia[c] 2005	2740	7369	30 410	166 577	5351	14 094
Asia[c] 2006	2832	7457	31 649	172 087	5595	15 073
Asia[c] 2007	2942	7573	32 340	174 669	5699	16 119
Asia[c] 2008	3029	7719	32 843	182 224	5804	16 701
Asia[c] 2008/2000	1.35	1.21	1.17	1.32	1.18	1.59

[a] $Gg\,NO_2\,yr^{-1}$. [b] Asian Russia. [c] Asia in this table include all target countries and sub-regions in REAS 2.1. [d] $Tg\,yr^{-1}$.

Table 3. Proportion of total Asian emissions from sub-regions of Asia (% 2000/2008).

	SO_2	NO_x	CO	NMVOC	PM_{10}	$PM_{2.5}$
China	54.3/58.8	40.9/50.1	53.0/56.2	33.8/39.6	55.9/59.4	54.3/58.7
India	15.5/17.7	20.3/20.5	17.3/17.2	24.8/23.3	18.8/18.3	20.3/19.8
East Asia except for China	4.3/2.8	13.7/7.7	5.4/3.4	7.9/4.5	2.8/1.9	2.3/1.5
Southeast Asia	8.8/7.4	10.2/10.3	13.5/13.4	21.9/21.9	11.1/8.5	12.1/9.2
South Asia except for India	2.8/2.5	4.6/3.8	4.5/4.2	6.1/5.4	3.5/3.3	4.5/4.1
Asian part of Russia and Central Asia	14.2/10.8	10.3/7.6	6.2/5.5	5.4/5.4	7.9/8.6	6.5/6.8
	BC	OC	NH_3	CH_4	N_2O	CO_2
China	50.7/52.5	39.6/39.9	44.8/45.2	35.4/41.7	43.9/45.9	43.3/56.8
India	20.6/23.5	27.6/29.6	28.8/28.7	19.8/16.2	30.5/29.7	14.7/12.6
East Asia except for China	4.0/2.2	0.8/0.6	3.9/3.2	3.3/2.6	5.1/4.3	18.8/12.5
Southeast Asia	14.1/12.3	20.4/18.4	12.6/12.8	17.9/15.9	10.7/10.3	10.6/8.9
South Asia except for India	7.1/6.3	8.9/8.7	9.1/9.4	6.3/5.8	8.9/9.0	2.8/2.3
Asian part of Russia and Central Asia	3.5/3.2	2.7/2.7	0.8/0.7	17.2/17.9	0.9/0.8	9.9/6.9

Table 4. Proportions of total Southeast Asian emissions from individual Southeast Asian countries (%, average of 2000 and 2008).

	SO_2	NO_x	CO	NMVOC	PM_{10}	$PM_{2.5}$
Brunei	0.2	0.2	0.0	0.2	0.1	0.0
Cambodia	0.6	1.9	2.4	1.6	1.8	2.4
Indonesia	38.8	42.9	43.3	44.8	40.9	42.4
Laos	1.4	1.2	0.9	0.6	0.8	1.0
Malaysia	7.2	11.4	6.9	11.2	6.6	5.9
Myanmar	2.1	4.5	6.1	4.9	5.2	6.7
Philippines	13.8	8.2	5.6	6.5	6.6	6.2
Singapore	5.8	2.8	0.3	2.1	0.3	0.3
Thailand	19.6	17.9	17.5	16.1	17.9	13.5
Vietnam	10.5	8.9	17.0	12.0	19.8	21.7
	BC	OC	NH_3	CH_4	N_2O	CO_2
Brunei	0.0	0.0	0.2	2.1	0.4	0.5
Cambodia	3.2	3.0	2.8	2.6	3.4	1.2
Indonesia	45.5	47.9	39.5	39.8	36.0	38.9
Laos	1.2	1.2	1.9	1.2	1.9	0.5
Malaysia	3.9	2.4	5.4	9.7	7.9	12.4
Myanmar	8.6	8.7	9.6	10.3	10.7	3.5
Philippines	5.0	5.6	10.4	6.0	11.6	7.8
Singapore	0.3	0.0	0.2	0.2	0.5	3.2
Thailand	9.6	9.5	13.5	14.6	15.3	19.3
Vietnam	22.6	21.7	16.5	13.5	12.5	12.9

Table 5. Published estimates of emissions of SO_2, NO_x, CO, NMVOC, BC, and OC from China ($Tgyr^{-1}$).

	2000	2001	2002	2003	2004	2005	2006	2007	2008
SO_2									
Streets et al. (2003a)	20.3								
Ohara et al. (2007)	27.6	29.3	31.9	36.6					
Zhang et al. (2009a)		22.9					31.0		
Lu et al. (2010)	21.7	22.7	24.0	26.7	28.8	32.3	33.2	32.3	31.3
Lu et al. (2011)	21.1				30.5				32.0
EDGAR 4.2	20.1	19.9	20.7	23.7	27.6	30.1	32.5	34.7	40.3
This work	23.0	23.3	25.2	28.1	31.1	35.2	35.5	34.6	33.5
NO_x									
Streets et al. (2003a)	10.5								
Ohara et al. (2007)	11.2	11.8	12.7	14.5					
Zhang et al. (2007)	12.6	13.2	14.4	16.2	18.6				
Zhang et al. (2009a)		13.4					20.8		
EDGAR 4.2	11.2	11.1	11.5	13.1	14.7	15.8	17.0	18.1	20.0
This work	12.7	13.3	14.3	15.8	17.9	20.4	22.0	23.9	25.1
CO									
Streets et al. (2003a)	100.0								
Streets et al. (2006)		141.7							
Ohara et al. (2007)	137.0	140.6	146.3	158.3					
Zhang et al. (2009a)		141.6					166.9		
EDGAR 4.2	73.9	73.5	74.7	77.6	84.2	89.1	93.8	98.6	105.1
This work	141.8	142.8	149.4	158.1	168.5	186.6	190.2	196.1	202.0
NMVOC									
Klimont et al. (2002a)	15.6								
Streets et al. (2003a)	14.7								
Ohara et al. (2007)	14.7	15.5	16.1	17.2					
Bo et al. (2008)	10.2					15.6			
Wei et al. (2008)						19.2			
Zhang et al. (2009a)		18.1					23.2		
EDGAR 4.2	16.8	17.0	17.4	18.0	19.1	19.8	20.6	21.1	22.2
This work	15.8	16.7	17.8	19.3	21.8	23.3	25.3	26.8	27.1
BC									
Streets et al. (2003a)	0.94								
Cao et al. (2006)	1.40								
Ohara et al. (2007)	1.09	1.10	1.11	1.14					
Zhang et al. (2009a)		1.60					1.81		
Lu et al., 2011	1.16				1.47				1.68
Lei et al., 2011a	1.18				1.51				
Qin and Xie, 2012	1.14	1.20	1.29	1.36	1.35	1.52	1.55	1.56	1.61
This work	1.14	1.18	1.27	1.33	1.37	1.46	1.52	1.56	1.59
OC									
Streets et al. (2003a)	2.66								
Cao et al. (2006)	3.82								
Ohara et al. (2007)	2.56	2.58	2.60	2.62					
Zhang et al. (2009a)		2.83					3.22		
Lu et al., 2011	2.41				3.13				3.37
Lei et al., 2011a	2.54				3.19				
This work	2.53	2.66	2.91	2.98	3.07	3.13	3.13	3.08	3.08

Table 6. Published estimates of emissions of SO_2, NO_x, CO, NMVOC, BC, and OC from India ($Tgyr^{-1}$).

	2000	2001	2002	2003	2004	2005	2006	2007	2008
SO_2									
Streets et al. (2003a)	5.46								
Garg et al., 2006	4.05					4.59			
Ohara et al. (2007)	6.14	6.47	6.74	7.02					
Zhang et al. (2009a)							5.60		
Lu et al., 2011	5.77				6.45				7.99
EDGAR 4.2	5.76	5.79	6.05	6.18	6.46	6.77	7.36	7.90	8.52
This work	6.57	6.88	7.10	7.29	7.77	8.47	9.21	9.81	10.08
NO_x									
Streets et al. (2003a)	4.05								
Garg et al., 2006	3.64					4.37			
Ohara et al. (2007)	4.73	4.75	4.86	4.97					
Zhang et al. (2009a)							4.86		
EDGAR 4.2	4.55	4.57	4.93	5.02	5.30	5.42	5.74	6.09	6.39
This work	5.83	6.13	6.26	6.76	7.23	7.69	8.26	9.00	9.68
CO									
Streets et al. (2003a)	51.1								
Garg et al., 2006	40.3					41.7			
Ohara et al. (2007)	79.4	80.6	83.0	84.4					
Zhang et al. (2009a)							61.1		
EDGAR 4.2	41.6	41.8	43.3	44.1	43.5	44.1	44.7	45.7	46.3
This work	46.3	47.5	49.0	50.4	51.8	53.9	55.9	58.7	61.8
NMVOC									
Streets et al. (2003a)	8.63								
Ohara et al. (2007)	8.64	8.95	9.34	9.68					
Zhang et al.,2009a							10.77		
EDGAR 4.2	9.57	9.66	10.02	10.27	10.14	10.22	10.37	10.56	10.61
This work	11.62	12.03	12.35	12.89	13.36	14.00	14.69	15.29	15.95
BC									
Streets et al. (2003a)	0.517								
Ohara et al. (2007)	0.795	0.802	0.819	0.832					
Zhang et al. (2009a)							0.344		
Lu et al., 2011	0.680				0.772				0.892
This work	0.462	0.486	0.506	0.536	0.560	0.590	0.616	0.665	0.713
OC									
Streets et al. (2003a)	2.19								
Ohara et al. (2007)	3.27	3.32	3.37	3.42					
Zhang et al. (2009a)							0.89		
Lu et al., 2011	1.71				1.99				2.18
This work	1.76	1.81	1.85	1.91	1.95	2.00	2.05	2.16	2.29

Table 7a. Published estimates of emissions of SO_2, NO_x, CO, NMVOC, BC, and OC in East Asia outside China ($Tg\,yr^{-1}$).

	2000	2001	2002	2003	2004	2005	2006	2007	2008
SO_2									
Streets et al. (2003a)	2.31								
Ohara et al. (2007)	2.56	2.52	2.43	2.39					
Zhang et al. (2009a)							1.79		
EDGAR 4.2	6.84	6.67	6.70	6.26	6.12	6.05	5.85	5.77	5.47
This work	1.82	1.82	1.83	1.81	1.79	1.73	1.75	1.66	1.60
NO_x									
Streets et al. (2003a)	4.33								
Ohara et al. (2007)	4.43	4.44	4.57	4.67					
Zhang et al. (2009a)							4.66		
EDGAR 4.2	5.47	5.44	5.59	5.23	5.22	5.16	5.08	5.04	4.77
This work	4.65	4.67	4.74	4.85	4.85	4.71	4.59	4.35	3.98
CO									
Streets et al. (2003a)	15.0								
Ohara et al. (2007)	15.3	15.5	15.5	17.0					
Zhang et al. (2009a)							11.7		
EDGAR 4.2	21.1	20.7	20.4	20.0	19.7	19.1	18.8	18.5	17.4
This work	14.4	14.2	14.0	13.9	13.9	13.9	13.6	12.5	12.3
NMVOC									
Streets et al. (2003a)	3.73								
Ohara et al. (2007)	3.74	3.78	3.82	3.89					
Zhang et al. (2009a)							3.93		
EDGAR 4.2	7.41	7.33	7.28	7.22	7.18	7.14	7.10	7.08	6.95
This work	3.68	3.58	3.49	3.44	3.46	3.31	3.25	3.26	3.07
BC									
Streets et al. (2003a)	0.103								
Ohara et al. (2007)	0.164	0.160	0.153	0.152					
Zhang et al. (2009a)							0.182		
This work	0.090	0.091	0.090	0.088	0.085	0.081	0.077	0.072	0.066
OC									
Streets et al. (2003a)	0.196								
Ohara et al. (2007)	0.241	0.238	0.206	0.206					
Zhang et al. (2009a)							0.145		
This work	0.054	0.056	0.056	0.056	0.055	0.053	0.051	0.050	0.046

Table 7b. Published estimates of emissions of SO_2, NO_x, CO, NMVOC, BC, and OC in Southeast Asia ($Tg\,yr^{-1}$).

	2000	2001	2002	2003	2004	2005	2006	2007	2008
SO_2									
Streets et al. (2003a)	3.15								
Ohara et al. (2007)	3.65	3.85	3.83	3.81					
Zhang et al. (2009a)							5.48		
EDGAR 4.2	4.11	4.18	4.17	4.11	4.51	4.64	4.74	4.88	4.99
This work	3.71	3.80	3.74	3.72	3.95	4.13	4.13	4.19	4.21
NO_x									
Streets et al. (2003a)	3.06								
Ohara et al. (2007)	3.77	3.97	4.15	4.29					
Zhang et al. (2009a)							5.51		
EDGAR 4.2	3.29	3.41	3.42	3.46	3.71	3.76	3.74	3.88	3.84
This work	3.00	3.13	3.24	3.33	3.62	3.89	4.15	4.70	4.97
CO									
Streets et al. (2003a)	34.0								
Ohara et al. (2007)	54.5	55.0	57.1	59.1					
Zhang et al. (2009a)							44.6		
EDGAR 4.2	32.7	32.9	31.5	31.5	32.7	32.8	32.7	32.8	32.4
This work	36.2	36.8	37.8	39.1	40.7	42.1	43.8	46.5	48.3
NMVOC									
Streets et al. (2003a)	11.1								
Ohara et al. (2007)	11.1	11.3	11.9	12.4					
Zhang et al. (2009a)							14.1		
EDGAR 4.2	9.28	9.35	9.05	9.15	9.39	9.50	9.54	9.67	9.64
This work	10.23	10.54	10.99	11.54	12.13	12.71	13.32	14.28	15.00
BC									
Streets et al. (2003a)	0.320								
Ohara et al. (2007)	0.413	0.419	0.430	0.436					
Zhang et al. (2009a)							0.386		
This work	0.315	0.320	0.327	0.332	0.340	0.345	0.352	0.365	0.371
OC									
Streets et al. (2003a)	1.37								
Ohara et al. (2007)	1.83	1.85	1.90	1.92					
Zhang et al. (2009a)							1.58		
This work	1.30	1.31	1.33	1.34	1.35	1.36	1.38	1.40	1.42

Table 7c. Published estimates of emissions of SO_2, NO_x, CO, NMVOC, BC, and OC in South Asia outside India ($Tg\,yr^{-1}$).

	2000	2001	2002	2003	2004	2005	2006	2007	2008
SO_2									
Streets et al. (2003a)	1.63								
Ohara et al. (2007)	1.34	1.36	1.36	1.28					
Zhang et al. (2009a)							3.16		
EDGAR 4.2	1.03	1.02	0.98	0.69	0.82	0.93	1.17	1.21	1.15
This work	1.19	1.18	1.17	1.03	1.17	1.23	1.38	1.52	1.41
NO_x									
Streets et al. (2003a)	0.71								
Ohara et al. (2007)	0.99	1.02	1.03	1.07					
Zhang et al. (2009a)							0.97		
EDGAR 4.2	0.86	0.88	0.88	0.86	0.95	0.97	1.02	1.13	1.10
This work	1.14	1.03	1.13	1.24	1.32	1.36	1.42	1.50	1.56
CO									
Streets et al. (2003a)	11.2								
Ohara et al. (2007)	19.1	19.5	19.9	20.6					
Zhang et al. (2009a)							13.9		
EDGAR 4.2	10.0	10.0	9.8	10.2	10.3	10.5	10.7	11.1	11.1
This work	12.1	12.4	12.7	13.3	13.7	14.1	14.6	15.2	15.2
NMVOC									
Streets et al. (2003a)	2.04								
Ohara et al. (2007)	2.04	2.10	2.14	2.28					
Zhang et al. (2009a)							2.60		
EDGAR 4.2	2.58	2.59	2.55	2.61	2.65	2.71	2.75	2.83	2.86
This work	2.87	2.94	3.04	3.19	3.29	3.39	3.49	3.61	3.71
BC									
Streets et al. (2003a)	0.142								
Ohara et al. (2007)	0.234	0.239	0.243	0.246					
Zhang et al. (2009a)							0.191		
This work	0.158	0.155	0.161	0.169	0.173	0.178	0.181	0.186	0.192
OC									
Streets et al. (2003a)	0.626								
Ohara et al. (2007)	0.967	0.986	1.007	1.025					
Zhang et al. (2009a)							0.707		
This work	0.565	0.571	0.587	0.603	0.618	0.632	0.647	0.660	0.674

Table 8. Published estimates of NH_3, CH_4, and N_2O emissions from China, India, and the rest of the Asian countries.

	2000	2001	2002	2003	2004	2005	2006	2007	2008
NH_3									
China									
Streets et al. (2003a)	13.3								
Ohara et al. (2007)	12.6	12.6	12.6	12.6					
EDGAR 4.2	8.7	8.8	9.6	9.4	10.1	10.2	10.6	10.9	11.2
This work	12.5	12.6	13.6	13.4	13.1	13.6	14.3	14.6	14.8
India									
Streets et al. (2003a)	7.23								
Ohara et al. (2007)	8.48	8.52	8.56	8.60					
EDGAR 4.2	3.86	3.96	3.79	3.96	4.08	4.08	4.15	4.19	4.24
This work	8.08	8.21	8.04	8.23	8.44	8.71	9.02	9.25	9.42
Other Asia									
Streets et al. (2003a)	6.03								
Ohara et al. (2007)	7.02	7.08	7.14	7.20					
EDGAR 4.2	4.06	4.02	4.24	4.31	4.38	4.46	4.54	4.63	4.70
This work	7.16	7.18	7.42	7.62	7.92	7.89	8.04	8.28	8.34
CH_4									
China									
Streets et al. (2003a)	37.8								
Ohara et al. (2007)	33.1	33.1	33.1	33.2					
EDGAR 4.2	49.8	50.2	51.8	54.7	59.7	63.3	66.4	68.9	73.3
This work	48.8	49.9	53.4	58.0	62.4	66.9	69.2	70.9	76.0
India									
Streets et al. (2003a)	32.4								
Ohara et al. (2007)	25.7	25.8	25.8	25.9					
EDGAR 4.2	26.4	26.7	26.7	27.1	27.2	27.6	27.8	28.2	28.6
This work	27.3	27.3	27.0	27.3	27.3	27.5	28.8	28.4	29.4
Other Asia									
Streets et al. (2003a)	33.5								
Ohara et al. (2007)	31.1	31.1	31.2	31.2					
EDGAR 4.2	39.4	39.8	40.6	41.5	42.5	43.8	45.1	46.2	47.1
This work	38.0	38.1	38.7	39.4	40.3	41.6	42.8	43.5	44.4
N_2O									
China									
EDGAR 4.2	1.38	1.40	1.50	1.51	1.59	1.63	1.69	1.73	1.76
This work	2.16	2.18	2.31	2.30	2.31	2.41	2.55	2.59	2.66
India									
EDGAR 4.2	0.68	0.69	0.68	0.70	0.72	0.72	0.73	0.75	0.76
This work	1.51	1.53	1.48	1.52	1.54	1.59	1.65	1.70	1.72
Other Asia									
EDGAR 4.2	0.82	0.85	0.91	0.83	0.88	0.91	1.03	0.87	0.86
This work	1.22	1.22	1.24	1.26	1.32	1.31	1.35	1.36	1.37

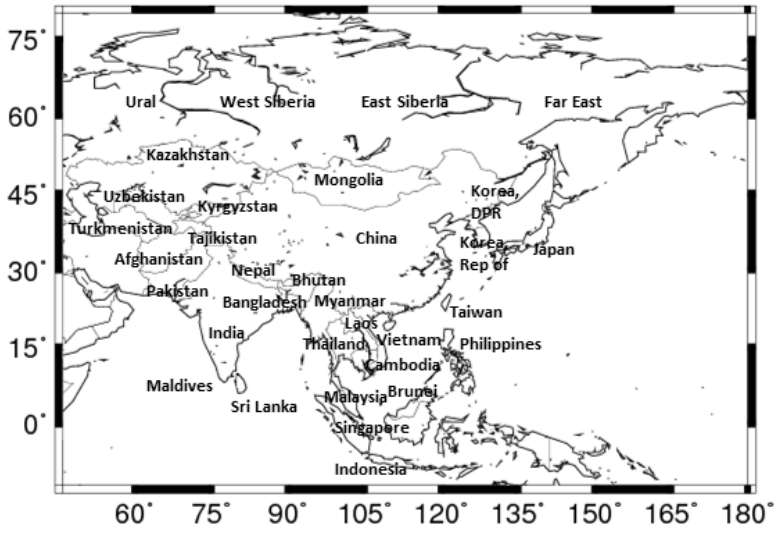

Fig. 1. Inventory domain of REAS 2.1 showing names of all included countries. Western boundary of the Ural region is 60° E.

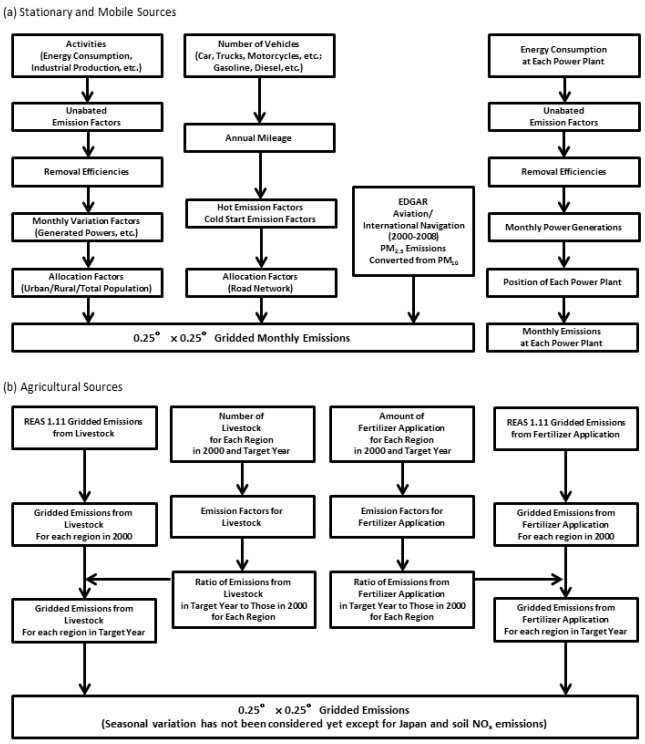

Fig. 2. Schematic flow diagrams showing **(a)** estimation of emissions from stationary and mobile sources and **(b)** extrapolation of gridded agricultural emissions based on REAS 1.1 data for the year 2000.

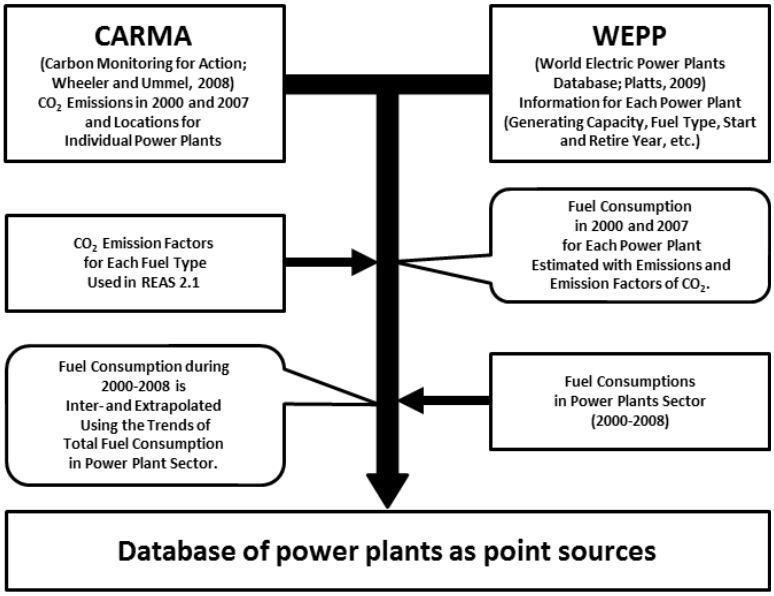

Fig. 3. Schematic flow diagram for developing a database of basic data for power plants as point sources.

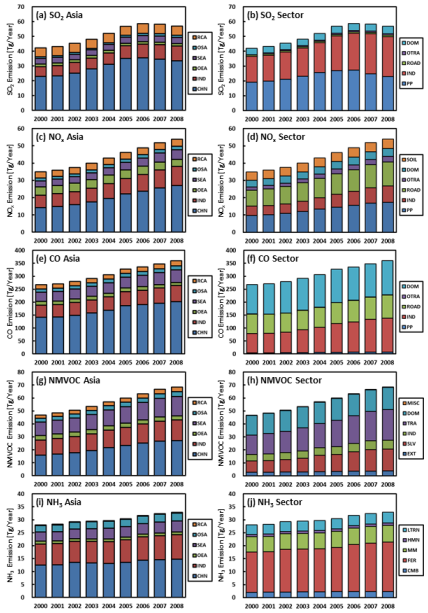

Fig. 4. Annual emissions of SO_2, NO_x, CO, NMVOC, and NH_3 in Asia from 2000 to 2008 for each region (left panels) and sector (right panels). Regions: CHN = China, IND = India, OEA = East Asia outside China, SEA = Southeast Asia, OSA = South Asia outside India, and RCA = Asian Russia and Central Asia. Sectors: PP = Power plants, IND = Industry, ROAD = Road transport, OTRA = Other transport, DOM = Domestic, SOIL = Soil, EXT = Extraction processes, SLV = Solvent and paint use, WASTE = Waste treatment, CMB = Combustion, FER = Fertilizer application, MM = Manure management, and HMN = Human perspiration and respiration, and LTRN = Latrines.

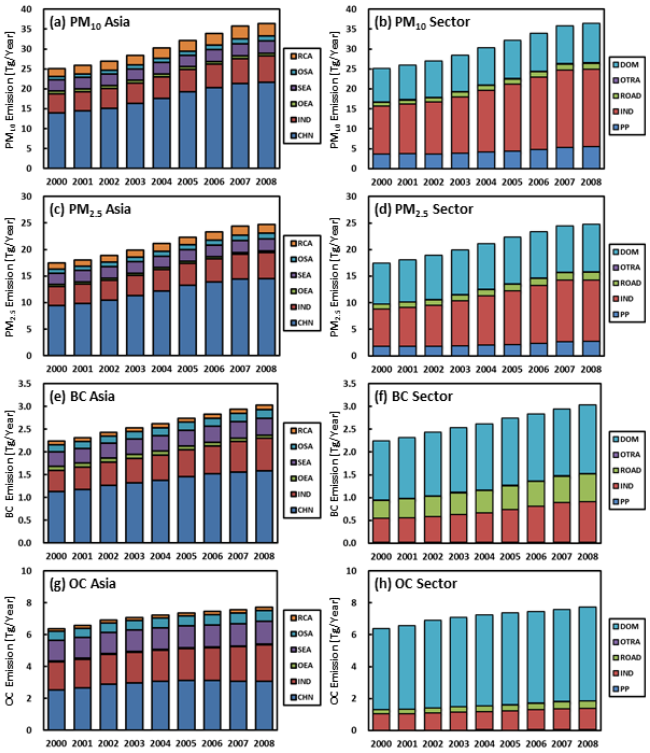

Fig. 5. Annual emissions of aerosols (PM_{10}, $PM_{2.5}$, BC, and OC) in Asia from 2000 to 2008 for each region (left panels) and sector (right panels). Abbreviations are the same as in Fig. 4.

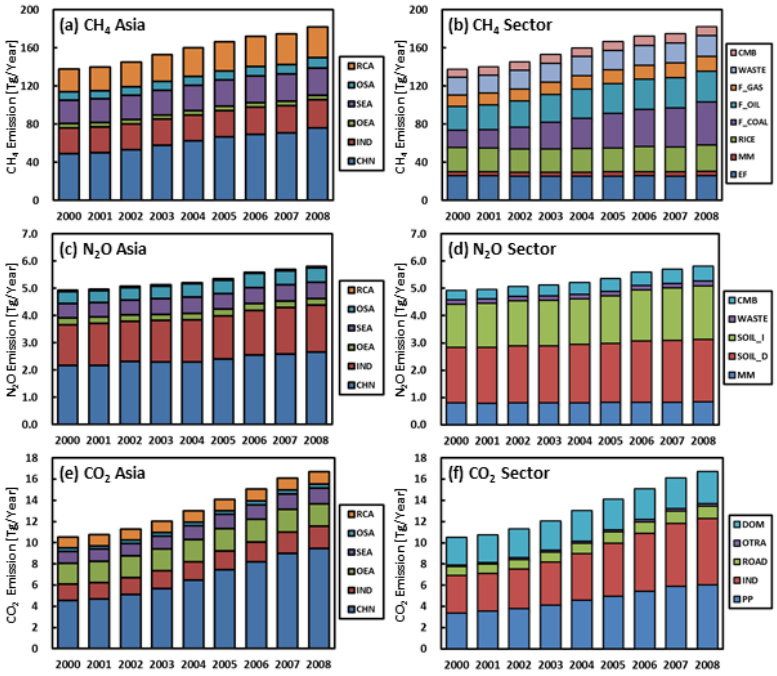

Fig. 6. Annual emissions of greenhouse gases (CH_4, N_2O, and CO_2) in Asia from 2000 to 2008 for each region (left panels) and sector (right panels). Abbreviations are the same as Fig. 4, plus EF = Enteric fermentation, RICE = Rice cultivation, F_COAL/F_OIL/F_GAS = Fugitive emissions related to coal/oil/gas, SOIL_D = Direct soil emissions, and SOIL_I = Indirect soil emissions.

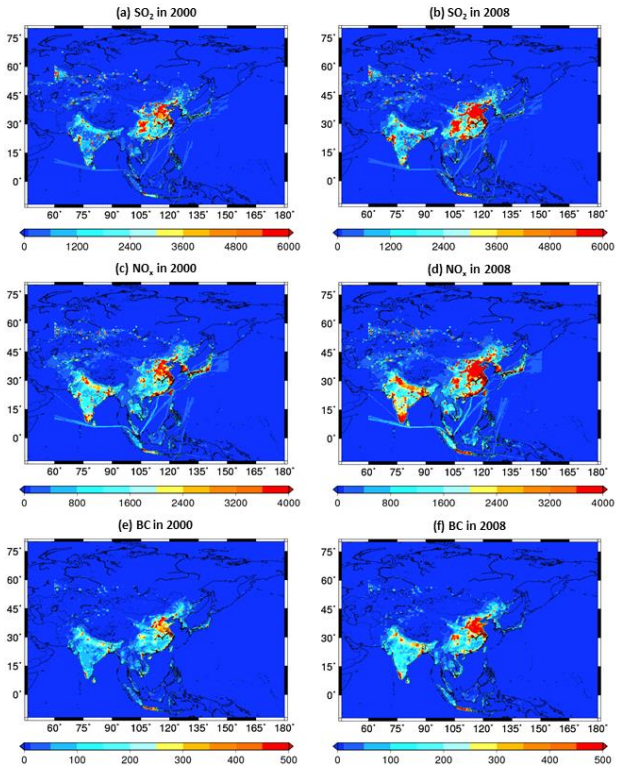

Fig. 7a. Spatial distributions of annual emissions ($Mg\,yr^{-1}$ per grid cell) of SO_2, NO_x, BC, NMVOC, NH_3, and CH_4 in 2000 and 2008.

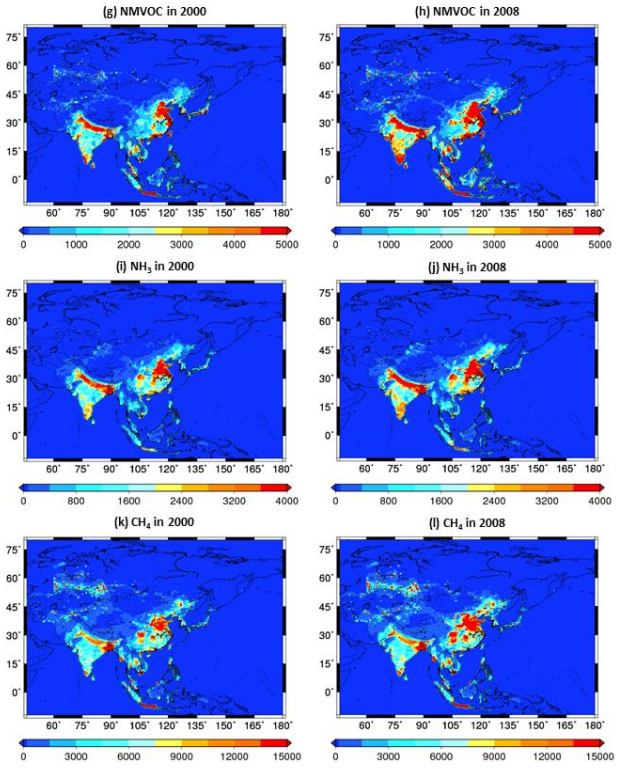

Fig. 7b. Caption on previous page.

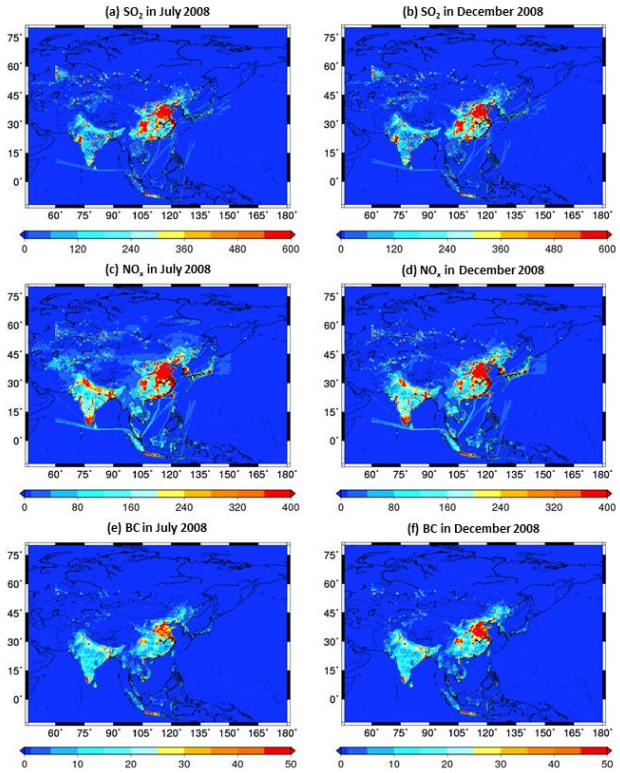

Fig. 8. Spatial distributions of monthly emissions (Mg month^{-1} per grid cell) of SO_2, NO_x, and BC in July and December 2008.

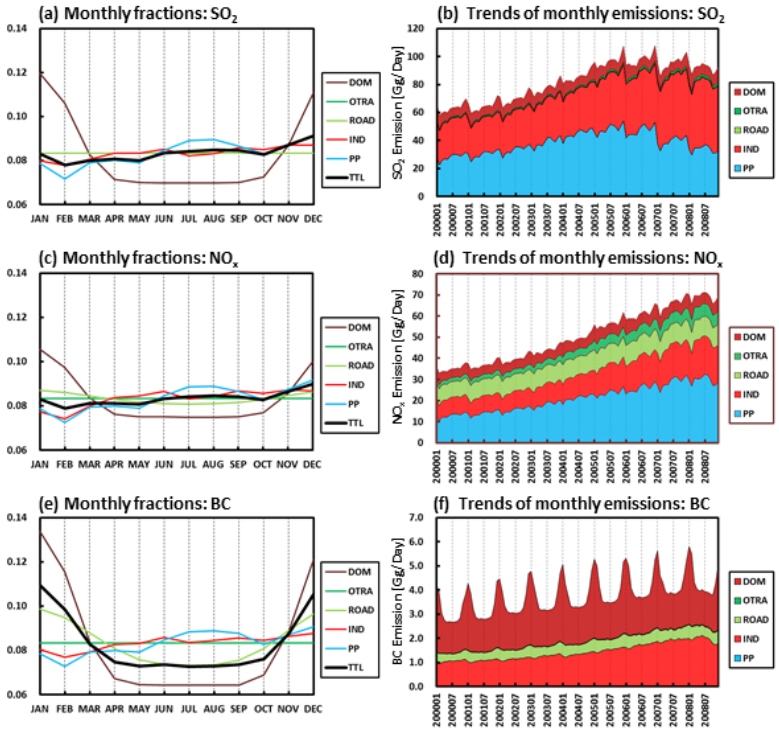

Fig. 9. Fractions (left panels) and trends (right panels) of monthly anthropogenic emissions of SO_2, NO_x, (other than soil) and BC in China (Gg day^{-1}). Monthly fractions are average of all values between 2000 and 2008. Abbreviations are the same as Fig. 4; plus TTL = Total.

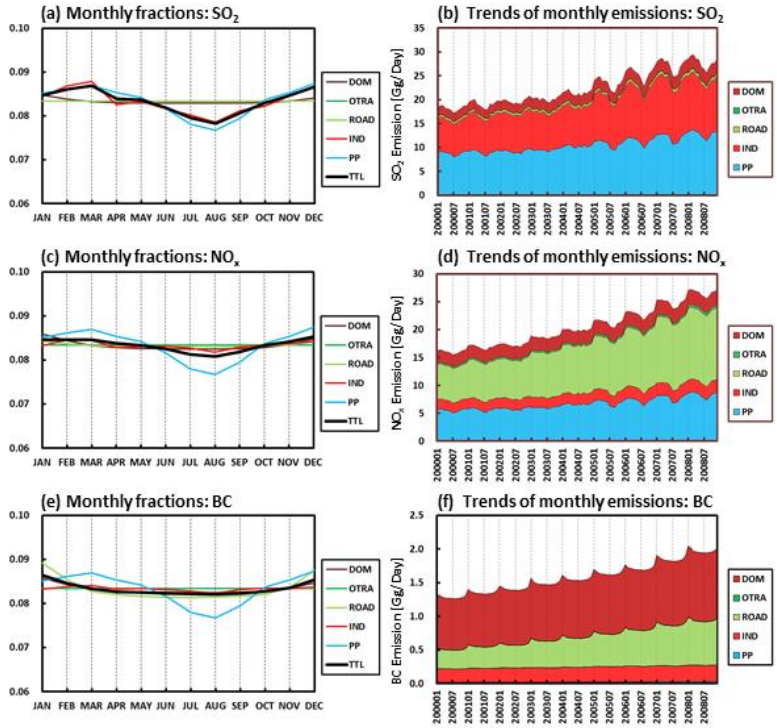

Fig. 10. Fractions (left panels) and trends (right panels) of monthly anthropogenic emissions of SO_2, NO_x, (other than soil) and BC in India ($Gg\,day^{-1}$). Monthly fractions are average of all values between 2000 and 2008. Abbreviations are the same as Fig. 9.

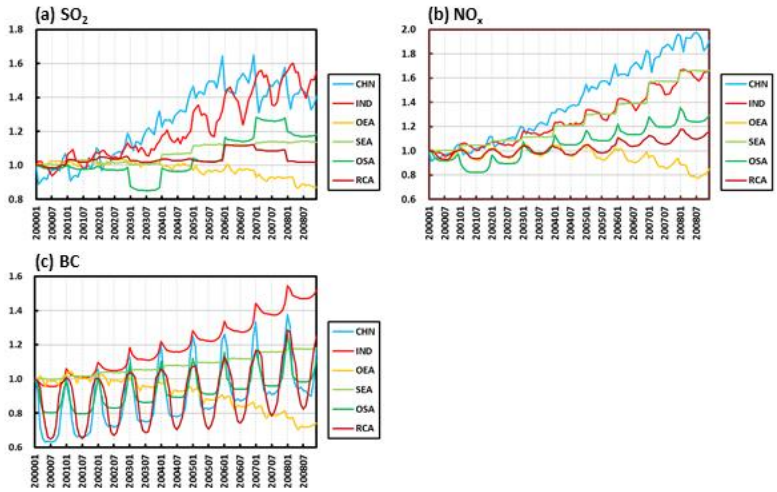

Fig. 11. Trends of monthly anthropogenic emissions of SO_2, NO_x (other than soil), and BC in each region from 2000 to 2008. Values are normalized at emissions in January 2000. Abbreviations are the same as Fig. 4.

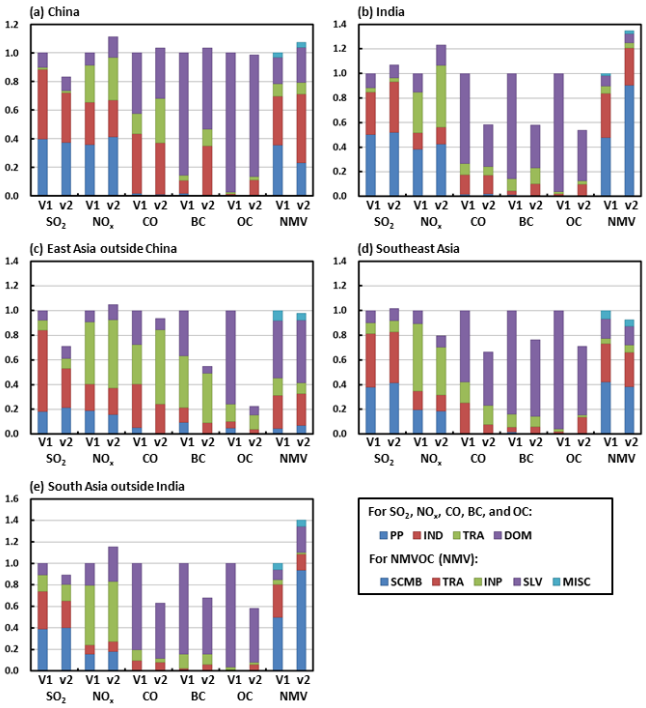

Fig. 12. Comparison of SO_2, NO_x (other than soil), CO, BC, OC, and NMVOC emissions in 2000 in REAS version 1 (v1) and 2 (v2) for **(a)** China, **(b)** India, **(c)** East Asia outside China, **(d)** Southeast Asia and **(e)** South Asia outside India for various sectors (PP = Power plants, IND = Industry, TRA = Transport, DOM = Domestic, SCMB = Stationary combustion, INP = Industrial processes, SLV = Solvent and paint use, MISC = Miscellaneous). Values are normalized to those in REAS version 1.