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MIX: a mosaic Asian anthropogenic emission inventory for the MICS-Asia and the HTAP projects

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34813

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poral profiles, etc.), introducing large uncertainties in emission estimates for a specific region (He et al., 2007; Kurokawa et al., 2009).

The other is the “mosaic” approach that harmonizes various emission inventories of different regions into one emission data product at large scale, by normalization of source categories, species, and spatial and temporal resolution from different inventories and providing emission data with uniform format. Available emission inventories always differ in geographic region, time period, source classification, species, and spatial and temporal resolution, introducing complexities in inter-comparisons of emissions and model results with different emission inputs. By involving the state-of-the-art local emission inventories developed with local knowledge and harmonizing them to uniform format, this approach can provide a reference on magnitude and spatial distribution of emissions for different regions, while there is always trade-off in spatial/temporal coverage and resolution due to inconsistencies among involved inventories.

Recent studies (e.g., Zhang et al., 2009; Kurokawa et al., 2013) tend to use the mosaic approach to supplement the Asian emission inventory developments. To support NASA’s INTEX-B (the Intercontinental Chemical Transport Experiment-Phase B) mission (van Donkelaar et al., 2008; Adhikary et al., 2010), Zhang et al. (2009) developed a new emission inventory for Asia for the year 2006 as an update and improvement of the TRACE-P inventory (Streets et al., 2003). Compared to the TRACE-P inventory, the INTEX-B inventory improved emission estimates for China by introducing a technology-based methodology, and incorporated several local inventories including BC and OC emissions for India from Reddy et al. (2002a, b), a Japan emission inventory from Kannari et al. (2007), and official emission inventories for the Republic of Korea and Taiwan. All of these emission data were harmonized and processed to $0.5^\circ \times 0.5^\circ$ resolution. In the updated version 2.1 of the REAS inventory (Kurokawa et al., 2013), a few regional inventories developed with local knowledge are also incorporated to improve the accuracy (see Sect. 2.2.1 for details).

In order to support the MICS-Asia III and other regional modeling activities with the best available anthropogenic emission dataset over Asia, we develop a new Asian an-

34817

thropogenic emission inventory, named MIX, by harmonizing different local emission inventories with the mosaic approach. The mosaic inventory developed in this work will provide (1) a more complete and state-of-the-art understanding of anthropogenic emissions over Asia with better estimates from local inventories, (2) a reference dataset with moderate accuracy and resolution that can support both scientific research and mitigation policy-making, and (3) broader application of the best available local inventories in modeling studies by processing them to model-ready format and including them in a publicly available emission dataset.

The target year of the MIX inventory is 2010, in accordance with base year simulations in MICS-Asia III. It should be noted that MIX is not comparable to INTEX-B and TRACE-P to derive an emission trend due to differences in methodology and underlying data. In this paper, we also provided Asian emissions for 2006 and 2008 using the same methodology, partly resolving the problems of trend analysis in mosaic inventories. The MIX emission data for the years 2008 and 2010 are then incorporated into the HTAP v2.2 global emission inventory (Janssens-Maenhout et al., 2015) to support the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), providing a consistent emission input for global and regional modelling activities.

Figure 1 presents the definition of the MIX domain and emission datasets used for each country and region. The domain of MIX covers 30 countries and regions, stretching from Kazakhstan in the west to Far East Russia in the east, and from Indonesia in the south to Siberia in the north. Emissions are aggregated into five sectors: power, industry, residential, transportation, and agriculture. Ten chemical species are included in the MIX inventory, including both gaseous species and aerosol species: SO_2 , NO_x , CO, NMVOC (non-methane volatile organic compounds), NH_3 (ammonia), PM_{10} (particulate matter with diameter less than or equal to $10\ \mu\text{m}$), $\text{PM}_{2.5}$ (particulate matter with diameter less than or equal to $2.5\ \mu\text{m}$), BC (black carbon), OC (organic carbon) and CO_2 . Only emissions from anthropogenic sources are included in MIX. NMVOC emissions are speciated into model-ready inputs for two chemical mechanisms: CB05 (the Carbon Bond mechanism, Yarwood et al., 2005) and SAPRC-99 (the

34818

REAS2 also incorporated a few regional inventories developed by local agencies with detailed activity data and emission factors, including the JEI-DB inventory (Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base, JPEC, 2012a–c) for all anthropogenic sources in Japan excluding shipping, OPRF (Ocean Policy Research Foundation, OPRF, 2012) for shipping emissions in Japan, CAPSS emission inventory for Korea (Lee et al., 2011), and official emission data from the Environmental Protection Administration of Taiwan for Taiwan (Kurokawa et al., 2013). All these regional datasets were then harmonized to the same spatial and temporal resolution in REAS2. In this work, we processed the CAPSS emission data separately as an individual data source, which is presented in Sect. 2.2.5, and adopted Japan and Taiwan emissions directly from the REAS2 product.

The REAS2 inventory is provided with monthly gridded emission data by sectors at $0.25^\circ \times 0.25^\circ$ resolution. We aggregated the 11 REAS2 sectors to five sectors provided in the MIX inventory. Monthly variations are developed for power plants, industry, residential sources and cold-start emissions from vehicles by various monthly profiles (Kurokawa et al., 2013). In REAS2, power plants with annual CO_2 emissions larger than 1 Tg were provided as point sources with coordinates of locations, while emissions for other sectors were processed as area sources and gridded at $0.25^\circ \times 0.25^\circ$ resolution using maps of rural, urban and total populations and road networks.

2.2.2 MEIC

We use anthropogenic emission data generated from the MEIC (Multi-resolution Emission Inventory for China) model to override emissions in mainland China. MEIC is a bottom-up emission inventory framework developed and maintained by Tsinghua University, which uses a technology-based methodology to calculate air pollutant and CO_2 emissions for more than 700 anthropogenic emitting sources for China from 1990 to the present. With the detailed source classification, the MEIC model can represent emission characteristics from different sectors, fuels, products, combustion/process technologies, and emission control technologies. The MEIC model improved the bottom-up

34821

emission inventories developed by the same group (Streets et al., 2006; Zhang et al., 2007a, b, 2009; Lei et al., 2011) and integrated them into a uniform framework. The major improvements include a unit-based power plant emission database (Zhao et al., 2008; Wang et al., 2012; Liu et al., 2015), a high-resolution vehicle emission modeling approach (Zheng et al., 2014), an explicit NMVOC speciation assignment methodology (Li et al., 2014), and a unified, on-line framework for emission calculation, data processing, and data downloading (available at <http://www.meicmodel.org>).

Power plant emissions in MEIC were derived from the China coal-fired Power plant Emissions Database (CPED), in which emissions were estimated for each generation unit based on the unit-specific parameters including fuel consumption rates, fuel quality, combustion technology, and emission control technology. With detailed information of over 7600 generation units in China, CPED improved the spatial and temporal resolution of the power plant emission inventory compared to previous studies (Liu et al., 2015). For the on-road transportation sector, MEIC used the new approach developed by Zheng et al. (2014), which estimated vehicle emissions with high spatial resolution by using vehicle population and emission factors at county level. County-level emissions were further allocated to high-resolution grids based on a digital road map and weighting factors of vehicle kilometers traveled (VKT) by vehicle and road type.

MEIC provides lumped speciated NMVOC emissions for different chemical mechanisms, e.g., SAPRC-99, SAPRC-07, CBIV, CB05, and RADM2. Following the speciation assignment approach developed by Li et al. (2014), emissions of individual NMVOC species were calculated for each source category by splitting the total NMVOC emissions with corresponding source profiles. Emissions were then assigned to various mechanisms using species mapping tables.

MEIC delivers monthly emissions at various spatial resolutions through an open-access, online framework (<http://www.meicmodel.org>). Monthly variations and gridded emissions were generated by sector using different temporal profiles and spatial profiles. Users can define the metadata (species, domain range, time period, sectors, spatial resolution, and chemical mechanisms), calculate gridded emissions, and download

34822

speciated NMVOC emissions were then calculated by linking emissions to speciation profiles with cross-referencing.

3 Results

3.1 Asian anthropogenic emissions in 2010

5 Based on the mosaic approach and candidate inventories described in Sect. 2, gridded anthropogenic emissions for ten species were generated over Asia and called the “MIX” emission inventory. In the MIX inventory, Asian anthropogenic emissions in 2010 are estimated as follows: 51.3 Tg SO₂, 52.1 Tg NO_x, 336.6 Tg CO, 67.0 Tg NMVOC, 28.8 Tg NH₃, 31.7 Tg PM₁₀, 22.7 Tg PM_{2.5}, 3.5 Tg BC, 8.3 Tg OC and 17.3 Pg CO₂. Figure 4 presents the emission distributions among sectors over Asia in 2010. Among the different sectors, the industrial sector has the largest contribution to SO₂ (50 % of total), NMVOC (38 %), PM₁₀ (48 %), and CO₂ (40 %) emissions. Power plants have significant contributions for SO₂ (38 % of total), NO_x (29 %), and CO₂ (34 %) emissions.

15 Asian emissions in 2010 for ten species are listed in Table 3 by country and the shares of 2010 emissions by each sub-region are presented in Fig. 5. China is the largest contributor for most species except NH₃, with more than 50 % contribution for SO₂, NO_x, CO, PM₁₀, PM_{2.5} and CO₂ emissions. Following China, India is the largest contributor for NH₃ emissions (34 % of total) and the second largest contributor for all other species. As shown in Fig. 5, Southeast Asia and Other South Asia contribute more than 20 % to NMVOC, NH₃, OC and CO emissions, and around 10 % for other species, representing, in particular, a high contribution from biofuel emissions. Contributions from other Asian regions are less than 10 % for all species.

20 Table 4 presents Asian 2010 emissions by region and by sector. China’s anthropogenic emissions in 2010 are estimated as follows: 28.7 Tg SO₂, 29.1 Tg NO_x, 170.9 Tg CO, 23.6 Tg NMVOC, 9.8 Tg NH₃, 16.6 Tg PM₁₀, 12.2 Tg PM_{2.5}, 1.8 Tg BC, 3.4 Tg OC and 10.1 Pg CO₂. Overall, industry is the largest emitter of China’s anthro-

34827

pogenic emissions, contributing 49 % of the total CO₂ emissions and 59, 39, 61 %, and 50 % of SO₂, NO_x, NMVOC, and PM_{2.5} emissions, respectively. The dominance of the industrial sector on China’s anthropogenic emissions reflects the fact that China has developed a huge industrial capacity, which has led to very high levels of energy use and emissions. For example, China produced 44 and 70 % of global iron and cement, respectively, in 2010 (World Steel Association, 2011; United Nations, 2011). As a result, industrial SO₂ emissions in China in 2010 surpassed SO₂ emissions from the US and Europe combined. Power plants contributed 32 % of the total CO₂ emissions and 28, 33 %, and 7 % of SO₂, NO_x, and PM_{2.5} emissions, respectively. Emission ratios of SO₂/CO₂ and PM_{2.5}/CO₂ are lower in power plants than in the industrial sector, reflecting the better emission control facilities operated in power plants, such as flue-gas desulfurization devices (FGD). The residential sector dominates emissions for pollutants from incomplete combustion, given that large amounts of solid fuels (coal and biomass) were burned in small stoves in China’s homes. The residential sector shared 13 % of China’s total CO₂ emissions in 2010, but contributed to 45 % of CO, 27 % of NMVOC, 51 % of BC, and 81 % of OC emissions, respectively. The transportation sector accounted for 25, 12, 11 %, and 16 % of NO_x, CO, NMVOC, and BC emissions, respectively. The contribution of the transportation sector to China’s CO and NMVOC emissions has substantially decreased during recent years, which will be further discussed in the next section.

20 In the MIX inventory, Indian emissions in 2010 are estimated as follows: 9.3 Tg SO₂, 9.6 Tg NO_x, 67.4 Tg CO, 16.9 Tg NMVOC, 9.9 Tg NH₃, 7.1 Tg PM₁₀, 5.2 Tg PM_{2.5}, 1.0 Tg BC, 2.5 Tg OC and 2.3 Pg CO₂. In India, the industrial sector has much lower contribution to emissions compared to China, while higher emission contributions from the residential sector are estimated. The differences of the emission patterns between China and India can be attributed to differences in the stage of economic development and the composition of the energy structure. In India, the residential sector is the second largest contributor for CO₂ emissions and the largest contributor for CO, NMVOC, PM_{2.5}, BC, and OC emissions, in which more than 70 % of those emissions

34828

are contributed by biofuel combustion. With the rapid growth of coal-fired generation units, SO₂ emissions from Indian power plants are estimated to be 5.5 Tg in 2010, contributing 59 % of the total SO₂ emissions. The SO₂/CO₂ emission ratio in Indian power plants is significantly higher than that of China, representing the low penetration rates of FGD in Indian power plants (Lu et al., 2011). The transportation sector contributes 55 % of NO_x and 36 % of NMVOC emissions in India. These large shares are caused by the high emission factors used in REAS2, in which relatively poor emission control measures are in place (Kurokawa et al., 2013).

3.2 Changes of Asian emissions from 2006 to 2010

In this work, we also developed Asian emissions for 2006 and 2008 following the same approach of MIX, to illustrate the changes in Asian emissions from 2006 to 2010. Table 5 presents Asian emissions in 2006 by country. For the whole of Asia, emission growth rates from 2006 to 2010 are estimated as follows: -8.0 % for SO₂, +19 % for NO_x, +4 % for CO, +15 % for NMVOC, +2 % for NH₃, -3 % for PM₁₀, -2 % for PM_{2.5}, +6 % for BC, +2 % for OC and +20 % for CO₂. Growth in CO₂ emissions represents the continuously increasing energy use across Asia during 2006–2010, while different trends among species represents differences in the emission control level among sectors and regions. Compared to the increasing emission trends of all species during 2001–2006 (Zhang et al., 2009), the relatively flat or even decreasing emission trends in many species indicates the effectiveness of emission control measures in recent years (Gu et al., 2013; Lin et al., 2010; Wang et al., 2013).

Changes in Asian emissions are dominated by changes in China and India. Figure 6a demonstrates the changes in SO₂ emissions among Asian regions from 2006 to 2010. Wide installation of flue-gas desulfurization (FGD) in China's coal-fired power plants is the main driving factor of SO₂ emission changes over Asia. SO₂ emissions in China's power plants decreased from 17.2 Tg in 2006 to 8.2 Tg in 2010, contributing to most of the total SO₂ emission reduction over Asia. In contrast, SO₂ emissions in India increased by 27 % during 2006–2010, owing to dramatic construction of new

34829

power plants and lack of emission control facilities (Garg et al., 2001, 2006). As a consequence, the Indian share of the total Asian SO₂ emissions increased from 13 % in 2006 to 18 % in 2010. NO_x and NMVOC emissions increased in all Asian regions except Other East Asia, indicating less effective emission control measures for those two species over Asia. Increases of NO_x and NMVOC emissions are mainly driven by the industry and transportation sectors. Emission changes of other species are relatively small (i.e., within 6 %) during 2006–2010. For CO, PM₁₀, and PM_{2.5}, emission reductions in China were partly offset by increases of emissions in the South and Southeast Asian regions. CO emissions in China decreased by 5 % during 2006–2010 (see Fig. 6b), mainly due to improved combustion efficiency, recycling of industrial coal gases, and strengthened vehicle emission standards. The implementation of new vehicle emission standards and retirement of old vehicles has reduced China's transportation CO and NMVOC emissions by 20 and 30 % respectively during 2006–2010. The downward trend of CO emissions over China has been confirmed by both in-situ and satellite observations (Wang et al., 2010; Worden et al., 2010; Yumimoto et al., 2014; Yin et al., 2015). While in India, Other South Asia and Southeast Asia, CO emissions increased by 21, 11, and 16 %, respectively, between 2006 and 2010.

3.3 Speciated NMVOC emissions

Figure 7 presents 2010 Asian NMVOC emissions of different chemical groups by region and by sector. Similar to Asian emissions estimated in previous work (Klimont et al., 2002; Li et al., 2014), alkanes and alkenes are the largest contributors to total Asian NMVOC emissions in 2010 (27 and 26 % of the total respectively), followed by aromatics (20 %), OVOCs (oxygenated volatile organic compounds, 17 %), and alkynes (7 %). Regionally, shares of alkanes and aromatics are higher in East Asia, Central Asia, and Russia Asia than other regions, due to large contributions from the industrial sector. Shares of alkynes in Central Asia and Russia Asia are significantly lower than other regions due to a low contribution from biofuel emissions. Sectoral contribution of emissions vary significantly by different chemical groups. Over Asia, the industrial sector is

34830

the major source of emissions of alkanes and aromatics, while the residential sector has a high contribution of OVOCs, alkynes, and alkenes. The sectoral contribution to different chemical groups also varies with region. For example, the residential sector dominates emissions for all species in the Other South Asia region, as a consequence of the low economic development in that region.

Among different regions, China, India and Southeast Asia are the largest contributors to NMVOC emissions in Asia, with contributions varying by chemical groups. China contributes more than 40 % of alkanes, alkynes, and aromatics in Asia, compared to 35 % contribution of the total Asian NMVOC emissions. India contributes high to emissions of alkenes, alkynes and OVOCs, constituting about 30 % of Asian emissions. Southeast Asia shares around 20 % of the emissions of alkanes, alkenes, aromatics and OVOCs.

3.4 Seasonality

Monthly emissions are provided in MIX. As documented in Section 2.2, we used monthly emissions from each component inventory where available, and assumed no monthly variation in emissions when the component inventory only provided annual emissions. Monthly emissions by sector and by Asian region are provided in Tables S1 and S2 in the Supplement. Monthly profiles in emissions are highly sector-dependent, given that monthly activity rates vary among different sectors. Figure 8 illustrates the monthly variations of Asian SO₂, CO, PM_{2.5}, and CO₂ emissions by sector for the year 2010. Different species generally show similar monthly emission patterns within the same sector, indicating that monthly emission profiles of each sector are dominated by monthly variations in activity rates. For example, industrial emissions are higher in the second half of the year induced by larger industrial productions to meet the annual total production target. The most significant monthly variation with a winter peak was found in the residential sector, reflecting the higher energy demand for residential heating in winter. Residential SO₂ emissions in winter are even higher than other species, because SO₂ emissions from China dominate residential emissions in Asia (70 % of total), of which

34831

coal consumption in winter is higher than other regions for heating. Monthly profiles of CO emissions are different from other species for the transportation sector. This is because the CO emission factor in winter is higher than in other seasons due to additional emissions from the cold-start process (Kurokawa et al., 2013; Zheng et al., 2014).

Figure 9 presents monthly variations of SO₂, CO, PM_{2.5}, and CO₂ emissions by Asian region. Compared to other species, CO emissions are much higher in winter in high-latitude regions due to residential heating and additional vehicle emissions from cold starts. Winter PM_{2.5} emissions in China are higher than other regions, representing large emissions from solid fuel use in residential homes.

3.5 Gridded emissions

In the MIX inventory, gridded emissions for ten gaseous and aerosol species were developed at 0.25° × 0.25° resolution. Emission maps of all species in 2010 are shown in Fig. 10. Compared to the previous gridded Asian emission inventories, we believe the spatial patterns are improved because several local high-resolution emission datasets are incorporated, such as CPED for China and JEI-DB and OPRF for Japan. However, for sectors in which emissions are dominated by spatially scattered sources (e.g., residential combustion, solvent use), the spatial distributions in emissions are still uncertain.

MIX emission inventory can be accessed publicly from the website of <http://www.meicmodel.org/dataset-mix>. Both 2008 and 2010 emissions of ten species with monthly variation at a spatial resolution of 0.25° × 0.25° are available from the website, including SO₂, NO_x, CO, NH₃, NMVOC, PM₁₀, PM_{2.5}, BC, OC, and CO₂. Speciated NMVOC Emissions for CB05 and SAPRC-99 chemical mechanisms are provided at the same spatial and temporal resolution. The MIX inventory has been re-gridded to 0.1° × 0.1° resolution using area-weighting approach and then incorporated to the HTAP v2 gridded emission inventory (Janssens-Maenhout et al., 2015). The HTAP v2 emission inventory can be downloaded from the EDGAR website (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123).

34832

4 Comparison with other inventories

4.1 MIX, REAS2 and EDGAR v4.2 over Asia

In this section, we compare the MIX inventory with REAS2 and EDGAR v4.2 (EC-JRC/PBL, 2011, hereafter EDGAR), the two widely used inventories, to highlight the new findings from the mosaic inventory and identify the potential sources of uncertainties. We choose the year of 2008 to conduct the comparison because emissions after 2008 are not available in either REAS2 or EDGAR. Asian anthropogenic emissions of MIX, REAS2 and EDGAR in 2008 are tabulated in Table 6. Over Asia, MIX and REAS2 differ within 10 % for most species, except for NH_3 (18 % higher in REAS), PM_{10} (13 % higher), and BC (13 % lower). It is not surprising that the total Asian emission budgets in MIX and REAS2 are similar, given that MIX used emissions estimates in REAS2 for Asian regions except China and India. On the other hand, REAS2 has incorporated several different emission inventories for China (Kurokawa et al., 2013). The differences between REAS and MIX over China and India will be discussed in the following sections.

Larger discrepancies are observed between MIX and EDGAR. Compared to MIX, 2008 Asian emissions in EDGAR are 29 % higher for SO_2 , but 20, 33, 11, 27 % lower for NO_x , CO, NMVOC, and NH_3 , respectively. PM_{10} and CO_2 emissions agree well between the two inventories. Figure 11 details the differences by region and by sector. Regionally, the differences can be largely attributed to disagreements in emission estimates for China and India, as presented in Table 6. Discrepancies are large at the sector level. EDGAR's estimates for SO_2 emissions from power plants are 60 % higher than estimates in MIX, most likely due to underestimation of FGD penetration (Kurokawa et al., 2013). Large discrepancies for the residential and transportation sectors are found for NO_x , CO, and NMVOC estimates in the two inventories. For instance, EDGAR estimates lower NO_x emissions of transportation sector by 27 and by 48 % for the residential sector compared to MIX. Similarly, residential CO emissions in EDGAR are about a factor of 1.5 lower than in MIX, leading to 35 % lower estimates of CO

34833

emissions in EDGAR compared to MIX. Underestimates of CO emissions in EDGAR inventory have been confirmed by top-down constraints (Pétron et al., 2004; Fortems-Cheiney et al., 2011). As the statistical differences of energy use are usually within 30 % at sector level (Guan et al., 2012), the huge discrepancy by sector could only be attributed to differences in emission factors. Although a point-by-point comparison of emission factors between EDGAR and MIX is not feasible, we can still speculate that EDGAR may overestimate the combustion efficiency and emission control measures in Asia by using an emission factor database from developed countries. NH_3 emissions in EDGAR are 26 % lower than in MIX, with a large difference in residential emissions. The differences are mainly from high emission estimates of wastewater treatment sources in REAS2 which were incorporated into MIX for Asian regions except China. MIX estimated 3.4 Tg NH_3 emissions from wastewater treatment in Asia in 2008, which are more than two orders of magnitude higher than EDGAR estimates. Differences in PM_{10} emissions at the sector level are also huge; similar estimates of PM_{10} emissions in the two datasets are rather a coincidence than real agreements.

4.2 China

4.2.1 Power plants

Both MIX and REAS2 processed power plants emissions as point sources. As presented in Sect. 2.2, MIX used a high-resolution emission database for China (CPED, Liu et al., 2015) to derive emissions and locations of China's power plant emissions at unit level. In REAS2, emissions of individual power plants are estimated by combining information from two global databases, CARMA and WEPP. MIX and REAS2 showed good agreements on power plant emissions in China for 2008 for major species (differences within 30 % for NO_x and 10 % for SO_2 and CO_2 , respectively), implying similar values for energy consumption and emission factors used in the two inventories. Liu et al. (2015) found that CARMA has omitted information of small plants and overestimated emissions from large plants by wrongly allocating fuel consumptions of small

34834

plants to large ones. REAS2 included 380 power plants for China, 84% lower than 2411 plants in MIX. In REAS2, large plants (defined as plants with CO₂ emissions higher than 5 Tg) contributed 77% of power plant CO₂ emissions in China, compared to a 47% contribution of large plants in MIX.

5 Figure 12a compares CO₂ emissions from power plants between MEIC and REAS2 in Shanxi province where a large amount of coal is extracted and combusted in power plants. EDGAR emissions are also presented in Fig. 12a as a reference. For Shanxi province, MIX, REAS2, and EDGAR included 134, 22, and 24 coal-fired power plants, respectively, demonstrating the omission of many small power plants in REAS2 and EDGAR. In REAS2, only plants with annual CO₂ emissions higher than 1 Tg were processed as point sources (Kurokawa et al., 2013). In the three datasets, a total of 6, 13, and 12 power plants in Shanxi province have annual CO₂ emissions higher than 5 Tg, respectively, indicating significant emission overestimates for large plants in REAS2 and EDGAR. Moreover, the locations of power plants are not accurate in EDGAR, given that CARMA used city centers as the approximate coordinates of power plants (Wheeler and Ummel, 2008). In contrast, coordinates in CPED are obtained from official sources and crosschecked by Google Earth (Liu et al., 2015); the positions of large power plants in REAS2 are also checked manually (Kurokawa et al., 2013).

20 Figure 12b further compares the emission ratios of SO₂/CO₂ in the three inventories for individual power plants over Shanxi. Large deviations of SO₂/CO₂ ratios in MIX are driven by variations of fuel quality, combustion efficiency, and FGD removal efficiency in each plant, which are precisely represented in CPED. In CPED, there is a tendency towards a decrease in SO₂/CO₂ emission ratio with increase of plant size (presented as CO₂ emissions), in accordance with the legislation that large units were required to be equipped with FGD during 2005–2010 (Zhang et al., 2012). Smaller deviations in SO₂/CO₂ emission ratios are found in REAS2, because power plant SO₂ emissions in REAS2 were estimated by using the average FGD penetration rates at provincial level (Kurokawa et al., 2013). EDGAR presented constant ratios for all power plants, indicating that uniform SO₂ and CO₂ emission factors are used.

34835

4.2.2 Agriculture

The agriculture sector is a dominant source of NH₃ emissions, mainly contributed by fertilizer applications and manure managements. MIX incorporated the PKU-NH₃ inventory for China, which estimated agricultural NH₃ emissions using a process-based model to represent the dynamic impact of fertilizer use patterns, meteorological factors, and soil properties (Huang et al., 2012). The new inventory improved on previous studies which used uniform emission factors across time and region. Table 7 compares agricultural NH₃ emissions in China estimated in different emission inventories. Compared to other work, PKU-NH₃ yields lower estimates for fertilizer application but higher estimates for manure management. The differences are mainly because PKU-NH₃ used local correction factors for fertilizer volatilization and manure loss rate (Huang et al., 2012). Top-down inversion of NH₃ emissions by adjoint model and deposition fluxes agrees well with Huang et al. (2012), confirming the validity of the process-based model (Paulot et al., 2014).

15 Besides the magnitude of emissions, a process-based model may also better represent the spatial and temporal variations in emissions. As an example, Fig. 13 compares NH₃ agricultural emissions for MEIC and the PKU-NH₃ inventory for different climate zones. MEIC agrees well with PKU-NH₃ in temperate zones but is significantly higher than PKU-NH₃ in tropical zones. The differences in spatial distributions can be explained by the discrepancies in derived emission factors in the two inventories, given that they used the same activity data from the National Bureau of Statistics of China (NBSC). MEIC used a higher loss rate of NH₃ (20% for urea) for tropical zones and a lower one (15%) for temperate zones following Klimont (2001). With full consideration of fertilization method and soil acidity by grids and by month, PKU-NH₃ estimated 20 9% average NH₃ loss rate for urea for tropical zones and 14% for temperate zone.

34836

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34846

Table 1. Summary of the MIX Asian anthropogenic emission inventory.

Item	Description
Domain	30 countries and regions in Asia
Species	SO ₂ , NO _x , CO, NMVOC, NH ₃ , PM ₁₀ , PM _{2.5} , BC, OC, CO ₂
VOC Speciation	by chemical mechanisms: CB05, SAPRC-99
Sectors	power, industry, residential, transportation, agriculture
Spatial Resolution	0.25° × 0.25°
Seasonality	Monthly
Year	2008, 2010
Data Access	http://www.meicmodel.org/dataset-mix

34849

Table 2. List of regional emission inventories used in this work.

	MEIC v.1.0	PKU-NH ₃	CAPSS	JEI-DB+OPRF	ANL-India	REAS2
Seasonality	Monthly	Monthly	Annual	Monthly	Monthly	Annual
Resolution	0.25°*	1 km	0.25°	1 km	0.1°	0.25°*
CO ₂	X		X	X	X	X
NO _x	X		X	X		X
CO	X		X	X		X
NMVOC	X		X	X		X
NH ₃	X	X		X		X
PM ₁₀	X		X	X		X
PM _{2.5}	X			X		X
BC	X			X	X	X
OC	X			X	X	X
CO ₂	X		X	X		X
NMVOC speciation	X					X

* Power plant emissions are developed with specific geophysical locations and allocated into 0.25° × 0.25° grids.

34850

Table 5. Asian emissions in 2006 based on the same methodology of MIX (Units: Tg for CO₂ and Gg for other species)*.

Regions	SO ₂	NO _x	CO	NMVOG	NH ₃	PM ₁₀	PM _{2.5}	BC	OC	CO ₂
China	34 597 (0.83)	23 719 (1.23)	1 621 (0.95)	20 715 (1.14)	11 203 (0.88)	19 342 (0.86)	13 752 (0.89)	1 771 (1.00)	3 486 (0.97)	7 827 (1.29)
Japan	838 (0.85)	2352 (0.81)	5 555 (0.73)	1538 (0.77)	507 (0.94)	149 (0.76)	109 (0.74)	32 (0.63)	12 (0.68)	1 241 (0.89)
Korea, DPR	233 (0.91)	293 (0.81)	5 430 (0.83)	175 (0.79)	108 (1.02)	319 (0.83)	139 (0.83)	16 (0.86)	18 (0.94)	84 (0.85)
Korea, Rep of	446 (0.94)	1 270 (0.84)	827 (1.01)	794 (1.07)	184 (1.03)	65 (1.91)	42 (2.04)	15 (1.55)	12 (0.69)	510 (1.06)
Mongolia	71 (1.39)	45 (1.37)	523 (1.4)	37 (1.29)	103 (0.93)	75 (1.46)	31 (1.49)	1 (1.58)	2 (1.72)	11 (1.38)
Other East Asia	1 588 (0.90)	3 961 (0.83)	12 668 (0.82)	2 544 (0.87)	903 (0.97)	607 (1.01)	321 (1.02)	65 (0.92)	44 (0.84)	1 846 (0.94)
India	7 476 (1.24)	7 484 (1.28)	55 910 (1.21)	14 685 (1.15)	9 015 (1.09)	5 874 (1.21)	4 327 (1.21)	887 (1.15)	2 415 (1.05)	1 892 (1.20)
Afghanistan	2 (1.33)	111 (1.60)	279 (1.64)	96 (1.46)	131 (1.10)	14 (1.49)	13 (1.48)	5 (1.48)	7 (1.37)	2 (1.25)
Bangladesh	102 (1.30)	283 (1.30)	2 332 (1.10)	711 (1.11)	889 (1.14)	283 (1.21)	203 (1.15)	30 (1.09)	113 (1.07)	67 (1.25)
Bhutan	4 (1.32)	11 (1.21)	256 (1.18)	43 (1.15)	42 (0.99)	21 (1.23)	18 (1.18)	3 (1.14)	12 (1.13)	4 (1.17)
Maldives	3 (0.97)	8 (0.98)	144 (1.05)	7 (1.21)	0 (1.07)	0 (1.28)	0 (1.27)	0 (1.45)	0 (1.40)	2 (1.00)
Nepal	28 (1.08)	72 (1.16)	1 985 (1.06)	405 (1.09)	242 (1.05)	138 (1.08)	128 (1.08)	25 (1.08)	98 (1.08)	32 (1.07)
Pakistan	1 128 (1.24)	816 (1.16)	8 298 (1.12)	1 871 (1.13)	1 543 (1.20)	542 (1.11)	503 (1.11)	103 (1.11)	358 (1.09)	231 (1.14)
Sri Lanka	108 (1.23)	120 (0.96)	1 274 (1.04)	347 (1.06)	112 (1.09)	123 (1.23)	98 (1.13)	15 (1.00)	58 (1.02)	29 (1.08)
Other South Asia	13 76 (1.24)	14 21 (1.20)	14 568 (1.11)	3 481 (1.12)	2 959 (1.16)	1 121 (1.15)	964 (1.12)	181 (1.10)	647 (1.08)	365 (1.15)
Brunei	9 (1.22)	10 (1.15)	6 (0.91)	32 (0.98)	7 (1.13)	1 (0.56)	1 (0.58)	0 (0.67)	0 (0.53)	8 (1.18)
Cambodia	26 (0.99)	46 (1.00)	976 (1.05)	198 (1.07)	121 (1.11)	55 (1.06)	53 (1.06)	11 (1.04)	42 (1.04)	16 (1.04)
Indonesia	16 76 (1.17)	19 99 (1.29)	19 379 (1.23)	6 134 (1.30)	16 34 (1.19)	12 37 (0.96)	9 44 (1.00)	164 (1.08)	663 (1.04)	520 (1.07)
Laos	133 (1.13)	35 (1.17)	388 (1.02)	80 (1.06)	78 (1.12)	24 (1.01)	22 (1.01)	4 (1.02)	16 (1.00)	6 (1.03)
Malaysia	290 (1.26)	505 (1.25)	3 117 (1.20)	1 504 (1.17)	222 (1.15)	191 (1.13)	126 (1.05)	14 (1.13)	33 (1.05)	175 (1.15)
Myanmar	71 (0.94)	76 (1.21)	2 594 (1.04)	654 (1.25)	392 (1.08)	155 (1.06)	149 (1.04)	30 (1.04)	121 (1.03)	47 (1.05)
Philippines	474 (1.06)	288 (1.26)	2 269 (1.03)	812 (1.07)	404 (1.02)	160 (1.21)	114 (1.08)	15 (0.97)	70 (0.98)	94 (1.27)
Singapore	191 (0.92)	112 (1.03)	138 (1.18)	290 (1.15)	12 (0.85)	7 (0.96)	6 (0.97)	1 (1.08)	1 (1.12)	39 (1.08)
Thailand	796 (0.77)	740 (1.09)	7 555 (1.13)	2 031 (1.15)	533 (1.22)	508 (0.97)	285 (0.96)	33 (1.06)	134 (1.11)	271 (1.09)
Vietnam	463 (1.24)	337 (1.31)	7 419 (1.11)	1 584 (1.41)	624 (1.07)	594 (1.20)	485 (1.16)	79 (1.09)	302 (1.06)	184 (1.26)
Southeast Asia	4 129 (1.08)	4 149 (1.23)	43 841 (1.16)	13 319 (1.25)	4 027 (1.14)	2 933 (1.04)	2 184 (1.04)	352 (1.07)	1 383 (1.05)	1 380 (1.12)
Kazakhstan	1 775 (0.59)	499 (1.12)	2 107 (1.59)	423 (1.28)	39 (1.06)	381 (1.16)	192 (1.16)	10 (1.26)	24 (1.17)	191 (1.07)
Kyrgyzstan	30 (0.90)	27 (1.32)	224 (1.66)	30 (1.34)	12 (1.00)	60 (1.03)	27 (1.05)	1 (1.44)	2 (1.21)	5 (1.19)
Tajikistan	11 (1.24)	16 (1.82)	122 (1.57)	25 (1.17)	15 (1.06)	28 (0.80)	15 (0.86)	1 (1.78)	1 (1.34)	3 (1.03)
Turkmenistan	45 (1.42)	97 (1.28)	298 (1.40)	174 (1.37)	13 (1.10)	54 (1.18)	25 (1.20)	2 (1.37)	2 (1.28)	42 (1.22)
Uzbekistan	590 (0.84)	241 (0.94)	808 (1.11)	287 (1.08)	53 (0.94)	325 (1.15)	143 (1.15)	3 (1.03)	9 (1.13)	129 (0.94)
Central Asia	2 451 (0.67)	879 (1.10)	3 558 (1.47)	940 (1.24)	131 (1.01)	847 (1.14)	402 (1.14)	17 (1.27)	39 (1.17)	370 (1.04)
East Siberia	1 711 (0.96)	482 (1.11)	2 437 (1.18)	351 (1.12)	24 (0.97)	380 (0.97)	199 (0.99)	11 (1.28)	19 (1.06)	178 (1.03)
Far East	349 (1.02)	410 (1.19)	2 284 (1.17)	268 (1.13)	20 (0.91)	228 (0.98)	120 (1.03)	17 (1.34)	22 (1.13)	109 (1.10)
Ural	1 510 (0.98)	412 (1.11)	3 757 (1.07)	551 (1.07)	22 (0.99)	1 042 (1.01)	580 (1.03)	17 (1.09)	69 (1.09)	174 (1.07)
West Siberia	647 (1.05)	815 (1.14)	5 399 (1.12)	1 206 (1.09)	43 (0.99)	484 (0.96)	275 (0.98)	27 (1.19)	50 (1.03)	308 (1.10)
Russia Asia	4 217 (0.99)	2 119 (1.13)	13 678 (1.12)	2 376 (1.09)	108 (0.97)	2 132 (0.99)	1 173 (1.01)	72 (1.21)	160 (1.07)	770 (1.08)
Asia	55 832 (0.92)	43 732 (1.19)	324 049 (1.04)	58 059 (1.15)	28 348 (1.02)	32 857 (0.97)	23 124 (0.98)	3 345 (1.06)	8 174 (1.02)	14 430 (1.20)

* Numbers in the parentheses represent emission ratios of 2010 to 2006.

34853

Table 6. Inter-comparisons of emissions among MIX, REAS2 and EDGAR v4.2 for 2008.

Unit: Tg yr ⁻¹	SO ₂	NO _x	CO	NMVOG	NH ₃	PM ₁₀	PM _{2.5}	BC	OC	CO ₂
MIX	49.00	46.38	317.11	60.26	27.66	30.16	21.71	3.40	8.04	15 145
REAS2	52.82	46.17	344.20	65.94	32.74	34.21	23.51	2.95	7.55	15 271
EDGAR v4.2	63.26	36.73	212.16	53.43	20.08	31.08				15 282
China										
MIX	31.41	26.55	175.64	22.10	9.80	17.63	12.74	1.76	3.38	8955
REAS2	33.58	25.55	202.71	27.78	15.00	21.69	14.57	1.60	3.09	9085
EDGAR v4.2	41.35	20.66	106.10	22.60	11.11	14.76				8647
India										
MIX	8.42	8.86	61.80	15.95	9.42	6.65	4.88	0.98	2.48	2103
REAS2	10.08	9.68	61.80	15.95	9.42	6.65	4.88	0.71	2.29	2103
EDGAR v4.2	8.42	6.37	45.58	10.58	4.14	10.80				2307

34854

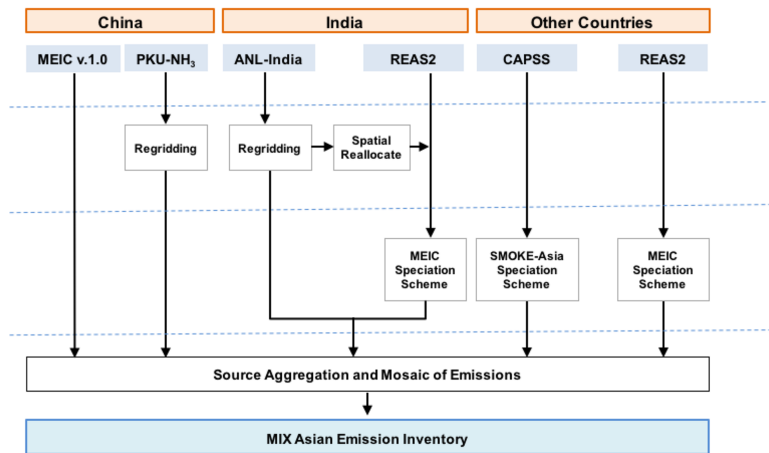


Figure 2. Schematic methodology of the MIX emission inventory development.

34857

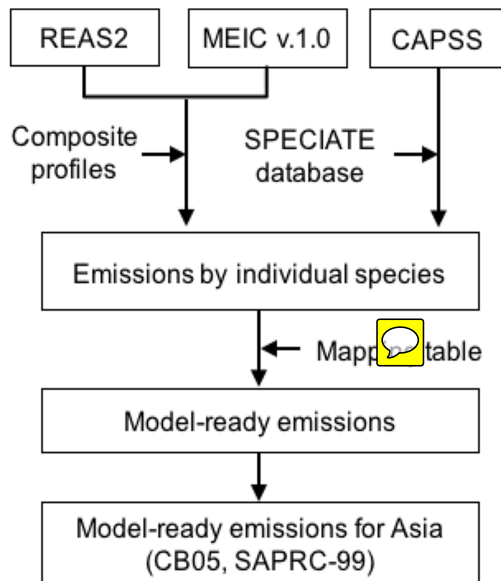


Figure 3. NMVOC speciation scheme used in the MIX inventory development.

34858

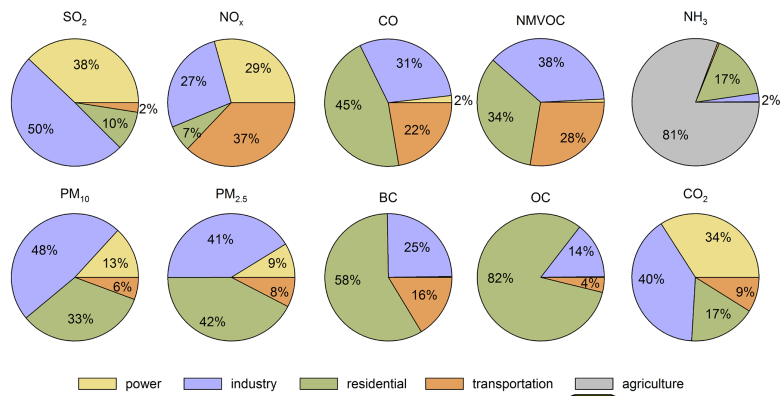


Figure 4. Emission distributions among sectors in Asia in 2010.

34859

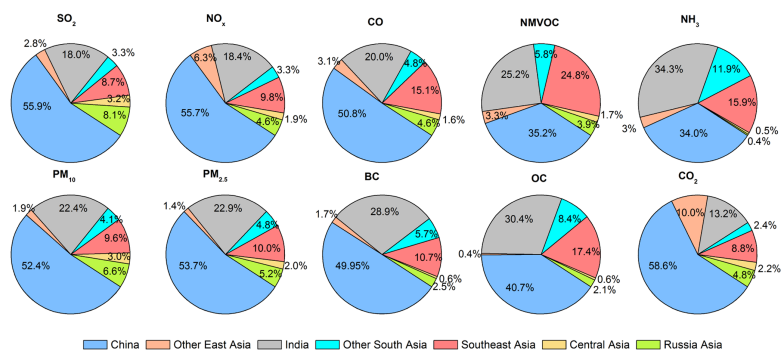


Figure 5. Emissions distributions by Asian regions in 2010.

34860

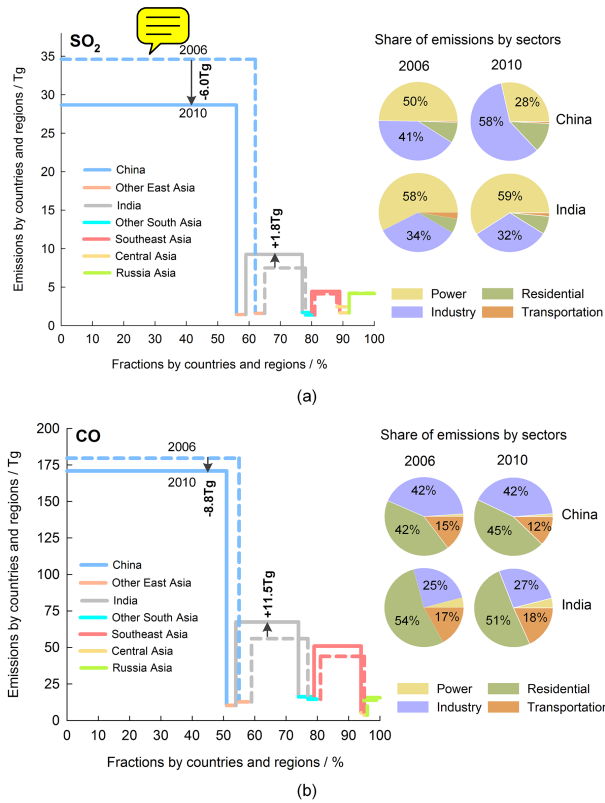


Figure 6. Emission changes from 2006 to 2010 by Asian regions for SO₂ (a) and CO (b).

34861

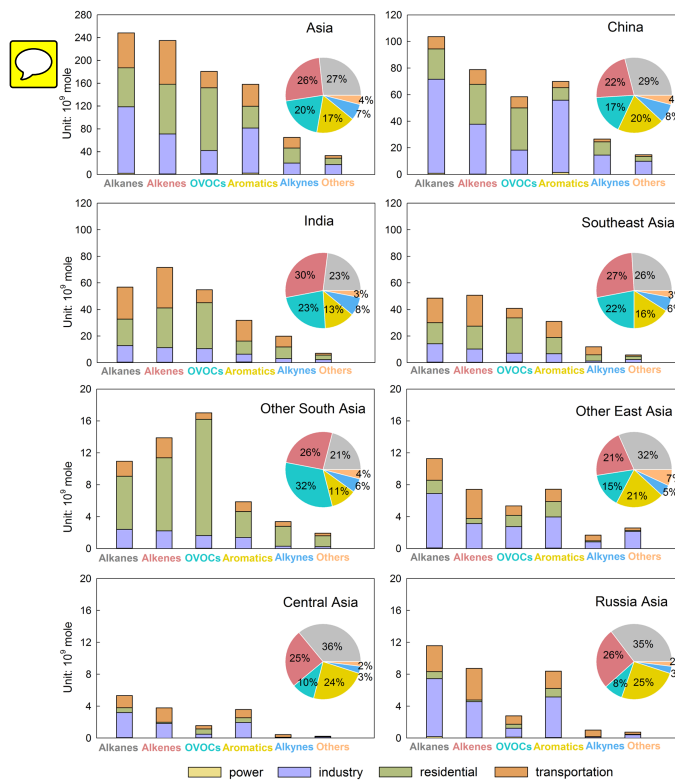


Figure 7. Speciated NMVOC Emissions for the year 2010 by chemical group and by Asian regions.

34862

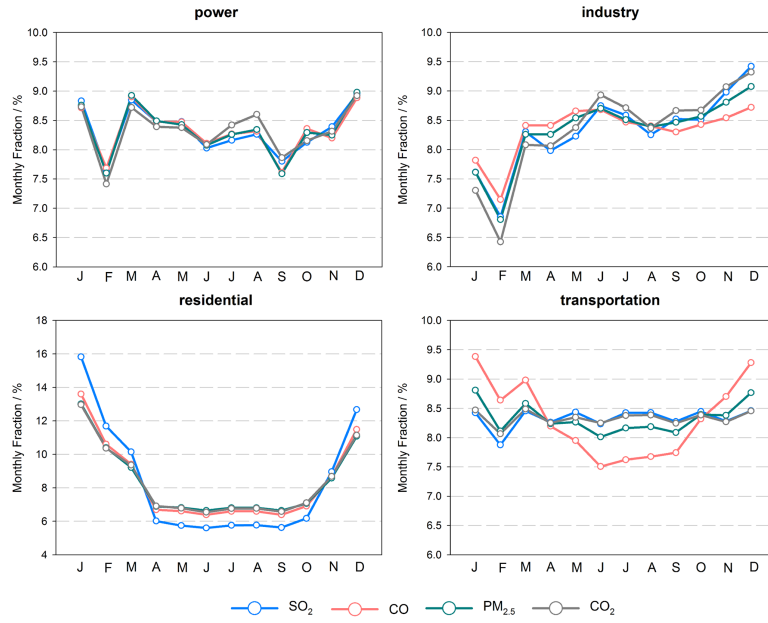


Figure 8. Monthly variations of Asian SO₂, CO, PM_{2.5}, and CO₂ emissions by sector for the year 2010.

34863

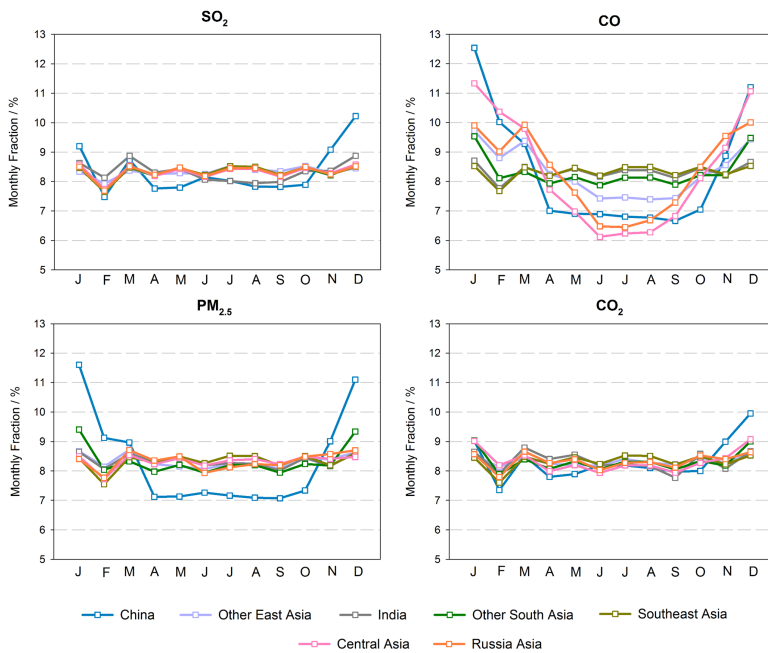


Figure 9. Monthly variations of SO₂, CO, PM_{2.5}, and CO₂ emissions by Asian region for the year 2010.

34864

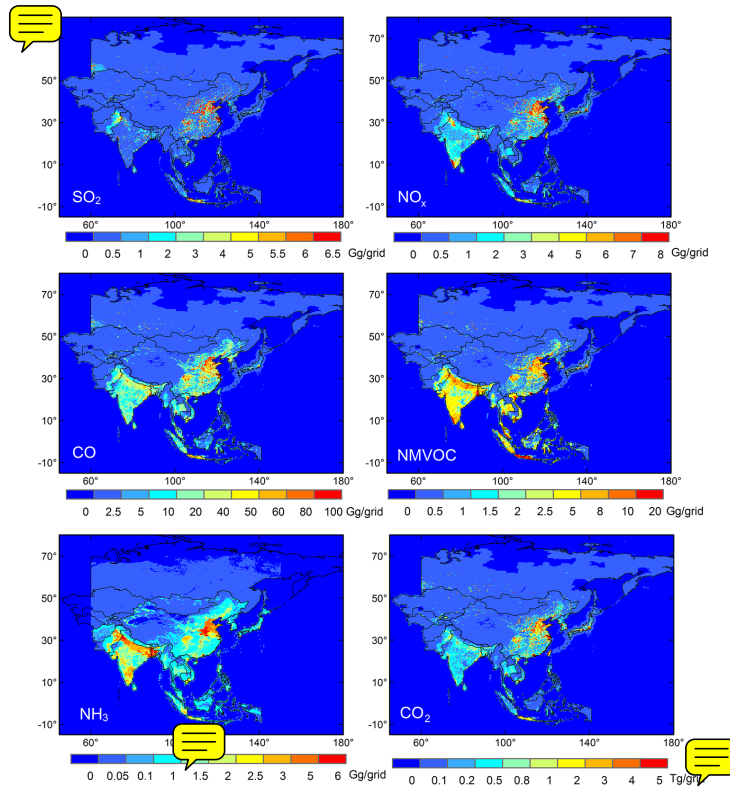


Figure 10.

34865

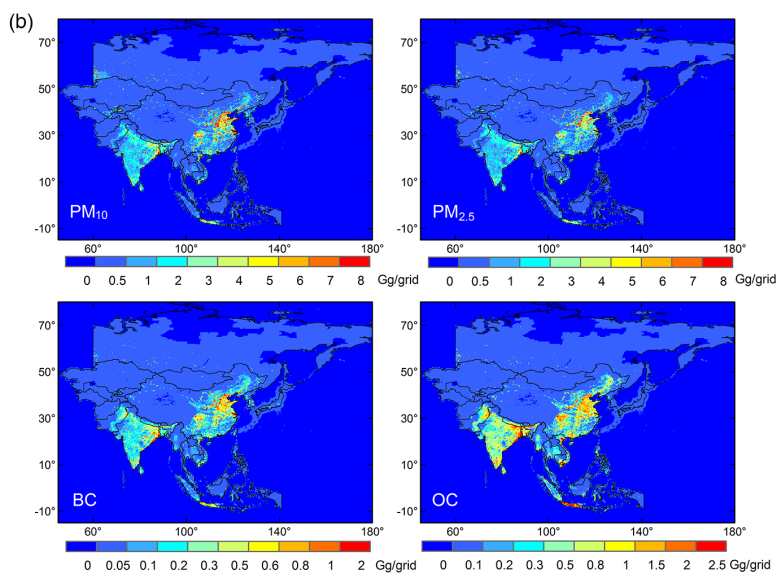


Figure 10. Grid maps for gaseous (a) and aerosol (b) species in the MIX Asia emission inventory, 2010.

34866

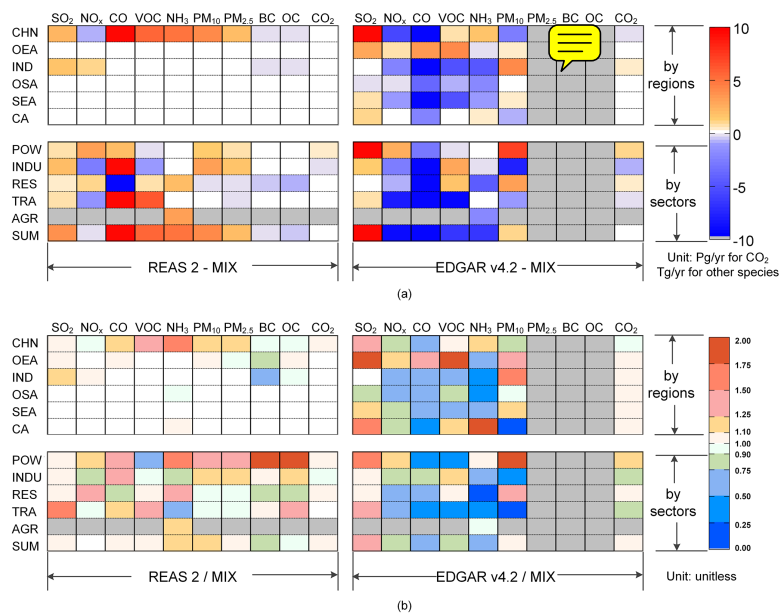


Figure 11. Comparisons of emission estimates between MIX, REAS2 and EDGAR v4.2 by Asian regions and sectors. **(a)** Absolute differences of emission estimates. **(b)** Ratio of emission estimates. Grey grids indicate that the comparison is not available due to absence of emission estimates in EDGAR. Abbreviations of Asian countries and regions are the same as Fig. 5. Abbreviations of sectors are as follows: "POW": power plants; "INDU": industry; "RES": residential; "TRA": transportation; "AGR": agriculture; "SUM": total. "Russia" is not included in the comparison.

34867

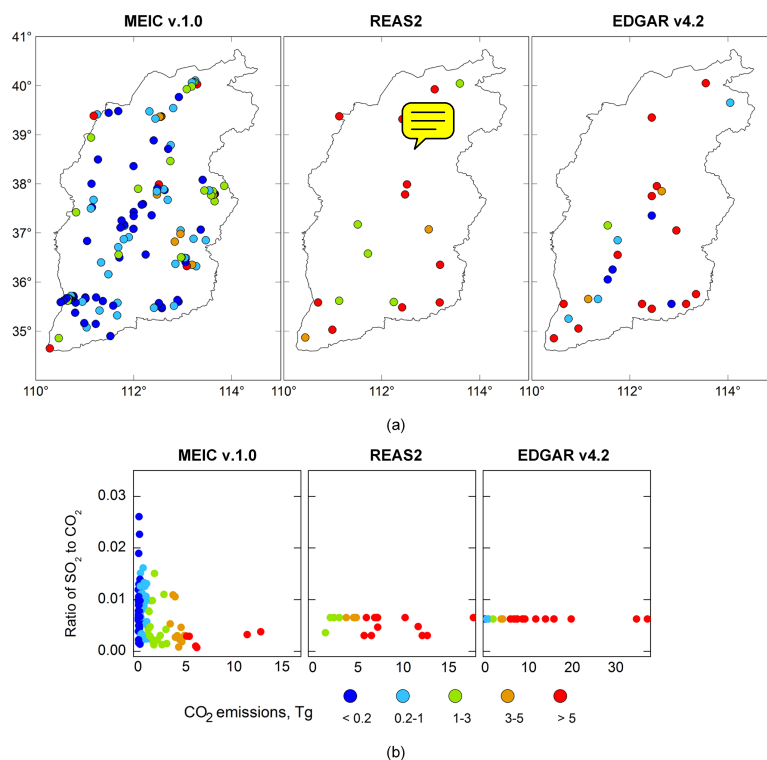


Figure 12. Comparison of 2008 power plants emission estimates between MEIC v.1.0, REAS2 and EDGAR v4.2 for Shanxi province, China. **(a)** Spatial distribution of CO₂ emissions, and **(b)** emission ratio of SO₂ to CO₂.

34868

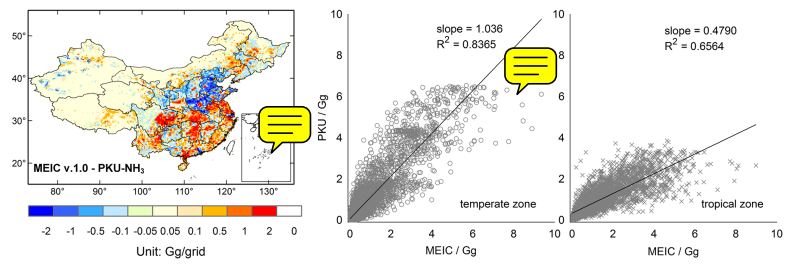


Figure 13. Comparisons of spatial distribution of NH_3 agricultural emissions between MEIC v.1.0 and PKU- NH_3 . Provinces that included in the tropical zones are: Fujian, Guangdong, Hainan, Guangxi, Guizhou, Hubei, Hunan, Yunnan, Sichuan, Jiangxi, Anhui, Zhejiang and Jiangsu. Other provinces are treated as temperate ones.