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

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## Abstract

An anthropogenic emission inventory for Asia is developed for the years 2008 and 2010 to support the Model Inter-Comparison Study for Asia (MICS-Asia) and the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) projects by a mosaic of up-to-date regional emission inventories. Emissions are estimated for all major anthropogenic sources in 30 countries and regions in Asia. We conducted detailed comparisons of different regional emission inventories and incorporated the best-available ones for each region into the mosaic inventory at a uniform spatial and temporal resolution. We estimate the total Asian emissions of ten species in 2010 as follows: 51.3 Tg SO<sub>2</sub>, 52.1 Tg NO<sub>x</sub>, 336.6 Tg CO, 67.0 Tg NMVOC (non-methane volatile organic compounds), 28.8 Tg NH<sub>3</sub>, 31.7 Tg PM<sub>10</sub>, 22.7 Tg PM<sub>2.5</sub>, 3.5 Tg BC, 8.3 Tg OC and 17.3 Pg CO<sub>2</sub>. Emissions from China and India dominate the emissions of Asia for most of the species. We also estimated Asian emissions in 2006 using the same methodology of MIX. The relative change rates of Asian emissions for the period of 2006–2010 are estimated as follows: –8.0 % for SO<sub>2</sub>, +19 % for NO<sub>x</sub>, +4 % for CO, +15 % for NMVOC, +2 % for NH<sub>3</sub>, –3 % for PM<sub>10</sub>, –2 % for PM<sub>2.5</sub>, +6 % for BC, +2 % for OC and +20 % for CO<sub>2</sub>. Model-ready speciated NMVOC emissions for SAPRC-99 and CB05 mechanisms were developed following a profile-assignment approach. Monthly gridded emissions at a spatial resolution of 0.25° × 0.25° are developed and can be accessed from <http://www.meicmodel.org/dataset-mix>.

## 1 Introduction

The Model Inter-Comparison Study for Asia (MICS-Asia) project is currently in Phase III. During the previous two phases, studies have been focused on long-range transport and deposition of pollutants, global inflow of pollutants to Asia, model sensitivities to aerosol parameterization, and emissions over Asia (Carmichael et al., 2002, 2008; Han et al., 2008; Hayami et al., 2008; Holloway et al., 2008; Wang et al., 2008). MICS-Asia

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Phase III aims to conduct further inter-comparisons of atmospheric modeling for Asia and analyze the disagreement of model output and relative uncertainties. With this regard, common meteorological fields, emission data, and boundary conditions should be used. One of the key tasks in MICS-Asia Phase III is to develop a reliable Asian emission inventory as common inputs for model inter-comparisons through integration of state-of-the-art knowledge on Asian emissions.

A reasonable understanding of anthropogenic emissions is essential for atmospheric chemistry and climate research (Xing et al., 2013; Keller et al., 2014). Hence, the community has put tremendous efforts on developing better emission inventories (Granier et al., 2011). For a large geographic region like Asia, compiling a bottom-up emission inventory is a challenging task because it requires a huge amount of local information on energy use, technologies, and environmental regulations for many different countries.

Generally, there are two common approaches to develop a bottom-up emission inventory at regional level. One is using a unified framework of source categories, calculating method, chemical speciation scheme (if applicable), and spatial and temporal allocations (e.g., Streets et al., 2003; Ohara et al., 2007; Lu et al., 2011). Using the unified approach, emissions are estimated in a consistent way with attainable resources. Several Asian emission inventories widely used in the community were developed by the unified approach. Streets et al. (2003) first developed a comprehensive Asian emission inventory for a variety of gaseous and aerosol species for the year 2000 to support the TRACE-P (Transport and Chemical Evolution over the Pacific) campaign (Carmichael et al., 2003), which was subsequently used for MICS-Asia Phase II. Ohara et al. (2007) developed the Regional Emission inventory in Asia (REAS) version 1.1 covering emissions of major species over Asia from 1980 to 2003, which provides estimates of Asian emissions for a long-term period. However, with the unified approach, many region-dependent parameters are shared among different regions due to lack of resources and local knowledge (e.g., emission factors, chemical profiles, spatial proxies, and tem-

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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-

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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:  $\text{SO}_2$ ,  $\text{NO}_x$ , CO, NMVOC (non-methane volatile organic compounds),  $\text{NH}_3$  (ammonia),  $\text{PM}_{10}$  (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  $\text{CO}_2$ . 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

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State Air Pollution Research Center 1999 version, Carter, 2000). Monthly emissions are provided by sector at  $0.25^\circ \times 0.25^\circ$  resolution. Gridded emissions are available from <http://www.meicmodel.org/dataset-mix>. The key elements of the MIX inventory are summarized in Table 1.

5 This paper documents the methodology and emission datasets of the MIX Asian anthropogenic emission inventory. The regional/national inventories used to develop MIX gridded datasets and the mosaic methodology are presented in Sect. 2. Section 3 presents Asian emissions in 2010 and spatial and temporal variations in emissions. Changes in Asian emissions between 2006 and 2010 are also discussed. Section 4  
10 highlights the major improvements in the new inventory by comparing MIX with other Asian emission inventories. Concluding remarks are provided in Sect. 5.

## 2 Compilation of the MIX emission inventory

### 2.1 Methodology

Five emission inventories are selected and incorporated into the mosaic inventory, as  
15 listed in the following: REAS inventory version 2.1 for the whole of Asia (referred to as REAS2 hereafter, Kurokawa et al., 2013), the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University (<http://www.meicmodel.org>), a high-resolution  $\text{NH}_3$  emission inventory by Peking University (referred to as PKU- $\text{NH}_3$  inventory hereafter, Huang et al., 2012), an Indian emission inventory developed by Ar-  
20 gonne National Laboratory (referred to as ANL-India hereafter, Lu et al., 2011; Lu and Streets, 2012), and the official Korean emission inventory from the Clean Air Policy Support System (CAPSS) (Lee et al., 2011).

We then selected different emission datasets for various species for each country. REAS2 was used as the default where local emission data are absent. We used PKU-  
25  $\text{NH}_3$  for  $\text{NH}_3$  and MEIC v.1.0 for all other species over mainland China. For India, emissions of a few species ( $\text{SO}_2$ , BC, OC, and power plant  $\text{NO}_x$ ) were taken from

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ANL-India and REAS2 was used to supplement the missing species in ANL-India. The mosaic process of India emissions is documented in Sect. 2.3. Emissions of the Republic of Korea were provided by CAPSS. Table 2 lists the information of each inventory used in MIX.

5 Figure 2 illustrates the mosaic process for the MIX inventory development. Each dataset was reprocessed to  $0.25^\circ \times 0.25^\circ$  resolution with monthly variations when necessary. NMVOC emissions were speciated to SAPRC-99 and CB05 speciation following the explicit species mapping approach documented in Li et al. (2014) (see Fig. 3). Finally, emissions were aggregated to the five MIX sectors and then assembled to  
10 monthly emission grid maps over Asia with a uniform spatial resolution of  $0.25^\circ \times 0.25^\circ$ .

### 2.2 Components of the MIX emission inventory

#### 2.2.1 REAS2

We used anthropogenic emissions from REAS2 (Kurokawa et al., 2013) to fill the gap where local emission data are not available. REAS2 updated the REAS version 1.1  
15 for both activity data and emission factors by each country and region using global and regional statistics and recent regional specific studies on emissions factors. Improved from its previous version, power plant emissions in REAS2 were estimated by combining information on generation capacity, fuel type, running years, and  $\text{CO}_2$  emissions from the Carbon Monitoring for Action database (CARMA, Wheeler and Ummel,  
20 2008) and the World Electric Power Plants database (WEPP, Platts, 2009). REAS2 extended the domain to include emissions of Central Asia and the Asian part of Russia (referred to as Russia Asia). Readers can refer to Kurokawa et al. (2013) for detailed data sources of activity rates and emission factors assignments for each country and source type. REAS2 is available for the period of 2000–2008. In this work, we updated  
25 the REAS2 to the year 2010, following the same approach documented in Kurokawa et al. (2013).

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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  $\text{CO}_2$  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  $\text{CO}_2$  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

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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 proxies. Users can define the metadata (species, domain range, time period, sectors, spatial resolution, and chemical mechanisms), calculate gridded emissions, and download

data from the website. Monthly emissions at  $0.25^\circ \times 0.25^\circ$  generated from MEIC v.1.0 (referred to as MEIC hereafter) were used in MIX. Emissions were aggregated to four MIX sectors: power, industry, residential, and transportation. Agriculture  $\text{NH}_3$  emissions in MEIC were replaced by PKU- $\text{NH}_3$ , which will be discussed in the next section.

### 5 2.2.3 PKU- $\text{NH}_3$ for China

We used a high-resolution  $\text{NH}_3$  emission inventory in China compiled by Peking University (PKU- $\text{NH}_3$ , Huang et al., 2012) to replace China's  $\text{NH}_3$  emissions in MEIC. MEIC used annual and regional average  $\text{NH}_3$  emission factors to calculate emissions from each source category, while PKU- $\text{NH}_3$  used a process-based model to estimate  $\text{NH}_3$  emissions which parameterized the spatial and temporal variations of emission factors with consideration of ambient temperature, soil property, and other factors. For  $\text{NH}_3$  emissions from fertilizer applications, fertilizer type, soil property, fertilizer application method, application rate, and ambient temperature were used to develop monthly and gridded emission factors. For livestock wastes, emissions were estimated based on a mass-flow methodology by tracing the migration and volatilization of nitrogen from each stage of livestock manure management.

PKU- $\text{NH}_3$  estimated  $\text{NH}_3$  emissions in China (including mainland China and Hong Kong, Macao, and Taiwan) in 2006 for the following sources: livestock wastes, farmland ecosystem, biomass burning, excrement from rural population, chemical industry, waste disposal, and transportation. Open biomass burning was considered as a natural emission source and excluded in the MIX inventory. PKU- $\text{NH}_3$  is available at  $1 \text{ km} \times 1 \text{ km}$  resolution with monthly variation. We then regridded PKU- $\text{NH}_3$  monthly emissions to  $0.25^\circ \times 0.25^\circ$ . In the MIX inventory, 2006 emissions from PKU- $\text{NH}_3$  are used for both 2008 and 2010 since 2006 is the most recent year for emissions in PKU- $\text{NH}_3$  when the MIX inventory was developed. As the major drivers of  $\text{NH}_3$  emissions, synthetic fertilizer consumption and animal population increased by 4 and 9% from 2006 to 2010 respectively, much smaller than the growth rates of coal consumption and vehicle population for the same period.

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### 2.2.4 ANL emission inventories for India

A high-resolution Indian emission inventory developed by ANL (referred to as ANL-India hereafter; Lu et al., 2011) was used in the MIX inventory. ANL-India used a technology-based methodology to estimate  $\text{SO}_2$ , BC, and OC emissions in India for the period of 1996–2010. Major anthropogenic sources including both fossil-fuel and biofuel combustion are covered in ANL-India. Time-dependent trends in emission factors were developed by taking account of the impact of technology changes on emissions (e.g., Habib et al., 2004; Venkataraman et al., 2005). Lu and Streets (2012) further updated power plant emissions in India by calculating emissions at the generating unit level ( $\sim 800$  units in total) based on information from the reports of the Central Electricity Authority (CEA), including geographical location, capacity, fuel type, electricity generation, time the plant was commissioned/decommissioned, etc. The exact location of each power plant was obtained from the Global Energy Observatory (<http://globalenergyobservatory.org>) and crosschecked through Google Earth. The updated unit-based power plant emissions in ANL-India are available for  $\text{SO}_2$ ,  $\text{NO}_x$ , BC, and OC.

ANL-India is available for the period of 1990–2010 at  $0.1^\circ \times 0.1^\circ$  resolution with monthly variations. Emissions are presented by sectors, i.e., power, industry, residential, transportation, and open biomass burning. Monthly variations in ANL-India were developed by sector using various surrogates (Lu et al., 2011). As ANL-India only covers some of the required MIX species ( $\text{SO}_2$ , BC, and OC for all sectors, and  $\text{NO}_x$  for power plants), monthly emissions by sector from ANL-India were first regridded to  $0.25^\circ \times 0.25^\circ$  and then merged with REAS2 before being implemented in MIX. The merge process will be presented in Sect. 2.3.

### 25 2.2.5 CAPSS inventory for the Republic of Korea

For the Republic of Korea, we used the CAPSS emission inventory developed by the National Institute of Environmental Research of Korea (Lee et al., 2011). CAPSS esti-

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mated emissions with four levels of source classifications. We mapped emissions from 12 first-level aggregated source categories (SCC1) to five sectors in MIX. The CAPSS inventory included emissions for five regulated air pollutants,  $\text{SO}_2$ ,  $\text{NO}_x$ , CO, NMVOC, and  $\text{PM}_{10}$ . We derived sector-specific emission ratios between  $\text{PM}_{10}$  and the other aerosol components from Lei et al. (2011) and applied those ratios to estimate  $\text{PM}_{2.5}$ , BC and OC emissions. In the MIX inventory, we used the 2008 and 2009 CAPSS inventories to represent 2008 and 2010 emissions of the Republic of Korea, because 2009 is the most recent year of CAPSS inventory at the time the MIX inventory was developed. In the CAPSS inventory, point sources, area sources, and mobile sources were processed using different spatial allocation approaches (Lee et al., 2011). We used the  $0.25^\circ \times 0.25^\circ$  emission product from CAPSS as input for the MIX inventory. Only annual total emissions were presented in the CAPSS inventory. In the MIX inventory, we assume no monthly variation in emissions in the Republic of Korea.

### 2.3 Mosaic of Indian emission inventory

In this work, REAS2 is used to supplement the missing species in ANL-India. To reduce possible inconsistencies from implementation of the two different inventories, we have reprocessed ANL-India and REAS2 emissions over India in the following two ways. First, ANL-India is available for  $\text{SO}_2$ ,  $\text{NO}_x$ , BC, and OC for power plants. Because ANL-India used CEA reports to derive information of individual power generation units, while REAS2 used the CARMA and WEPP databases to get similar information, direct merging of the two products may introduce inconsistency due to a mismatch of unit information in the two databases. In this work, we first generated the spatial distribution of fuel consumption by type at  $0.25^\circ \times 0.25^\circ$  resolution by aggregating unit-level information in ANL-India, we then used these spatial proxies to reallocate total power plant emissions of CO, NMVOC,  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ , and  $\text{CO}_2$  in REAS2 by fuel type.

Second, we used BC and OC emissions from ANL-India but used  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  emissions from REAS2. In certain grids, the sum of BC and OC emissions may exceed  $\text{PM}_{2.5}$  emissions because the two inventories may use different activity data, emis-

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sion factors and spatial proxies. The so-called "PMfine" species in chemical transport models are usually calculated by subtracting BC and OC emissions from total  $\text{PM}_{2.5}$  emissions, leading to negative emissions of "PMfine" in those grids. In this case, we adjusted the emissions of  $\text{PM}_{2.5}$  to the sum of BC and OC emissions for each sector.

### 2.4 NMVOC speciation of the MIX inventory

In the MIX inventory, we provide model-ready speciated NMVOC emissions over Asia (except the Republic of Korea) for both CB05 and SAPRC-99 chemical mechanisms, by using the explicit species mapping approach and updated NMVOC profiles developed in Li et al. (2014), as illustrated in Fig. 3. Following Li et al. (2014), NMVOC emissions for CB05 and SAPRC-99 species are calculated as follows:

$$\text{EVOC}(i, k, m) = \sum_{j=1}^n \left[ \frac{\text{EVOC}(i, k) \times X(i, j)}{\text{mol}(j)} \times C(j, m) \right]$$

Where  $k$  is the region;  $m$  is species type in CB05 or SAPRC-99 mechanisms;  $n$  is the number of species emitted from source  $i$ . EVOC is the total NMVOC emissions by source type. In this work, emissions in China and other Asian countries were derived from MEIC and REAS2 respectively.  $X(i, j)$  is the mass fraction of species  $j$  in the total NMVOC emissions for source  $i$ , which is taken from the profiles developed by Li et al. (2014). Those profiles were constructed by grouping and averaging multiple profiles from both local measurements and the SPECIATE database (Hsu and Divita, 2009; Simon et al., 2010).  $\text{mol}(j)$  is the mole weight of species  $j$ ; and  $C(j, m)$  is the conversion factor between  $j$  and  $m$  obtained from Carter (2013).

For the Republic of Korea, the SMOKE-Asia model developed by Woo et al. (2012) was used to calculate model-ready NMVOC emissions for both CB05 and SAPRC-99 mechanisms. NMVOC emissions from the CAPSS were mapped to Source Classification Codes (SCCs) and country/state/county (FIPS) code in SMOKE-Asia model and

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

### 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 SO<sub>2</sub>, 52.1 Tg NO<sub>x</sub>, 336.6 Tg CO, 67.0 Tg NMVOC, 28.8 Tg NH<sub>3</sub>, 31.7 Tg PM<sub>10</sub>, 22.7 Tg PM<sub>2.5</sub>, 3.5 Tg BC, 8.3 Tg OC and 17.3 Pg CO<sub>2</sub>. Figure 4 presents the emission distributions among sectors over Asia in 2010. Among the different sectors, the industrial sector has the largest contribution to SO<sub>2</sub> (50 % of total), NMVOC (38 %), PM<sub>10</sub> (48 %), and CO<sub>2</sub> (40 %) emissions. Power plants have significant contributions for SO<sub>2</sub> (38 % of total), NO<sub>x</sub> (29 %), and CO<sub>2</sub> (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 NH<sub>3</sub>, with more than 50 % contribution for SO<sub>2</sub>, NO<sub>x</sub>, CO, PM<sub>10</sub>, PM<sub>2.5</sub> and CO<sub>2</sub> emissions. Following China, India is the largest contributor for NH<sub>3</sub> 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, NH<sub>3</sub>, 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 SO<sub>2</sub>, 29.1 Tg NO<sub>x</sub>, 170.9 Tg CO, 23.6 Tg NMVOC, 9.8 Tg NH<sub>3</sub>, 16.6 Tg PM<sub>10</sub>, 12.2 Tg PM<sub>2.5</sub>, 1.8 Tg BC, 3.4 Tg OC and 10.1 Pg CO<sub>2</sub>. Overall, industry is the largest emitter of China’s anthro-

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pogenic emissions, contributing 49 % of the total CO<sub>2</sub> emissions and 59, 39, 61 %, and 50 % of SO<sub>2</sub>, NO<sub>x</sub>, NMVOC, and PM<sub>2.5</sub> 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 SO<sub>2</sub> emissions in China in 2010 surpassed SO<sub>2</sub> emissions from the US and Europe combined. Power plants contributed 32 % of the total CO<sub>2</sub> emissions and 28, 33 %, and 7 % of SO<sub>2</sub>, NO<sub>x</sub>, and PM<sub>2.5</sub> emissions, respectively. Emission ratios of SO<sub>2</sub>/CO<sub>2</sub> and PM<sub>2.5</sub>/CO<sub>2</sub> 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 CO<sub>2</sub> 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<sub>x</sub>, 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.

25 In the MIX inventory, Indian emissions in 2010 are estimated as follows: 9.3 Tg SO<sub>2</sub>, 9.6 Tg NO<sub>x</sub>, 67.4 Tg CO, 16.9 Tg NMVOC, 9.9 Tg NH<sub>3</sub>, 7.1 Tg PM<sub>10</sub>, 5.2 Tg PM<sub>2.5</sub>, 1.0 Tg BC, 2.5 Tg OC and 2.3 Pg CO<sub>2</sub>. 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 CO<sub>2</sub> emissions and the largest contributor for CO, NMVOC, PM<sub>2.5</sub>, BC, and OC emissions, in which more than 70 % of those emissions

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are contributed by biofuel combustion. With the rapid growth of coal-fired generation units, SO<sub>2</sub> emissions from Indian power plants are estimated to be 5.5 Tg in 2010, contributing 59 % of the total SO<sub>2</sub> emissions. The SO<sub>2</sub>/CO<sub>2</sub> 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<sub>x</sub> 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 SO<sub>2</sub>, +19 % for NO<sub>x</sub>, +4 % for CO, +15 % for NMVOC, +2 % for NH<sub>3</sub>, -3 % for PM<sub>10</sub>, -2 % for PM<sub>2.5</sub>, +6 % for BC, +2 % for OC and +20 % for CO<sub>2</sub>. Growth in CO<sub>2</sub> 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 SO<sub>2</sub> 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 SO<sub>2</sub> emission changes over Asia. SO<sub>2</sub> 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 SO<sub>2</sub> emission reduction over Asia. In contrast, SO<sub>2</sub> emissions in India increased by 27 % during 2006–2010, owing to dramatic construction of new

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power plants and lack of emission control facilities (Garg et al., 2001, 2006). As a consequence, the Indian share of the total Asian SO<sub>2</sub> emissions increased from 13 % in 2006 to 18 % in 2010. NO<sub>x</sub> 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<sub>x</sub> 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, PM<sub>10</sub>, and PM<sub>2.5</sub>, 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., 2013; 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

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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 Sect. 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 SO<sub>2</sub>, CO, PM<sub>2.5</sub>, and CO<sub>2</sub> 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 SO<sub>2</sub> emissions in winter are even higher than other species, because SO<sub>2</sub> emissions from China dominate residential emissions in Asia (70 % of total), of which

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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 SO<sub>2</sub>, CO, PM<sub>2.5</sub>, and CO<sub>2</sub> 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<sub>2.5</sub> 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 SO<sub>2</sub>, NO<sub>x</sub>, CO, NH<sub>3</sub>, NMVOC, PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC, and CO<sub>2</sub>. 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](http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123)).

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## 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 REAS differ within 10 % for most species, except for  $\text{NH}_3$  (18 % higher in REAS),  $\text{PM}_{10}$  (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 recent 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  $\text{SO}_2$ , but 20, 33, 11, 27 % lower for  $\text{NO}_x$ , CO, NMVOC, and  $\text{NH}_3$ , respectively.  $\text{PM}_{10}$  and  $\text{CO}_2$  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  $\text{SO}_2$  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  $\text{NO}_x$ , CO, and NMVOC estimates in the two inventories. For instance, EDGAR estimates lower  $\text{NO}_x$  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

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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.  $\text{NH}_3$  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 REAS, which were incorporated into MIX for Asian regions except China. MIX estimated 3.4 Tg $\text{NH}_3$  emissions from wastewater treatment in Asia in 2008, which are more than two orders of magnitude higher than EDGAR estimates. Differences in  $\text{PM}_{10}$  emissions at the sector level are also huge; similar estimates of  $\text{PM}_{10}$  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  $\text{NO}_x$ , and 10 % for  $\text{SO}_2$  and  $\text{CO}_2$ , 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

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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 CO<sub>2</sub> emissions higher than 5 Tg) contributed 77 % of power plant CO<sub>2</sub> emissions in China, compared to a 47 % contribution of large plants in MIX.

5 Figure 12a compares CO<sub>2</sub> 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  
10 EDGAR. In REAS2, only plants with annual CO<sub>2</sub> 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 CO<sub>2</sub> 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  
15 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).

Figure 12b further compares the emission ratios of SO<sub>2</sub>/CO<sub>2</sub> in the three inventories  
20 for individual power plants over Shanxi. Large deviations of SO<sub>2</sub>/CO<sub>2</sub> 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 SO<sub>2</sub>/CO<sub>2</sub> emission ratio with increase of plant size (presented as CO<sub>2</sub> emissions), in accordance with the legislation that large units were required to  
25 be equipped with FGD during 2005–2010 (Zhang et al., 2012). Smaller deviations in SO<sub>2</sub>/CO<sub>2</sub> emission ratios are found in REAS2, because power plant SO<sub>2</sub> 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 SO<sub>2</sub> and CO<sub>2</sub> emission factors are used.

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#### 4.2.2 Agriculture

The agriculture sector is a dominant source of NH<sub>3</sub> emissions, mainly contributed by fertilizer applications and manure managements. MIX incorporated the PKU-NH<sub>3</sub> inventory for China, which estimated agricultural NH<sub>3</sub> emissions using a process-based  
5 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 NH<sub>3</sub> emissions in China estimated in different emission inventories. Compared to other work, PKU-NH<sub>3</sub> yields lower estimates for fertilizer application but higher estimates for manure management. The differences are mainly because PKU-NH<sub>3</sub> used  
10 local correction factors for fertilizer volatilization and manure loss rate (Huang et al., 2012). Top-down inversion of NH<sub>3</sub> 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).

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 NH<sub>3</sub> agricultural emissions for MEIC and the PKU-NH<sub>3</sub> inventory for different climate zones. MEIC agrees well with PKU-NH<sub>3</sub> in temperate zones but is significantly higher than PKU-NH<sub>3</sub> in tropical zones. The differences in spatial distributions can be explained by the discrepancies in derived emission factors in the two inventories, given  
20 that they used the same activity data from the National Bureau of Statistics of China (NBSC). MEIC used a higher loss rate of NH<sub>3</sub> (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-NH<sub>3</sub> estimated  
25 9 % average NH<sub>3</sub> loss rate for urea for tropical zones and 14 % for temperate zone.

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### 4.2.3 Other sectors

This section further discusses the differences between MIX and REAS2 over China. EDGAR is not compared here because references to the detailed underlying data used in EDGAR are not available. Figure S1 in the Supplement compares MIX and REAS2 estimates for China for 2008 by species and by sector. The two inventories generally agree well, given that both MEIC and REAS2 incorporate the most recent advances in emission inventory studies in China. The major differences between the two inventories are discussed below with explanation for possible reasons.

REAS2 estimates higher CO and PM emissions than MEIC for the industrial sector. This is probably because REAS2 underestimates the emission control progress in China's industrial sector after 2005. During the 11th Five-Year Plan (2005–2010), China has implemented a series of new standards to restrict industrial emissions, leading to a downward trend in emission factors after 2005 (Zhao et al., 2013). For the industrial sector, REAS2 adopted CO and PM emission factors from Streets et al. (2006) and Lei et al. (2011), respectively, which represent the real-world emission characteristics before the year 2005. Using those emission factors may have overestimated industrial emissions. Moreover, REAS2 estimated an increasing trend in China's CO emissions during 2005–2008, which is opposite to the downward trend derived from satellite-based constraints for the same period (Yumimoto et al., 2014; Yin et al., 2015), confirming that REAS2 may overestimate CO emissions in China after 2005. Transportation emissions in MEIC and REAS2 differ significantly for different species. Compared to REAS2, MEIC estimates much lower emissions for CO and NMVOC (dominated by gasoline vehicles) but higher emissions for NO<sub>x</sub> and PM (dominated by diesel vehicles).

### 4.3 India

For India, MIX used ANL-India for SO<sub>2</sub>, BC, and OC emissions and REAS2 for other species. Here we compare ANL-India and REAS2 for SO<sub>2</sub>, BC, and OC emissions, to

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evaluate the impact of using ANL-India. Both ANL-India and REAS2 used energy consumption data from the International Energy Agency (IEA), hence the differences are mainly from emission factors. Reasonable agreements are found in total emissions over India (differing by 8–28%), while discrepancies are large at the sector level. REAS2 estimates 50% higher SO<sub>2</sub> estimates for all sectors except power plants, most likely from different assumptions about the sulfur content of fuels. For BC and OC, the ratio between REAS2 and ANL-India varies from 0.4 to 11.8 at the sector level, indicating large differences in emission factor selections. ANL-India used emission factors from a global database (Bond et al., 2004) with updates of a few recent measurements (Lu et al., 2011), while REAS2 used a local database developed many years ago (Reddy and Venkataraman, 2002a, b). It should be noted that local emission measurements in India are still too few to support accurate emission estimates. More measurements should be conducted in the future to remedy this situation.

When implementing REAS2 to MIX over India, power plant emissions were redistributed using spatial distributions derived from ANL-India at 0.25° × 0.25° resolution (see Sect. 2.3). We believe that it will improve the accuracy because power plant emissions in ANL-India were estimated by each unit and allocated manually by Google Earth. A total of 68 power plants are identified in REAS2, compared to 145 plants in ANL-India. The two inventories generally agree well for the grids in which both inventories allocate power plant emissions. Lu and Streets (2012) found that the magnitudes and locations of power plant NO<sub>x</sub> emissions (from ANL-India) are matched well with satellite-based observations over India, providing confidence to the accuracy of ANL-India estimates. From all the comparisons discussed above, we can conclude that emissions are well depicted in MIX due to integration of the most-recent regional inventories.

## 5 Concluding remarks

In this work, we developed a new anthropogenic emission inventory for Asia for the years of 2008 and 2010 by constructing a mosaic of several regional and national emission inventories. MEIC, PKU-NH<sub>3</sub>, ANL-India, and CAPSS inventories are used to represent the best available emission data for China, India, and Korea, supplemented with REAS2 to fill gaps. By harmonizing these inventories, monthly emission grids maps for ten species over Asia were generated for five sectors (power, industry, residential, transportation, and agriculture) at a uniform spatial resolution of 0.25° × 0.25°. Gridded speciated NMVOC emissions for SAPRC-99 and CB05 mechanisms were also developed at the same temporal and spatial resolution. This new Asian emission inventory, named MIX, provides model-ready anthropogenic emissions for the MICS-Asia Phase III assessment. The MIX inventory has been also incorporated into the HTAP v2 gridded emission inventory (Janssens-Maenhout et al., 2015) to support the TF HTAP assessment. Gridded emissions are available from the following website: <http://www.meicmodel.org/dataset-mix>.

The MIX inventory provides a consistent emission input for Asian regions for global and regional modeling activities. Compared to previous global and regional inventories over Asia, MIX has improved the accuracy of emission estimates as well as spatial and temporal distributions due to extensive inclusion of local knowledge. For example, CPED has largely improved the accuracy of power plant emission estimates for China over other inventories that used the CARMA global power plant database. We expect that the MIX inventory can provide a good foundation for air quality modeling and can help to improve the model performance. On the other hand, the MIX inventory still has some limitations. It is very difficult to conduct quantitative uncertainty analysis for a mosaic inventory, which limits understanding of the reliability of the MIX inventory. Validation of the MIX inventory could be provided by comparing model predictions with in-situ and satellite observations. For MIX, the inter-comparison of emissions between regions is less valid because different methodologies were used. Harmonizing emis-

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sion inventory methodology among different regions and research groups could help to resolve this issue in the future.

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

Item	Description
Domain	30 countries and regions in Asia
Species	SO <sub>2</sub> , NO <sub>x</sub> , CO, NMVOC, NH <sub>3</sub> , PM <sub>10</sub> , PM <sub>2.5</sub> , BC, OC, CO <sub>2</sub>
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	<a href="http://www.meicmodel.org/dataset-mix">http://www.meicmodel.org/dataset-mix</a>

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

	MEIC v.1.0	PKU-NH <sub>3</sub>	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°*
SO <sub>2</sub>	X		X	X	X	X
NO <sub>x</sub>	X		X	X		X
CO	X		X	X		X
NMVOC	X		X	X		X
NH <sub>3</sub>	X	X		X		X
PM <sub>10</sub>	X		X	X		X
PM <sub>2.5</sub>	X			X		X
BC	X			X	X	X
OC	X			X	X	X
CO <sub>2</sub>	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.

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**Table 5.** Asian emissions in 2006 based on the same methodology of MIX (Units: Tg for CO<sub>2</sub> and Gg for other species)\*.

Regions	SO <sub>2</sub>	NO <sub>x</sub>	CO	NM VOC	NH <sub>3</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	BC	OC	CO <sub>2</sub>
China	34 597 (0.83)	23 719 (1.23)	179 626 (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)	5888 (0.73)	1538 (0.77)	507 (0.94)	149 (0.76)	109 (0.74)	32 (0.63)	12 (0.68)	1241 (0.89)
Korea, DPR	233 (0.91)	293 (0.81)	5430 (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)	1270 (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	1588 (0.90)	3961 (0.83)	12 668 (0.82)	2544 (0.87)	903 (0.97)	607 (1.01)	321 (1.02)	65 (0.92)	44 (0.84)	1846 (0.94)
India	7476 (1.24)	7484 (1.28)	55 910 (1.21)	14 685 (1.15)	9015 (1.09)	5874 (1.21)	4327 (1.21)	887 (1.15)	2415 (1.05)	1892 (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)	263 (1.30)	2332 (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)	1985 (1.06)	405 (1.09)	242 (1.05)	138 (1.08)	128 (1.08)	25 (1.08)	98 (1.08)	32 (1.07)
Pakistan	1128 (1.24)	816 (1.16)	8298 (1.12)	1871 (1.13)	1543 (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)	1274 (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	1376 (1.24)	1421 (1.20)	14 568 (1.11)	3481 (1.12)	2959 (1.16)	1121 (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	1676 (1.17)	1999 (1.29)	19 379 (1.23)	6134 (1.30)	1634 (1.19)	1237 (0.96)	944 (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)	3117 (1.20)	1504 (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)	2594 (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)	2269 (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)	7555 (1.13)	2031 (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)	7419 (1.11)	1584 (1.41)	624 (1.07)	594 (1.20)	485 (1.16)	79 (1.09)	302 (1.06)	184 (1.26)
Southeast Asia	4129 (1.08)	4149 (1.23)	43 841 (1.16)	13 319 (1.25)	4027 (1.14)	2933 (1.04)	2184 (1.04)	352 (1.07)	1383 (1.05)	1380 (1.12)
Kazakhstan	1775 (0.59)	499 (1.12)	2107 (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 (1.62)	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	2451 (0.67)	879 (1.10)	3558 (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	1711 (0.96)	482 (1.11)	2437 (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)	2284 (1.17)	268 (1.13)	20 (0.91)	228 (0.98)	120 (1.03)	17 (1.34)	22 (1.13)	109 (1.10)
Ural	1510 (0.98)	412 (1.11)	3757 (1.07)	551 (1.07)	22 (0.99)	1042 (1.01)	580 (1.03)	17 (1.09)	69 (1.09)	174 (1.07)
West Siberia	647 (1.05)	815 (1.14)	5399 (1.12)	1206 (1.09)	43 (0.99)	484 (0.96)	275 (0.98)	27 (1.19)	50 (1.03)	308 (1.10)
Russia Asia	4217 (0.99)	2119 (1.13)	13 678 (1.12)	2376 (1.09)	108 (0.97)	2132 (0.99)	1173 (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)	3345 (1.06)	8174 (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.

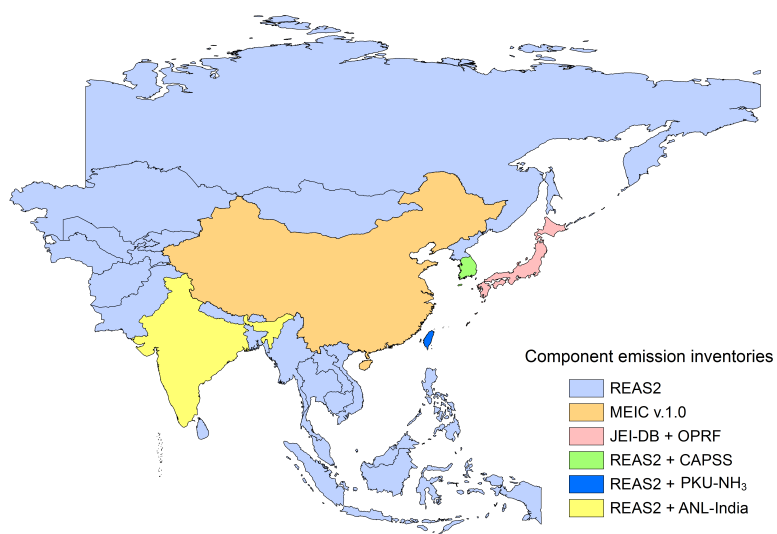
Unit: Tgyr <sup>-1</sup>	SO <sub>2</sub>	NO <sub>x</sub>	CO	NM VOC	NH <sub>3</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	BC	OC	CO <sub>2</sub>
Asia										
MIX	49.00	46.38	317.11	60.26	27.66	30.16	21.71	3.40	8.04	15 145
REAS2	52.82	46.17	344.20	65.94	32.74	34.21	23.51	2.95	7.55	15 271
EDGAR v4.2	63.26	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

**Table 7.** NH<sub>3</sub> agriculture emission estimates for China.

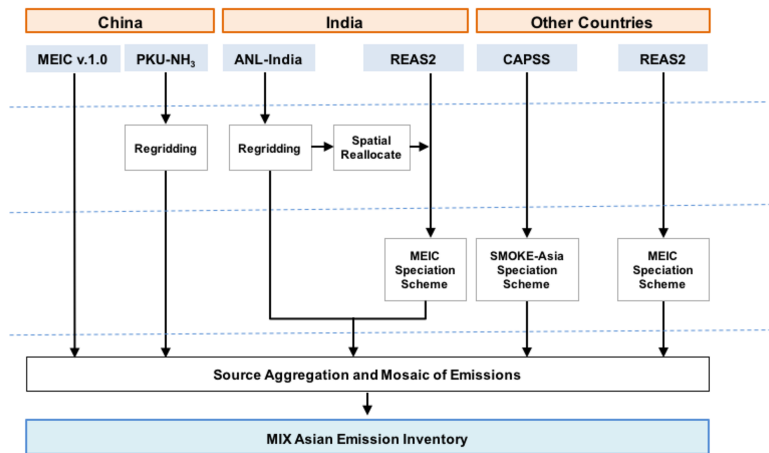
Unit: TgNH <sub>3</sub> yr <sup>-1</sup>	PKU-NH <sub>3</sub>	MEIC v.1.0	REAS2	EDGAR v4.2	MASAGE_NH <sub>3</sub>
Year	2006	2008	2008	2008	2005–2008
Fertilizer application	3.20	4.40	9.40	8.26	3.64
Live stock	5.30	5.30	2.80	2.31	5.83

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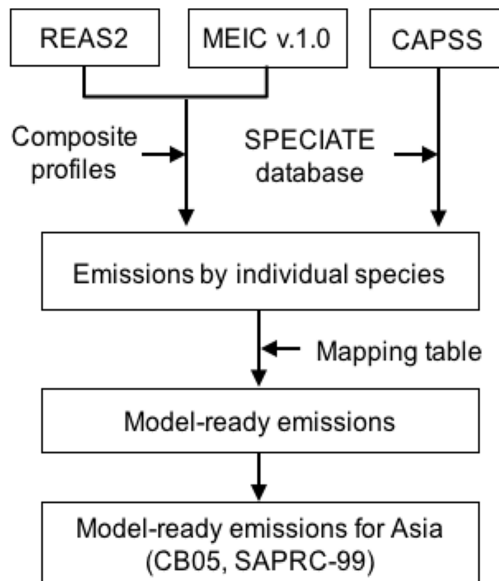
**Figure 1.** Domain and component of the MIX emission inventory.

34856



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

34857



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

34858

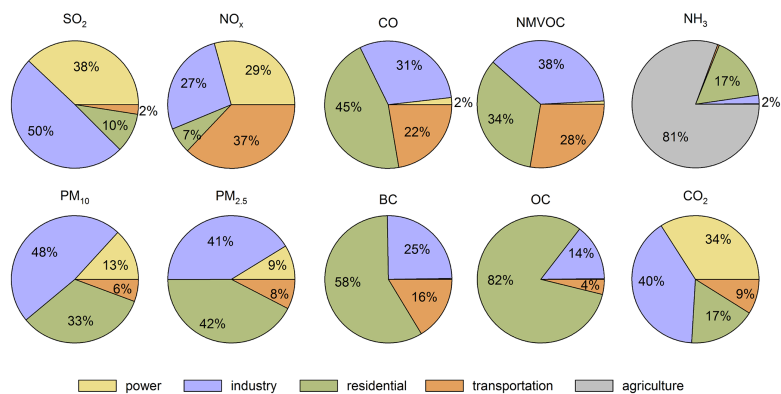


Figure 4. Emission distributions among sectors in Asia in 2010.

34859

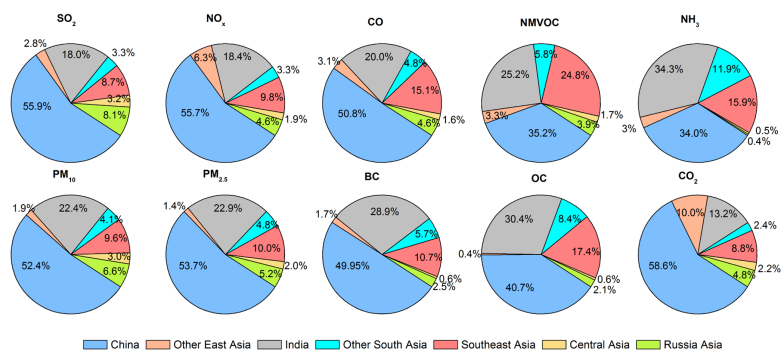


Figure 5. Emissions distributions by Asian regions in 2010.

34860



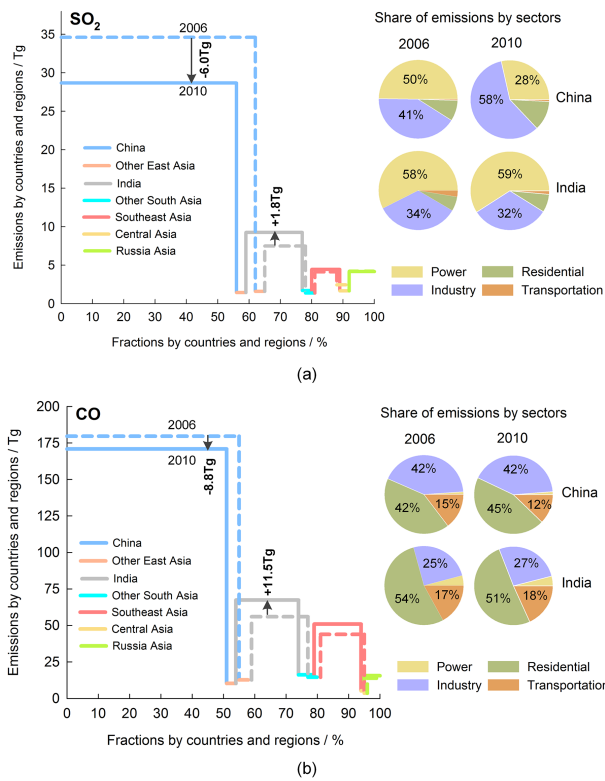


Figure 6. Emission changes from 2006 to 2010 by Asian regions for SO<sub>2</sub> (a) and CO (b).

34861

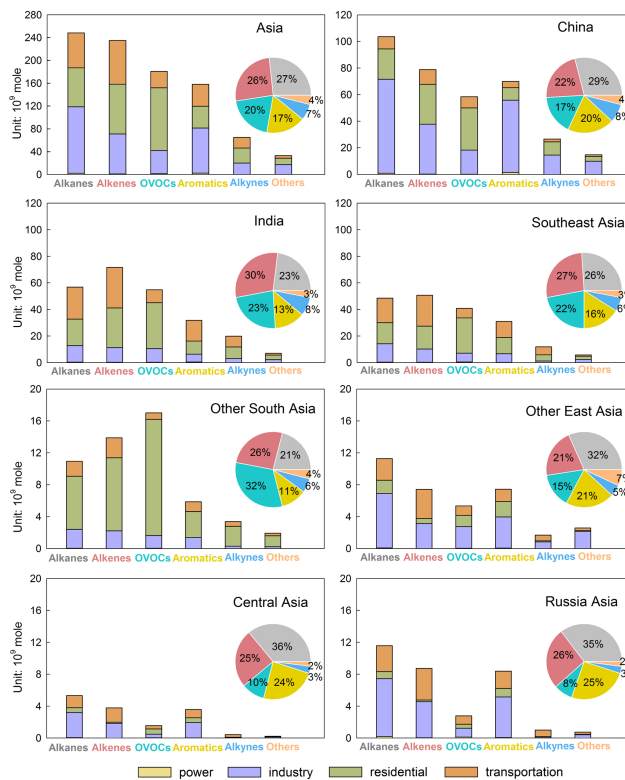
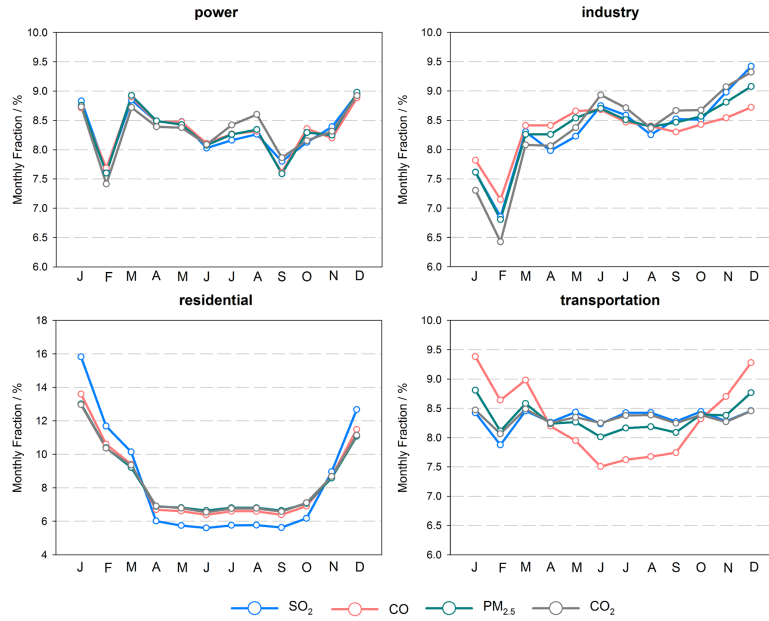


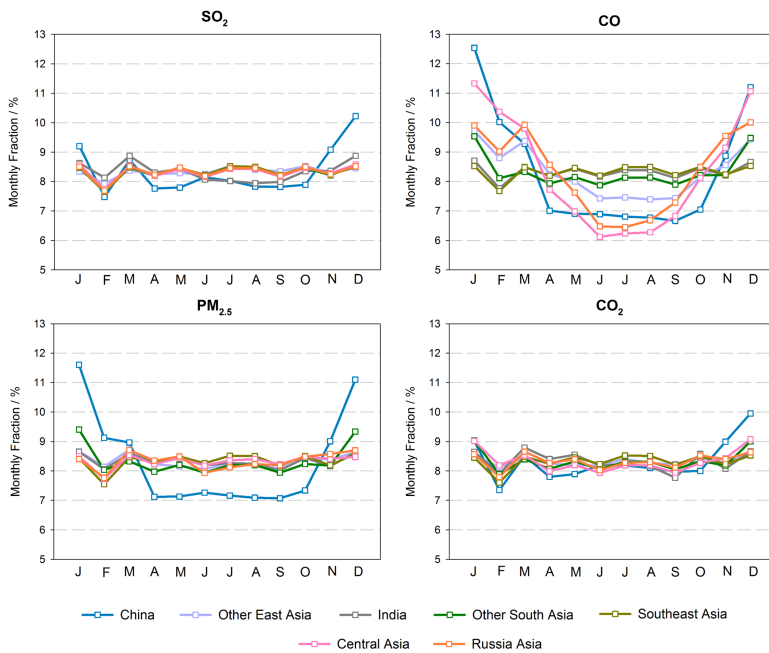
Figure 7. Speciated NMVOC Emissions for the year 2010 by chemical group and by Asian regions.

34862



**Figure 8.** Monthly variations of Asian SO<sub>2</sub>, CO, PM<sub>2.5</sub>, and CO<sub>2</sub> emissions by sector for the year 2010.

34863



**Figure 9.** Monthly variations of SO<sub>2</sub>, CO, PM<sub>2.5</sub>, and CO<sub>2</sub> emissions by Asian region for the year 2010.

34864

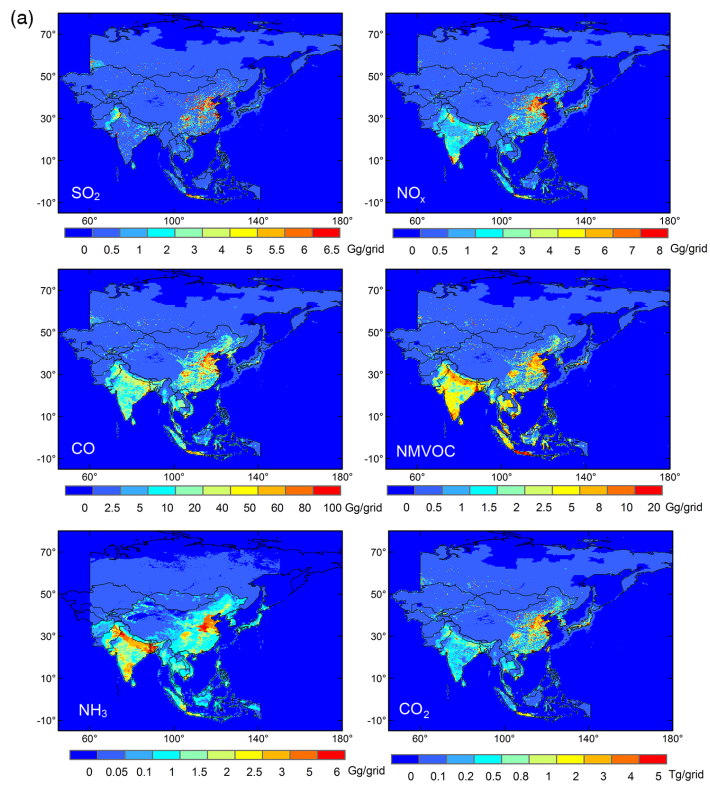


Figure 10.

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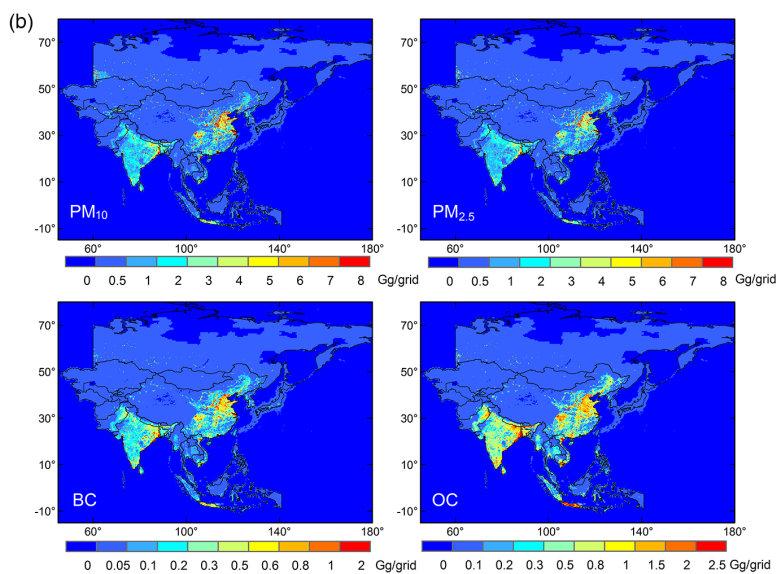
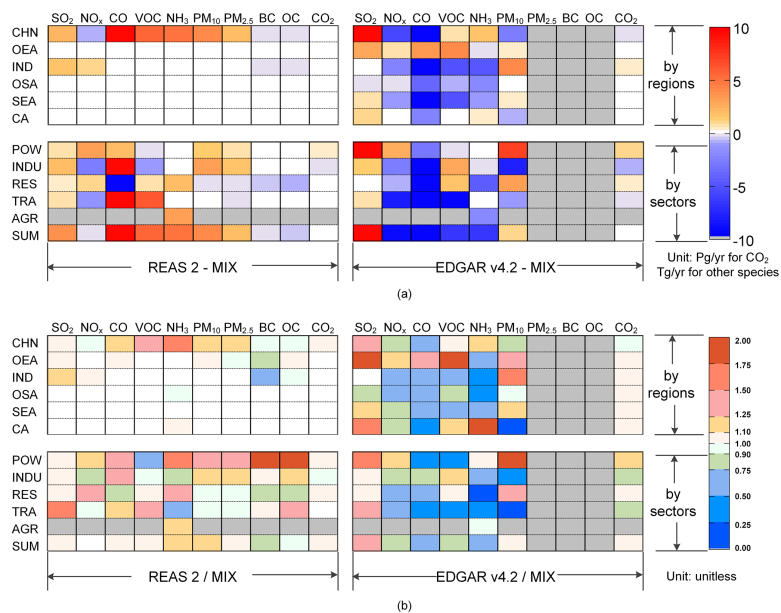


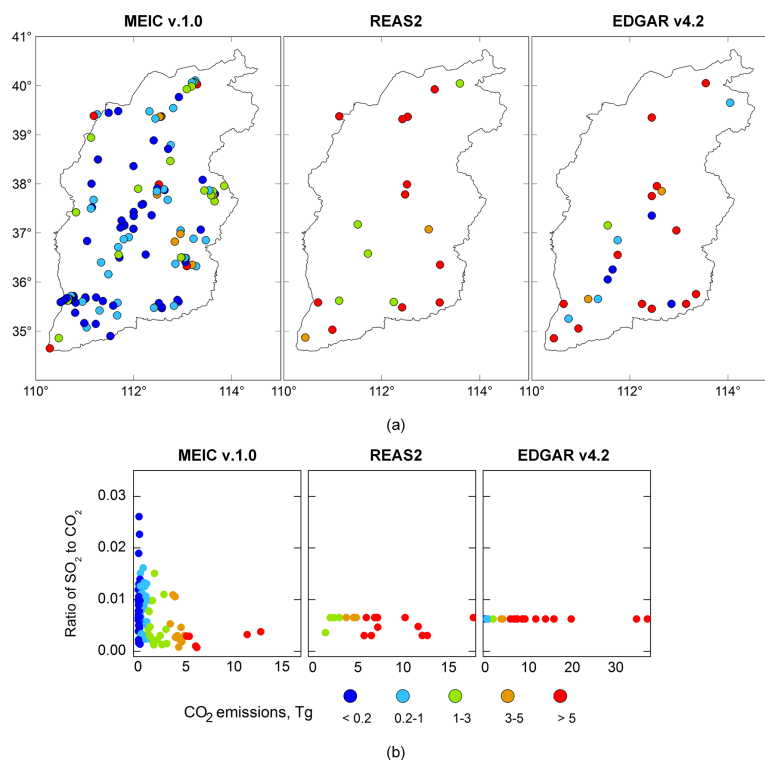
Figure 10. Grid maps for gaseous (a) and aerosol (b) species in the MIX Asian emission inventory, 2010.

34866



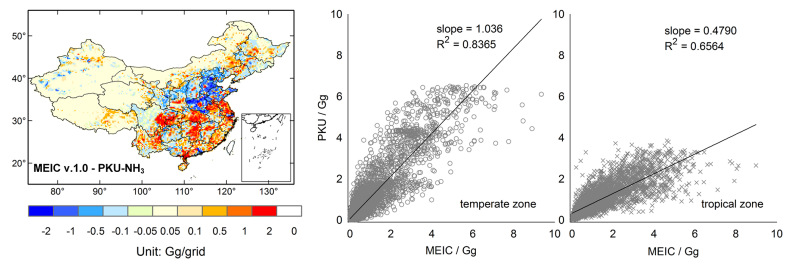
**Figure 11.** Inter-comparisons of emission estimates between MIX, REAS2 and EDGAR v4.2 by Asian regions and sectors. **(a)** Absolute differences of emission estimates. **(b)** Ratio of emission estimates. Grey grids indicate that the comparison is not available due to absence of emission estimates 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 Asia” is not included in the comparison.

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**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 CO<sub>2</sub> emissions, and **(b)** emission ratio of SO<sub>2</sub> to CO<sub>2</sub>.

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**Figure 13.** Comparisons of spatial distribution of  $\text{NH}_3$  agricultural emissions between MEIC v.1.0 and PKU- $\text{NH}_3$ . 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.