Response to referee #1

This paper presented a mosaic emission inventory of air pollutants for Asia, which is a combination of existing studies or progress in emission estimates by country and sector. Moreover the work made a comparison between selected inventories particularly for given countries and sectors. It is good to have such kind of results to support MICS-Asia and HTAP studies, as suggested by the authors. In general, the paper is well organized and clearly written. Some more explanations and discussions might be added to improve the work as follows.

Response: We thank the constructive comments given by the referee #1, which is very helpful to improve the manuscript. Our responses to each specific comment are presented below.

1. Methodology section. The reasons of inventory choice should be discussed. There are obvious overlaps in regions and species between current inventories, while the strategy of inventory choice was not sufficiently described. The readers would then question why the emissions of some species/regions were from a given inventory while the rest were from another. It would be clearer if the authors could present their preference when developing the mosaic MIX inventory.

Response: The following paragraph was added to Sect. 2.1 of the revised manuscript to indicate the hierarchy of the datasets.

"We then selected different emission datasets for various species for each country by the following hierarchy. REAS2 was used as the default where local emission data are absent. Emission inventories compiled by the official agencies or developed with more local information are selected to override REAS2, which include MEIC for mainland China, ANL-India for India, and CAPSS for the Republic of Korea. Detailed information and advantages of these inventories are presented in Sect. 2.2. As only a few species (SO_2, BC, OC, and power plant NO_x) were available from ANL-India, REAS2 was used to supplement the missing species. A mosaic process was then used to combine ANL-India and REAS2 into a single dataset for India emissions. It is worth noting that the REAS2 have incorporated local inventories for Japan and Taiwan, which are subsequently adopted in MIX for these two regions. PKU-NH_3 was further used to replace MEIC emissions for NH_3 over China, given that PKU-NH_3 was developed with a process-based model that represented the spatio-temporal variations in NH_3 emissions."

2. Section 3.2. It would be more interesting if the inter-annual trends in emissions could be analyzed by sector and species for countries other than China or India. It is well known that China started to conduct more and more stringent measures to control emissions since 2005, while such information is lacking or not well provided for other Asian countries. Moreover, the driving forces or reasons for the inter-annual trends should also be provided.

Response: In Sect. 3.2 of the revised manuscript, we added more discussions on inter-annual trend in emissions for different Asian regions.

3. For comparison section (Section 4), I understand it might be difficult to compare the detailed emission factors between MIX and EDGAR, but is it possible to make a more detailed comparison between MIX and REAS 2, for sectors/regions with different estimates in the two inventories?

Response: The estimates in MIX and REAS2 are only different for China, India, and Republic of Korea, where local emission inventories are incorporated to replace REAS2. MIX and REAS2 are same for other regions. Detailed comparisons for China and India between MIX and REAS2 are presented in the Sect 4.2 and 4.3 respectively.

4. Small issue: lines 22-24, P34833. Besides penetration, the removal efficiency that is also crucial for SO_2 estimates was assumed poorer than expected before 2010. Would that weaken the discussion here? I suggest a detailed quantitative comparison and analysis here for SO2 emission estimate.

Response: We have revised the statement as follows: "EDGAR's estimates for SO_2 emissions from power plants are 60% higher than estimates in MIX. For China, 70% of power generation capacities were equipped with FGD and the average SO_2 removal efficiency was 78% (Liu et al., 2015). The high estimates in EDGAR v4.2 most likely due to underestimation of FGD penetration or SO_2 removal efficiencies of FGD (Kurokawa et al., 2013)."

Response to referee #2

The paper documents an important emissions dataset MIX, which consists of monthly Asian gridmaps of air pollutant and aerosol emissions for 2008 and 2010, which are and will be used in international collaborations under the MICS-Asia and the HTAP Task Force. In view of the latter, this paper would be appropriate for the ACP special issue on Global and regional assessment of intercontinental transport of air pollution: results from HTAP, AQMEII and MICS. The paper does not go beyond a standard inter- comparison of emissions datasets and misses a section discussing uncertainties and border inconsistencies by compiling this mosaic of gridmaps, addressing the closure of mass balance for the aerosols and the NMVOC species per grid cell. It addresses changes in emissions from 2006 to 2010, which is an important period of increasing emissions in the Asian countries with emerging economy. However, it is not clear why then the MIX dataset is not completely covering 3 years 2006, 2008 and 2010.

Response: We appreciate the careful and extensive review given by the referee #2, which is crucial for improving the manuscript. In the revised manuscript, we added a new section entitled "Uncertainties and limitations" to discuss the uncertainties of the MIX inventory, including an overall qualitative discussion of uncertainties, issue of border inconsistencies, and mass balance closure for aerosols. The MIX dataset was developed to fulfill the needs of model simulations for the MICS-Asia and HTAP activities, in which both use 2008 and 2010 as base years. This is the main reason why the gridded data only covers 2008 and 2010 and we have clarified this in the revised manuscript. We agree that changes in emissions from 2006 to 2010 over Asia are of broad interests to the community. In this case, the magnitudes of emissions in 2006 were also collected and presented to support the analyses on emission trends and driving forces. Given that both MICS-Asia and HTAP community will not run the models for the year 2006, we feel that developing an additional gridded dataset for 2006 is less important for this study, especially considering that developing bottom-up emission inventory is always time consuming. We are now working on the more recent years, which might be more important for the community. In the revised manuscript, we further emphasized the purposed of the MIX inventory (in Sect. 2.1) and identified the limited coverage on time period as one of the limitations of current version of the MIX inventory (in Sect. 5).

Detailed responses to specific comments are provided below.

<u>*General Comments:*</u>

The documentation of the dataset could be considerably improved by: 1) Indicating a hierarchy of the datasets used for the compilation of the MIX dataset for the different countries and regions. 2) Giving an overview of the subsectors covered in the 5 source categories for each of the datasets used. 3) Giving a full documentation of the seasonality.

References for the monthly profiles used are missing 4) Giving a full documentation of the spatial distribution. References for the geo-spatial proxy datasets are missing, except for power plants.

Response: We thank the referee's comments on the improvement of the data documentation. The detailed responses to each comment are presented below.

1) **Hierarch of the datasets.** The following paragraph was added to Sect. 2.1 of the revised manuscript to indicate the hierarchy of the datasets.

"We then selected different emission datasets for various species for each country by the following hierarchy. REAS2 was used as the default where local emission data are absent. Emission inventories compiled by the official agencies or developed with more local information are selected to override REAS2, which include MEIC for mainland China, ANL-India for India, and CAPSS for the Republic of Korea. Detailed information and advantages of these inventories are presented in Sect. 2.2. As only a few species (SO_2, BC, OC, and power plant NO_x) were available from ANL-India, REAS2 was used to supplement the missing species. A mosaic process was then used to combine ANL-India and REAS2 into a single dataset for India emissions. It is worth noting that the REAS2 have incorporated local inventories for Japan and Taiwan, which are subsequently adopted in MIX for these two regions. PKU-NH_3 was further used to replace MEIC emissions for NH_3 over China, given that PKU-NH_3 was developed with a process-based model that represented the spatio-temporal variations in NH_3 emissions."

2) **Definition of subsectors.** In the supplement of the revised manuscript, a cross-walk table was provided with mapping information between subsectors in each regional inventory and the five aggregated sectors in the MIX inventory. In Sect. 2.1 of the revised manuscript, we added a note to identify the exclusion of specific subsectors from the MIX inventory. We hope the additional information may help the users to better understand the dataset.

3) **Seasonality.** When compiling the MIX inventory, we used monthly emissions from each regional emission inventory directly. In the revised manuscript, we added a subsection (Sect. 2.5) to briefly document the monthly profiles used in each component emission inventory. As the seasonality of emissions in the MIX inventory were taken from different regional inventories which have been documented previously, we provided corresponding references to those regional inventories instead of repeating the same information in this manuscript. A summary table of monthly profiles was also provided in the supplement of the revised manuscript. It should be noted that for some sub-sectors, the data sources of monthly profiles were not specified in the corresponding references.

4) **Spatial proxies.** We used gridded emissions from each regional emission inventory to compile the gridmaps of emissions. In this case, no spatial proxies were involved in

developing the MIX inventory. In the revised manuscript, we added a subsection (Sect. 2.6) to briefly document the spatial proxies used in each component emission inventory. Similarly, the spatial proxies used in different regional inventories have been documented in literatures and were not repeated here. But references to those regional inventories were provided in the revised manuscript. A summary table of spatial proxies was also provided in the supplement of the revised manuscript. It should be noted that for some sub-sectors, the data sources of spatial proxies were not specified in the corresponding references.

The structure of the paper could be improved by: 1) Explaining the different source categories (with emissions subsector-specification) in the methodology subsection 2.1 and then including a cross walk matrix of subsectors included in each of the different dataset components at the end of section 2. 2) Moving the subsection 3.4 and 3.5 on seasonality and gridding from the section 3 Results more upfront, documenting where the geo-spatial proxy and monthly profiles are coming from per subsector. 3) Discussing the aerosols and NMVOC speciation in more detail in separate section, following upon the Results section 3. That would allow to address also the consistency issues and issues with the closure of mass balance per grid cell, which is not trivial for a mosaic inventory.

Response: We thank the referee's comments on the improvement of the paper structure. The detailed responses to each comment are presented below.

1) **Explanation of source categories.** As suggested, we added a paragraph in the Sect. 2.1 of the revised manuscript, to identify the exclusion of specific subsectors from the MIX inventory. We believe may help the users to better understand the dataset. In the supplement of the revised manuscript, we also added a cross-walk table with mapping information between subsectors in each regional inventory and the five aggregated sectors in the MIX inventory.

2) **Documentation of seasonality and gridding.** In the revised manuscript, we added two subsections (Sect. 2.5 and 2.6) in the Methods section to briefly document the monthly profiles and spatial proxies respectively. We also added tables (in supplement) with full references of monthly profiles and spatial proxies used in each regional emission inventory. We prefer to keep current Sect. 3.4 and Sect. 3.5 because they provided analyses on the seasonality and spatial distributions of emissions, which may be better presented in the Results section rather than in the Methods section.

3) **Speciation of aerosol and NMVOC.** In the revised manuscript, we include a paragraph in the newly added section of "Uncertainties and limitations" to discuss the uncertainties induced from mass balance of aerosols. In the MIX inventory, speciated NMVOC emissions over the whole Asia were processed from total NMVOC emissions of each regional inventory by using a uniform, explicit species mapping framework developed by Li et al. (2014). In this case, no mass balance issue was involved.

The content of the paper could be enriched by: 1) Discussing separately the CO_2 emissions from MIX, REAS2, EDGARv4.2 and also using the national inventories reported to UNFCCC. 2) Elaborating more on the inter-comparison between Asian countries. How do the emission factors (per unit of activity) vary amongst the different countries of groups of countries? Which countries have similar per capita emissions for certain (sub)-sectors? 3) Elaborating the trend discussion using also the comparison with satellite data.

Response: We thank the referee's comments on the improvement of the content.

1) **CO_2 emissions.** We added a paragraph in the Sect. 4.1 of the revised manuscript to compare CO_2 emission estimates in different emission inventories. However, comparing CO_2 emissions with UNFCCC inventory is not feasible because the most recent year reported to UNFCCC is 2005 for Asian countries in non-Annex I Parties.

2) **Emissions per capita.** In the Sect. 3.1 of the revised manuscript, we compared per capita emissions for each country by sector and by species for the year 2010. Emissions are ranked by GDP per capita of each country. The correlations between emission intensity (per capita emissions) and economic development (GDP per capita) at country level are not always significant because emission intensities are affected by not only economic level but also by other factors such as industrial structure and dominant fuel type. Nevertheless, the changes in emission intensities in general follow the pattern of Kuznets curve for most species except NH_3, BC, and OC.

3) **Trend comparison with satellite data.** As suggested, comparison with satellite-based trend was added in the Sect. 3.2 of the revised manuscript.

<u>Specific comments</u>

1. Topic: MIX: a mosaic Asian anthropogenic emission inventory for the MICS-Asia and the HTAP project → I propose to rather talk about international collaborations under MICS-Asia and HTAP (HTAP is a task force, not really a project).

Response: We changed the title to "MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP".

2. Abstract:

a) P34815-Line1: "An anthropogenic emission inventory" → "The MIX inventory"

Response: Revised as suggested.

b) P34815-Line 3: "Task Force on Hemispheric Transport of Air Pollution (TF HTAP) projects" → delete "projects"

Response: Revised as suggested.

c) P34815-Line 6: "30 countries and regions in Asia" → Please put already here in a footnote the list of countries/ regions included.

Response: Usually footnote is not used in the abstract because abstract needs to be achieved separately. Instead, we added the list of countries in the main text (Table 1).

d) P34815-Line 14: "We also estimated Asian emissions in 2006 using the same methodology of MIX." → Why can 2006 not be full part of the dataset?

Response: The MIX dataset was developed to fulfill the needs of model simulations for the MICS-Asia and HTAP activities, in which both use 2008 and 2010 as base years. This is the main reason why the gridded data only covers 2008 and 2010 and we have clarified this in the revised manuscript. We agree that changes in emissions from 2006 to 2010 over Asia are of broad interests to the community. In this case, the magnitudes of emissions in 2006 were also collected and presented to support the analyses on emission trends and driven forces. Given that both MICS-Asia and HTAP community will not run the models for the year 2006, we feel that developing an additional gridded dataset for 2006 is less important for this study, especially considering that developing bottom-up emission inventory is always time consuming. We are now working on the more recent years, which might be more important for the community. In the revised manuscript, we further emphasized the purposed of the MIX inventory (in Sect. 1) and identified the limited coverage on time period as one of the limitations of current version of the MIX inventory (in Sect. 5).

e) P34815-Line 15: "The relative change rates of Asian emissions for the period of 2006–2010 are estimated as follows: -8.0 % for SO_2 , +19 % for NO_x , +4 % for CO," → Why only for SO_2 accurate to the first decimal behind the comma and not for all other substances (in particular for CO_2, I would expect a more accurate specification.)

Response: We unify the specification for all species to the first decimal behind the comma as SO_2.

f) P34815-Line 18: "Model-ready speciated NMVOC emissions for SAPRC-99 and CB05 mechanisms were developed" → Is it needed to specify these mechanisms already here in the abstract?

Response: We feel that it is an important message for modelers because these are actual emissions used in the chemical transport models (CTMs). One unique feature of the MIX inventory is that we provided speciated NMVOC emissions for the two widely used chemical mechanisms (SAPRC-99 and CB05) in the CTMs. We prefer to keep this in the abstract.

g) P34815-Line 10: "Monthly gridded emissions at a spatial resolution of 0.25x0.255are developed" → from the meic website monthly gridmaps are not available. we can access: - MIX v1.1 emissions by regions and sectors: xls file with total emissions by country/region for each pollutant and sector MIX v1.1 gridded emissions for each pollutant only two files are available e.g. for SO2 we can download only the following files "MICS_Asia_SO2_2008_0.25x0.25.nc and MICS_Asia_SO2_2010_0.25x0.25.nc " Monthly *gridmaps seem not to be available*

Response: The monthly gridded emissions are available in the NetCDF file. Users can extract the three-dimensional emissions data (lon × lat × month) by species, sectors and years from those .nc files.

3. Section 1 Introduction

a) P34815-Line 26: "Wang et al., 2008" → Please include also some more recent publications, such as Kulkarni et al. (2014)Atmos. Chem. Phys., 15, 1683-1705, 2015, http://www.atmos-chem-phys.net/15/1683/2015/, doi:10.5194/acp-15-1683-2015

Response: Here we refer to publications by MICS-Asia Phase I and Phase II, which was finished in 2008 or before. The paper suggested by the referee is not relevant to the MICS-Asia project.

b) P34817-Line 24: "All of these emission data were harmonized and processed to 0.5x0.5 resolution" → Please explain how you then go to 0.25deg x 0.25deg.

Response: Here we are discussing about the INTEX-B emission inventory, which is not used for the development of MIX. This sentence was removed from the revised manuscript to avoid misunderstanding.

c) P34818-Line4: "a more complete and state-of-the-art understanding of anthropogenic emissions over Asia with better estimates from local inventories" → best estimates

Response: Revised as suggested.

d) P34818-Line5: "(2) a reference dataset with moderate accuracy and resolution that can support both scientific research and mitigation policymaking," → since one of the purposes of the MICS-Asia (phase III) study is "to conduct further inter-comparisons of atmospheric modeling for Asia and analyze the disagreement of model output and relative uncertainties", can you provide some insights about emission uncertainties? or how is your study improving actual knowledge of emission estimate uncertainties? you might think to develop this discussion in section 4.

Response: In the revised manuscript, we added a new section entitled "Uncertainties and limitations" to discuss the uncertainties of the MIX inventory, including an overall qualitative discussion of uncertainties, issue of border inconsistencies, and mass balance closure for aerosols.

e) P34818-Line 14: "The MIX emission data for the years 2008 and 2010 are then incorporated into the HTAP v2.2 global emission inventory" → what is the final purpose of the MIX inventory? is it to develop and continuously maintain and update this inventory collecting the best available emission estimates from Asia or was it just an exercise for the years 2008 and 2010? it would be great if such estimates will be provided also for future years.

Response: We expect this is not just an exercise but a long-term international collaboration. Actually we have been talking with the EGDAR group to discuss the possibilities of more interactions between regional and global efforts.

P34818-Line 19: "The domain of MIX covers 30 countries and regions"→ Please give here the full list of countries and regions with name (e.g. Russia - Asian part defined by ...).

Response: We added a note to the full list of countries and regions.

f) P34818-Line 23: "including both gaseous species and aerosol species:" → delete the first "species"

Response: Revised.

g) P34818-Line 29: "NMVOC emissions are speciated into model-ready inputs for two chemical mechanisms" → Please specify here which groups of species are defined.

Response: The chemical mechanisms are developed by lumping individual NMVOC species based on similarities in chemical structure or reactivity, to characterize the atmospheric chemical reactions in the chemical transport models (Li et al., 2014). Descriptions of the SAPRC-99 and CB05 species are provided in the Tables S1-S2 of the revised manuscript.

h) P34819-Line 3: "The key elements of the MIX inventory are summarized in Table 1." → replace "elements" with "features"

Response: Revised as suggested.

4. Section2: compilation of the MIX emission inventory

2.2.1 REAS2

a) P34819-Line15: "Five emission inventories are selected and incorporated into the mosaic inventory, as listed in the following:" → Please provide as well which hierarchical order you used. When a regions is covered by more datasets, which one did you use? E.g. for the NH_3 of PKU, is this used at highest order, only for China, only for agriculture or also other regions and other sectors?

Response: We selected different emission datasets for various species for each country by the following hierarchy. REAS2 was used as the default where local emission data are absent. Emission inventories compiled by the official agencies or developed with more local information are selected to override REAS2, which include MEIC for mainland China, ANL-India for India, and CAPSS for the Republic of Korea. Detailed information and advantages of these inventories are presented in Sect. 2.2. As only a few species (SO_2, BC, OC, and power plant NO_x) were available from ANL-India, REAS2 was used to supplement the missing species. A mosaic process was then used to combine ANL-India and REAS2 into a single dataset for India emissions. It is worth noting that the REAS2 have incorporated local inventories for Japan and Taiwan, which are subsequently adopted in MIX for these two regions. PKU-NH_3 was further used to replace MEIC emissions for NH_3 over China, given that PKU-NH_3 was developed with a process-based model that represented the spatio-temporal variations in NH_3 emissions. The above clarifications have been added in the revised manuscript.

b) P34821-Line13: "We aggregated the 11 REAS2 sectors to five sectors provided in the MIX inventory." → Please indicate which (sub)sectors are NOT included in REAS (e.g. fuel transformation of charcoal is not included, certain agricultural sectors neither, what about the biomass burning, ...)

Response: Emissions from open-biomass burning, aviation, and international shipping were excluded from the REAS2 before incorporating into MIX. We have clarified this in the revised manuscript.

c) P34821-Line 18: "while emissions for other sectors were processed as area sources"→ it should be "areal sources"

Response: Revised as suggested.

d) P34821-Line19: "gridded at 0.25x0.258resolution using maps of rural, urban and total populations and road networks." → please specify the source of these data. (REAS). Could you please specify what proxy data were used to spatially distribute emissions by sector? was industry considered as areal source too?

Response: In the revised manuscript, we added a subsection (Sect. 2.6) to briefly document the spatial proxies used in each component emission inventory. A summary table of spatial proxies was also provided in the supplement of the revised manuscript.

5. Section 2.2.2 MEIC

a) P34822-Line8: "Power plant emissions in MEIC were derived from the China coal-fired Power plant Emissions Database (CPED)"→ are these data public available? in recent literature works, it is often criticized that the CARMA database collecting power plants information is not complete (especially for China). since the CPED database is fully documented in a specific publication, would you make this data available (maybe with some limitations etc.)

Response: Power plant emission data developed from the CPED database have been incorporated into the MEIC database and publicly available at 0.25×0.25 resolution from the MEIC website (www.meicmodel.org).

b) P34822-Line 14: "For the on-road transportation sector," → What about the non-road transportation sectors: inland waterways, domestic flights, off-road transport?

Response: Non-road transportation sector includes agricultural machinery, construction machinery, rural vehicles, and inland shipping, which are all processed areal sources.

c) P34822-Line 23: "emissions of individual NMVOC species were calculated for each source category by splitting the total NMVOC emissions with corresponding source profiles."

→ *please mention here or in section 2.4 the list of NMVOC species you are including in your work.*

Response: Using the explicit profile assignment approach developed in Li et al. (2014), we calculated NMVOC emissions for more than 700 individual chemical species, and then aggregated emissions of individual species to lumped species of two chemical mechanisms. We feel that it is difficult (and not necessary) to present the long list in this paper. Readers can refer to Li et al. (2014) for detailed information of individual NMVOC species.

d) P34823-Line 3: "Emissions were aggregated to four MIX sectors: power, industry, residential, and transportation" → Where is the waste sector included?

Response: Waste sector was aggregated to the residential sector.

e) P34823-Line 4: "Agriculture NH_3 emissions in MEIC were replaced by PKU-NH3, which will be discussed in the next section"→ Does MEIC include the NH_3 of non-agricultural sectors (e.g. from catalysts in road transport)?

Response: MEIC only includes NH_3 emissions for agriculture sector. Actually the PKU-NH_3 includes both agriculture and non-agricultural emissions for NH_3 and we incorporated these emissions in the MIX inventory. We have removed "agriculture" from the sentence.

6. Section 2.2.3 PKU-NH_3 for China

a) P34823-Line 20: "Open biomass burning was considered as a natural emission source and excluded in the MIX inventory." → open biomass burning cannot be fully considered as natural emission source. you should reformulate this sentence: e.g. open biomass burning emissions were excluded from the MIX inventory aggregation...is it because you needed to rely on a different database like GFED etc.?

Response: Yes, MICS-Asia III project decided to use GFED for biomass burning hence we removed open biomass burning emissions from all regional emission inventories. We revised the sentence as follows: "Open biomass burning was excluded from the MIX inventory aggregation since the MICS-Asia III project uses GFED dataset for biomass burning".

b) P34823-Line 24: "In the MIX inventory, 2006 emissions from PKU-NH3 are used for both 2008 and 2010" → When extrapolating in time, why also not extrapolating in space? Why is it not used for neighbouring countries?

Response: PKU-NH_3 is developed based on a process-based model by parameterizing NH_3 emissions with ambient temperature, fertilization method, application rate, soil acidity, fertilizer type, and etc. Extrapolating the methodology in other countries needs much more efforts, which seems exceed the scope of this work.

7. Section 2.2.4 ANL emission inventories for India

a) P34824-Line4: "ANL-India used a technology-based methodology to estimate SO2, BC, and OC emissions in India" → What for the other substances, NOx, NMVOC, CO, NH3?

What is used there?

Response: REAS2 is used as the default emission inventory to supplement emissions estimates that not included in the regional inventories. We further clarified this in the Sect. 2.1 of the revised manuscript.

b) P34824-Line 19: "Emissions are presented by sectors, i.e., power, industry, residential, transportation, and open biomass burning." → you should mention that open biomass burning was not included in the MIX inventory although available in the ANL database

Response: Revised as suggested.

c) P34824-Line 23: "monthly emissions by sector from ANL-India were first regridded to 0.25x0.25 and then merged with REAS2 before being implemented in MIX" → to cover all substances? to make the gapfilling? please specify what do you mean with "merged with REAS2" in this specific case.

Response: This sentence has been revised as follows: "monthly emissions by sector (excluding open biomass burning) from ANL-India were first regridded to 0.25º × 0.25º and then merged with REAS2 before being implemented in MIX to cover all species. The merge process is presented in Sect. 2.3."

8. Section 2.2.5 CAPSS inventory for the Republic of Korea

a) P34825-Line1: "We mapped emissions from 12 first-level aggregated source categories (SCC1) to five sectors in MIX." → you might think to provide these 12 levels of source categories and their aggregation to the 5 MIX sectors in the supplementary material

Response: Revised as suggested. The sector mapping table is provided in the supplement.

b) P34825-Line 6: "We derived sector-specific emission ratios between PM10 and the other aerosol components from Lei et al. (2011) and applied those ratios to estimate $PM_{2.5}$, BC and OC emissions" → how are CO_2 emissions estimated? using REAS2?

Response: The CO_2 emissions were obtained from CAPSS. We have clarified this in the revised manuscript.

c) P34825-Line 13: "In the MIX inventory, we assume no monthly variation in emissions in the Republic of Korea." → why? cannot you use the monthly profile for each source of another country like Japan or China?

Response: During the development of the MIX inventory, we assume no monthly variation in emissions when monthly profiles are absent from the regional emission inventories. As shown in Table S3, this not only for the case of the Republic of Korea but also for some sub-sectors for REAS2, MEIC, and PKU-NH_3. We acknowledge that it is not the best case but applying monthly profiles to all these sub-sectors will need much more efforts than what we can afford for this work.

9. Section 2.3 Mosaic of Indian emission inventory → this section could be a sub-section of

2.2.4 dealing only with ANL data

Response: Sect. 2.2 introduces the candidate emission inventories and the Section 2.3 document the mosaic process of the ANL-India inventory and the REAS2 inventory for India. For this case, we feel that it's better to keep Sect. 2.3 separately.

a) P34825-Line 25: "In this work, we first generated the spatial distribution of fuel consumption by type at 0.25x0.25 resolution by aggregating unit-level information in ANL-India, we then used these spatial proxies to reallocate total power plant emissions of CO, NMVOC, $PM_{2.5}$, PM_{10}, and CO_2 in REAS2 by fuel type." → Please clarify this procedure because it is not clear what you have done with the distribution of the fuel consumption and how did you check the consistency with the CARMA and WEPP databases. moreover it is not clear why did you apply this new proxy for power plants only for a subset of pollutants. would have not been possible to have the same spatial distribution for the same source for all pollutants?

Response: 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 could introduce inconsistency due to a mismatch of unit information in the two databases. In this work, we directly used ANL-India for SO_2, NO_x, BC, and OC emissions and used REAS for CO, NMVOC, $PM_{2.5}$, PM_{10}, and CO_2 but redistributed the total magnitudes of REAS2 power plant emissions by using the spatial distribution of power plants in the ANL-India inventory. We generated the spatial proxies of fuel consumption for each fuel type (coal, oil and gas) at 0.25×0.25 degree by aggregating fuel consumptions of each unit in the ANL-India inventory. We then applied the spatial proxy to the REAS2 estimates by fuel type for species that not included in ANL-India. We have clarified this in the revised manuscript.

10. Section 2.4 NMVOC speciation of the MIX inventory

a) P34826-Line 11: formula of EVOC (I,k,m) → Can the conversion factor from species j to m be assumed independent of the source category i and independent of the region k in general?

Response: The conversion factor was developed based on the lumping mechanism for various chemical mechanisms (e.g., SAPRC-99, CB05), which is dependent on the chemical species and mechanisms, and independent of the source categories and regions (Carter et al., 2013).

b) P34826-Line 12: "m is species type in CB05 or SAPRC-99 mechanisms" → please list these species.

Response: Descriptions of the SAPRC-99 and CB05 species are provided in the Tables S1-S2 of the revised manuscript.

c) P34827-Line 2 → Except for the MEIC inventory, the data source for the CO_2 inventory is not addressed in the subsections above. Where is it coming from? From the national *inventory reports to UNFCCC?*

Response: The CO_2 emissions of MIX were developed by mosaic of estimates from MEIC, CAPSS and REAS2 inventories. We have further clarified this in Sect. 2 of the revised manuscript.

11. Section 3.1 Asian anthropogenic emissions in 2010

a) P34828-Line 9: "28,33% and 7%" → 28%

Response: Revised as suggested.

b) P34828-Line 11: "reflecting the better emission control" → delete "the"

Response: Revised as suggested.

c) P34829-Line3: "contributing 59 % of the total SO_2 emissions" → insert "Indian" between "total" and "SO_2 emissions"

Response: Revised as suggested.

d) P34929-Line3: "The SO_2/CO_2 emission ratio in Indian power plants is significantly higher than that of China" → The ratios of air pollutants over CO_2 are of general interest. Please quantify these per country/ region and inter-compare these ratios for the different regions.

Response: In Sect. 3.1 of the revised manuscript, we compared the emission ratios of CO/CO_2 and SO_2/CO_2 to inform emission characteristics. SO_2/CO_2 ratio was used as an indicator of coal combustion and emission control levels (Li et al., 2007), and ratios of CO/CO_2 were used to inform combustion efficiency (Wang et al., 2010).

12. Section 3.2 Changes of Asian emissions from 2006 to 2010

a) P34829-Line 19: "the relatively flat or even decreasing emission trends in many species indicates" → Please specify per substance and region (eventually in a table)

Response: Emission ratios of 2010 to 2006 by country were presented in Table 5.

b) P34830-Line 4: "NMVOC emissions increased in all Asian regions except Other East Asia" → Please specify which countries the Other East Asia region includes

Response: The definition of each region could be found in Table 3 and Table 5.

c) P34830-Line 15: "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)." → Please elaborate on this with quantitative results

Response: We have revised the statement as follows: The downward trend of CO emissions over China in recent years 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). The decreasing rate of CO emissions over China is estimated to be -1.2% yr^{-1} from 2006 to 2010 in in the MIX inventory, consistent with the rates observed by multiple satellites in range of -1.0% yr^{-1} to -3.1% yr^{-1} during 2000-2012 (Table 6).

13. Section 3.3 speciated NMVOC emissions → solvent use is known to significantly contribute to NMVOC emissions especially in Asian regions. can you provide some details about this topic?

Response: Solvent use emissions are estimated to 12.7 Tg (19.0% of total) over Asia in 2010. Among different regions, China is the largest contributor (6.5 Tg) to solvent use emissions, which mainly from industrial paints, pesticide use, printing, and glue use.

a) P34830-Line 19: "Figure 7 presents 2010 Asian NMVOC emissions of different chemical groups" → Please specify how you grouped the substances (alcohols, ethane, propane, butanes, pentanes, hexanes and higher ethene, propene, ethyne, isoprene, terpenes, other alkenes and alkynes, benzene, toluene, xylene, trimethyl benzenes, other aromatics, esters, ethers, chlorinated HC, methanal (CH2O), other alkanals, ketones, acids, other VOC).

Response: We have added the information in the caption of the Fig. 7.

b) P34830-Line 30: "Over Asia, the industrial sector is the major source of emissions of alkanes and aromatics" → what type of industries?

Response: Alkanes emissions from industrial sector are mainly contributed by gas production and distribution (19.8% of total industrial emissions), coal combustion (17.1%), and oil refinery (15.0%), and aromatics emissions are mainly contributed by architectural paint use (21.0% of total industrial emissions), other industrial paint use (16.6%), and gas production and distribution (10.6%). We have clarified this in the revised manuscript.

c) P34831-Line 1: "while the residential sector has a high contribution of OVOCs" → from biofuel use? solvent use?

Response: Biofuel use. We have clarified this in the revised manuscript.

d) P3831-Line 10: "Among different regions, China, India and Southeast Asia are the largest contributors to NMVOC emissions in Asia, with contributions varying by chemical groups." → Moreover, interestingly, for India and Other South Asia the relative share of alkenes and OVOCs are considerably higher than in the other regions. Any explanation for this? as already mentioned, it would be interesting to have some details about the type of activities emitting NMVOC (and possibly providing regional differences in emitting sources)

Response: The high emissions of alkenes in South Asia (both India and Other South Asia) are mainly from contributions of biofuel combustions and motorcycles, and OVOC emissions are dominant by biofuel combustions.

14. Section 3.4 Seasonality

a) P34831-Line 14: "As documented in Sect. 2, we used monthly emissions from each component inventory where available" → Which references are documenting the monthly *profiles used?*

Response: In the revised manuscript, we added a subsection (Sect. 2.5) to briefly document the monthly profiles used in each component emission inventory. A summary table of monthly profiles was also provided in the supplement of the revised manuscript. It should be noted that for some sub-sectors, the data sources of monthly profiles were not specified in the corresponding references.

b) P34932-Line 8: "Winter $PM_{2.5}$ emissions in China are higher than other regions, representing large emissions from solid fuel use in residential homes" → why do we expect larger PM emission from the residential sector in China during wintertime compared to other asian countries? I guess in India or other countries residential emissions are even less regulated than the chinese ones...if it is associated with coal combustion in the residential sector, we should see the same effect in SO_2 emissions (while we see only very small difference between SO_2 in china from other countries). please try to give more explanations.

Response: This is because residential emissions contributed to 38.8% of the total primary $PM_{2.5}$ emissions over China, but only contributed to 12.2% of total SO_2 emissions. SO_2 emissions are mainly contributed by power and industry sector of which monthly variations are relatively small. In China, residential emissions in winter are much higher in other seasons due to heating. But in India, no heating is needed hence the monthly variations in residential emissions are very small.

15. Section 3.5 Gridded emissions

a) P34832-Line 15: "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." → These are only a few proxy datasets. Which geo-spatial proxy datasets are used for the transport sector, industry sector, residential sector? What about the possible inconsistency at borders because of the use of different proxy datasets?

5) **Response:** We used gridded emissions from each regional emission inventory to compile the gridmaps of emissions. In the revised manuscript, we added a subsection (Sect. 2.6) to briefly document the spatial proxies used in each component emission inventory. A summary table of spatial proxies was also provided in the supplement of the revised manuscript.

b) P34832-Line 15: "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." → so, how these emissions are distributed? please provide more information about the gridding procedure and the proxy data you used to spatially distribute emissions.

Response: Please see response above.

16. Section 4.1 MIX, REAS and EDGAR v4.2 over Asia

a) P34833-Line4: "the two widely used inventories" → delete "the"

Response: Revised as suggested.

b) P34833-Line5: "to highlight the new findings from the mosaic inventory and identify the potential sources of uncertainties." → unfortunately, you do not make here any uncertainty assessment, but you identify possible factors influencing emission calculations (e.g. use of different emission factors or abatement measures). using all your expertise and knowledge about Asian emissions, it would be great if you could try to constrain a bit the uncertainty of emission estimates in Asia (e.g. provide an uncertainty value for each pollutant and sector for macro-regions in Asia, or give a range of emissions for each region (min-max), or provide a number for uncertainty of emission factors, activity data, spatial distribution etc.)

Response: In the revised manuscript, we added a new section entitled "Uncertainties and limitations" to discuss the uncertainties of the MIX inventory, including an overall qualitative discussion of uncertainties, issue of border inconsistencies, and mass balance closure for aerosols.

c) P34833-Line7: "EDGAR"→ Please consistently refer to EDGARv4.2, in order to avoid confusion with other EDGAR datasets.

Response: Revised as suggested.

d) P34833-Line 14: "The differences between REAS and MIX over China and India will be discussed in the following section" → Make sure that you use the same "measure unit" for characterising something as "large discrepancy" or "good agreement", independently of the datasets you are comparing!!!

Response: We have carefully reworded the statement throughout the manuscript.

e) P34833-Line 16: "Larger discrepancies are observed between MIX and EDGAR" → How did you compare the EDGARv4.2 for the full MIX region with a part of Russia. How did you calculate this with the Russian total? Moreover, it would be more useful to compare the emissions per country!

Response: Russian emissions were not included in the comparisons between MIX, REAS2 and EDGARv4.2. As the MIX inventory contains emissions for 29 countries/regions and 10 species, we feel that compare emissions for each country in the text will make the paper difficult to read considering that the manuscript is already very lengthy.

f) P34833-Line 17: "20,33,11,27%" → 20%, 33%, 11%, 27%

Response: Revised as suggested.

g) P34834-Line 5: "the huge discrepancy by sector could only be attributed to differences in emission factors." → and abatement measures

Response: Revised as suggested.

h) P34834-Line 11: "The differences are mainly from high emission estimates of wastewater treatment sources in REAS," → Please refer to REAS2 and do not abbreviate to

REAS1 in order to avoid confusion about the version used.

Response: Revised as suggested.

17. Section 4.2 China → Please, before starting a detailed comparison for China and India, please use also other dataset, scientific literature for the comparison with other inventories. I suggest for SO_2 to look into Smith et al., ACP 2011 or Klimont, Smith Cofala, GRL, 2013)

Response: A comprehensive inter-comparison among different emission inventories over Asia was conducted by Kurokawa et al. (2013), including the literatures suggested by the referee. We feel that it's not necessary to repeat this in our paper. We added a note to Kurokawa et al. (2013) at the beginning of Sect. 4.1 of the revised manuscript.

a) P34834-Line 24: "(differences within 30% for NOx, and 10% for SO_2 and CO_2, respectively" → Please be consistent: when comparing MIX with EDGARv4.2 for NO_x and seeing a 20% difference, you characterised this as "large discrepancy", but when comparing MIX with REAS2 and seeing a 30% difference, you see a good agreement???

Response: We have revised the statement as follows: "MIX and REAS2 showed good agreements on power plant emissions in China for SO_2 and CO_2 (3% differences for SO_2, and 8% for CO_2) in 2008, implying similar estimates in energy consumption and emission factors in two inventories. Compared to MIX, REAS2 estimates lower emissions of NO_x, PM_{10}, $PM_{2.5}$ by more than 20%, mainly due to the differences in the emission factors used in compiling China's emissions."

b) P34835-Line1: "REAS2 included 380 power plants for China, 84 % lower than 2411 plants in MIX" → This % is not very meaningful. I suggest "REAS2 included 280 PP for China, which is much less than the 2411 PP in MIX and the yyy PP in EDGARv4.2, but these 280PP of REAS2 and yyy PP of EDGARv4.2 represent aaa% respectively bbb% of the power generation output accounted for with the 2411 PP in MIX.

Response: This sentence has been revised as follows: "REAS2 included 380 power plants for China, compared to the 2411 plants in MIX. While power plants in REAS2 are large ones which contributed 72% of CO_2 emissions in China."

c) P34835-Line24: "there is a tendency towards a decrease in SO_2/CO_2 emission ratio with increase of plant size (presented as CO_2 emissions)," → corresponding to higher CO_2 emissions?

Response: Revised as suggested.

d) P34835-Line25: "in accordance with the legislation that large units were required to be equipped with FGD during 2005–2010" → was the implementation of the legislation happening immediately or there was any delay? did you consider the real time of the implementation of the legislation or just the fulfill of the mandatory objectives in time?

Response: There was a delay of the implementation of the control measures after the legislation. We extracted the actual running time of FGD for each unit from the CPED database.

e) P34835-Line28: ". EDGAR presented constant ratios for all power plants, indicating that uniform SO_2 and CO_2 emission factors are used." → The constant ratio for all power plants in a given country for a given year, indicate that (i) the emission factors are not varied within the country and (ii) the spatial distribution treats all power plants equal.

Response: Revised as suggested.

18. Section 4.2.3 Other sectors

a) P34837-Line2: "EDGAR is not compared here because references to the detailed underlying data used in EDGAR are not available" → Is this a good reason? Please consistently refer to EDGARv4.2 and do not abbreviate to EDGAR, in order to avoid confusion with other EDGAR datasets.

Response: We have removed this statement from the revised manuscript. We also change EDGAR to EDGAR v4.2 throughout the manuscript.

b) P34837-Line12: "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)" → it would be interesting to have some details about these new standards...maybe you could add a table in the supplementary material

Response: Emission standards implemented during 2005-2010 are summarized in Table S15 of the revised manuscript.

19. Section 5 Concluding remarks

a) P34839-Line9: "Gridded speciated NMVOC emissions for SAPRC-99 and CB05 mechanisms were also developed" → Is it needed to specify these mechanisms here in the concluding section?

Response: We feel that it is an important message for modelers because these are actual emissions used in the chemical transport models (CTMs). One unique feature of the MIX inventory is that we provided speciated NMVOC emissions for the two widely used chemical mechanisms (SAPRC-99 and CB05) in the CTMs. We prefer to keep this message here.

b) P34839-Line 18: "MIX has improved the accuracy of emission estimates as well as spatial and temporal distributions due to extensive inclusion of local knowledge." → This needs a separate section to quantify this. Moreover, the local knowledge might cause artificial border effects. Can you elaborate also on this?

Response: The inter-comparison between MIX and REAS2 has demonstrated the improvement of emission estimates in MIX. We have removed the statement from the revised manuscript as we agree that the quality of a bottom-up emission inventory should be evaluated by independent approaches.

In the MIX inventory, the inconsistencies are expected at the country boarder of China and India. However, low populations and emissions are observed along the border of China, reducing the impact of cross-border grids on the accuracy of emissions. Also deriving country totals from the gridded emissions is not appropriate for small countries due to the impact from cross-board grids, especially for those grids with large point source emissions (Janssens-Maenhout et al., 2015). We have added these discussions in Sect. 5 of the revised manuscript.

c) P34839-Line30: "For MIX, the inter-comparison of emissions between regions is less valid because different methodologies were used." → actually, the inter-comparison you did is using emission independent estimates, so it should give you either an uncertainty assessment or in the best case comparable emissions among different inventories. using different methodologies does not mean having different emission estimates. please modify your sentence

Response: We revised the statement as follows: "The inter-comparison between MIX and other inventories indicated that significant differences in methodology and input data were used in different emission inventories were used. Harmonizing the efforts among different regions and research groups through international collaborations could help to resolve this issue in the future."

20. Comments on Tables:

a) Table1: Summary of the MIX Asian anthropogenic emission inventory.

→this is definitely too vague. Please specify the 30 countries.

Response: Revised as suggested.

→Also this is definitely too vague and needs to be specified more accurately. You might want to use the IPCC coding (CRF numbers) to specify the sectors.

Response: We add the source matrix table in the supplement, which specify the sectors in detail.

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

→add a column with the year of data availability

Response: Revised as suggested.

→Please include here also a row with header "region" so that the geo-coverage of each of the datasets can be given.

Response: Revised as suggested.

→Please include here which sectors are included in each of the datasets. Not all datasets cover all source categories (subsector levels)

Response: Sub-sectors and source mapping matrix are provided in the supplement.

c) *Table 3: National anthropogenic emissions in the MIX emission inventory in 2010*

→ *numbers are difficult to read...think about using Tg also for other species also in the following tables*

Response: Using Tg will make numbers of BC and OC emissions hard to read. Commas were added to the numbers to make them easy to read.

d) *Table 5: Asian emissions in 2006 based on the same methodology of MIX*

→ *Please provide in full 2006, 2008 and 2010 and combine table 3 and 5*

Response: Table 3 and Table 5 are already very large tables. Merging them into one table and adding 2008 emissions would make it difficult to fit into single journal page. We prefer to keep them separated. Emissions by regions and species for the years 2008 and 2010 are provided in the MIX website.

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

→ *here you use Tg for all species, so please use it also for the former tables. please add in the table caption that emissions come from all sectors...*

Response: The caption is revised as "Inter-comparisons of total anthropogenic emissions among MIX, REAS2 and EDGAR v4.2 for 2008."

→ *Please specify what Asia covers (either in footnote or caption). Please also specify which sectors are covered.*

Response: We classify the sectors and regions included in the comparison in the footnote.

f) *Table 7: NH_3 agriculture emission estimates for China*

→ *Please include for EDGARv4.2 also 2005, 2006, 2007。*

Response: Only EDGAR v4.2 estimates for 2008 were used for comparison. MASAGE_NH_3 represents the average top-down emission estimates during 2005-2008. We add a footnote for MASAGE_NH3 to avoid possible misunderstanding.

21. *Comments on Figures:*

a) *Figure1: Domain and component of the MIX emission inventory*

→ *replace the legend title with: MIX emission inventory components*

Response: Revised as suggested.

b) *Figure3: NMVOC speciation scheme used in the MIX inventory development*

→ *The mapping table is an interesting dataset of proxies for spatial distribution. Can this mapping table at least with references be documented?*

Response: We added the reference of the mapping table (Carter et al., 2013).

→ *please add here or in the supplement the NMVOC species list*

Response: NMVOC species list was added in the supplement.

c) Figure4: Emission distributions among sectors in Asia in 2010

→ *please verify that the sum of each pie chart gives 100% (e.g. for NMVOC some % are missing, BC, etc.)*

Response: Corrected.

d) Figure5: Emissions distributions by Asian regions in 2010

→ *It is more interesting to give the sector-specific distribution per region, combining figures 4 and 5.*

Response: Combing Fig. 4 and Fig. 5 will generate 70 pie charts, which are too much for a figure. Actually the sector-specific distribution per region could be derived from Table 4.

e) Figure6: Emission changes from 2006 to 2010 by Asian regions for SO_2 *(a) and CO (b)*

→ *the left part of this graph is not very clear. you might think to replace it with a more readable figure.*

Response: In the revised manuscript, we tried to explain the message more clearly in the figure caption. We hope the referee will satisfied with the revision.

f) Figure7: Speciated NMVOC Emissions for the year 2010 by chemical group and by Asian regions.

→ *How has this unit to be interpreted? Are these 10^9 mole species per year?*

Response: For each chemical group, the unit is 10^9 mole species per year, which is added in the Figure.

g) Figure9: Monthly variations of SO_2*, CO,* $PM_{2.5}$*, and* CO_2 *emissions by Asian region for the year 2010.*

→ *Again here, it is more useful to give the monthly variation per region and sector, combining figures 8 and 9.*

Response: Combing Fig. 8 and Fig. 9 will generate too much for a figure. Monthly emissions by sector for each region were provided in the supplement information.

h) Figure10: Grid maps for gaseous (a) and aerosol (b) species in the MIX Asian emission inventory, 2010.

→ *add in each graph the y-x labels (Lat, Lon)*

Response: Lat/Lon information is presented in the *y*-*x* labels.

→ *try to use the same color scale for most of the pollutants*

Response: In Fig. 10(a), we use the same color scale for SO_2, NO_x and NH_3. In Figure 10(b), we unify the color scale for BC and OC, PM_{10} and $PM_{2.5}$. As the magnitudes of emissions are quite different for different species, using the same color scale for all species will make the figure difficult to read.

→ *change with: Tg/grid cell*

Response: Changing units to Tg/grid will make most of numbers in figure in an unreadable decimal format (like 0.0005), especially for BC and OC with small emissions on each grid.

→ *in order to make figures more comparable, please use the same color scale (e.g.BC and OC up to 2.5...in the best case all PM components up to 8)*

Response: See response above.

i) Figure11: Inter-comparisons in Asia

→ *use grey shaded area in order to avoid confusion with values lower than -10*

Response: Revised as suggested.

→ *grey shaded grids*

Response: Revised as suggested.

→ *Please specify EDGARv4.2*

Response: Revised as suggested.

→ *please add "in", so that the text is "as in Fig.5"*

Response: Revised as suggested.

→ *why Russia Asia is not included?*

Response: The Russia Asia is not included in comparisons because emissions of Asian part of Russia are not separately estimated in EDGAR v4.2.

j) Figure12: Inter-comparisons in China, power plant sector

→ *power plants location is very different from both MEIC and Edgar. why?*

Response: MIX used a high-resolution emission database for China (CPED) to derive emissions and locations of China's power plant emissions at unit level. The coordinates in CPED are obtained from official sources and crosschecked by Google Earth (Liu et al., 2015). EDGAR v4.2 developed the power plant emissions using CARMA database. CARMA used city centers as the approximate coordinates of power plants (Wheeler and Ummel, 2008). We have explained this in Sect. 4.2.1.

→ *please put the legend of CO_2 emissions in the upper part of the graph, while add a new color scale for the SO_2 to CO_2 ratios (it cannot be the same since it is unitless)*

Response: The color scale in Fig. 13(b) also represents CO_2 emissions, and the SO_2 to CO_2 ratios are shown in *y*-axis.

→ *please specify the x axis label of the bottom figure (CO_2 emissions?)*

Response: The x-axis label of the bottom panel is CO_2 emissions, which is added now.

k) Figure13: Inter-comparisons in China, NH_3 emissions

→ *what is this square?*

Response: The square represents the island part of the China territory.

→ *these 2 graphs can be overlapped in one graph using different colors for points of temperate and tropical zone.*

Response: Revised as suggested.

→ *"Provinces that included in the tropical zones are", delete "the"*

Response: Revised as suggested.

Response to referee #3

The paper provides a useful description of the MIX inventory. The paper could be enhanced by providing additional detail for the data and inventory construction methodologies.

Response: We thank the constructive comments given by the referee #3 in improving our manuscript.

In addition to the suggestions from other reviewers I suggest the following: It appears that the mosaic inventory was constructed using the five sectors: power, industry, residential, transportation, and agriculture. This should be explicitly stated.

Response: In the abstract and the Sect. 2.1 of the revised manuscript, we clarified that the MIX inventory includes five sectors: power, residential, transportation, and agriculture.

The definition of these sectors should be provided (this could be in the supplement). Some of the issues that are potentially inconsistent between inventories include the sector assignment for: auto producer industrial emissions, mobile residential and commercial emissions, and off-road mobile emissions. A mapping between the summary sectors and IPCC/NFR categories would be useful.

Response: In the supplement of the revised manuscript, we provide the sub-sector information of different inventories and the mapping table between those sub-sectors and the five sectors of the MIX inventory. A mapping table between the five sectors and IPCC/NFR categories are also provided.

Some discussion of how consistent the sector definitions are across the different inventories used in MIX would also be helpful. For example, do all the inventories define these sectors in the same manner and include all sub-sectors?

Response: We agree with the referee that inconsistencies are always existed across different emission inventories. In the Sect. 2.1 of the revised manuscript, we added a paragraph to discuss the differences of sub-sectors in the component emission inventories.

It appears that some emissions, although somewhat small, may be missing (For example there are no emissions from agriculture listed except for NH3. I would expect NOx emissions, for example.)

Response: Some specific sources are excluded from the MIX inventory as they are not estimated in all of the component emission inventories. We clarified this in the Sect. 2.1 of the revised manuscript.

Section 4.2.1, line 24 ˇ 30% is a fairly large difference for China. I'm not sure this can be classified as "good" agreement.

Response: We revise the statement as follows:

For SO_2, and CO_2, MIX and REAS2 agreed well in power plant emission estimates over China (differences within 10% for 2008), implying similar estimates in energy consumption and emission factors in the two inventories. For NO_x, PM_{10}, and $PM_{2.5}$, REAS2 estimates are lower by more than 20% compared to MIX, mainly due to the differences in the emission factors.

Table 3 should clarify that the last line in each section is the sum for that set of countries.

Response: Revised as suggested.

Table 4 is appropriate for the main paper. A similar table by country with emissions by sector should be provided in the supplement in order to more fully document the dataset.

Response: Considering the size of the table is large, we put the table into the MIX online repository and added the link to the table in the revised manuscript.

MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP

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Abstract

The MIX inventory 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) by a mosaic of up-to-date regional emission inventories. Emissions are estimated for all major anthropogenic sources in 29 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. Emissions are aggregated to five anthropogenic sectors: power, industry, residential, transportation, and agriculture. We estimate the total Asian emissions of ten species in 2010 as follows: 51.3 Tg SO_2, 52.1 Tg NO_x, 336.6 Tg CO, 67.0 Tg NMVOC (non-methane volatile organic compounds), 28.8 Tg NH_3, 31.7 Tg PM_{10}, 22.7 Tg $PM_{2.5}$, 3.5 Tg BC, 8.3 Tg OC and 17.3 Pg CO_2. 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: -8.1% for SO_2, +19.2% for NO_x, +3.9% for CO, +15.5% for NMVOC, +1.7% for NH_3, -3.4% for PM_{10}, -1.6% for $PM_{2.5}$, +5.5% for BC, +1.8% for OC and +19.9% for CO_2. 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 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 temporal 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. (2002 a, b), a Japan emission inventory from Kannari et al. (2007), and official emission inventories for the Republic of Korea and Taiwan. 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 global and regional modeling activities with the best available anthropogenic emission dataset over Asia, we develop a new Asian anthropogenic 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 best 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 MIX inventory is developed for 2008 and 2010, in accordance with base year simulations in MICS-Asia III and the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). 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 using the same methodology, partly resolving the problems of trend analysis in mosaic inventories. The gridded 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 modelling activities in 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 29 countries and regions (the full list of country/region names are listed in Table 1), 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 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 μm), $PM_{2.5}$ (particulate matter with diameter less than or equal to 2.5 μm), 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 State Air Pollution Research Center 1999 version, Carter, 2000) (see Table S1 and Table S2). Monthly emissions are provided by sector at 0.25º × 0.25º resolution. Gridded emissions are available from http://www.meicmodel.org/dataset-mix. The key features of the MIX inventory are summarized in Table 1.

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 highlights the major improvements in the new inventory by comparing MIX with other Asian emission inventories. Uncertainties and limitations of the inventory are discussed in Sect. 5. Concluding remarks are provided in Sect. 6.

2. Compilation of the MIX emission inventory

2.1 Methodology

Five emission inventories are collected and incorporated into the mosaic inventory, as 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 NH_3 emission inventory by Peking University (referred to as PKU-NH_3 inventory hereafter, Huang et al., 2012), an Indian emission inventory developed by Argonne 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 by the following hierarchy. REAS2 was used as the default where local emission data are absent. Emission inventories compiled by the official agencies or developed with more local information are selected to override REAS2, which include MEIC for mainland China, ANL-India for India, and CAPSS for the Republic of Korea. Detailed information and advantages of these inventories are presented in Sect. 2.2. As only a few species (SO_2, BC, OC, and power plant NO_x) were available from ANL-India, REAS2 was used to supplement the missing species. A mosaic process was then used to combine ANL-India and REAS2 into a single dataset for India emissions. It is worth noting that the REAS2 have incorporated local inventories for Japan and Taiwan, which are subsequently adopted in MIX for these two regions. PKU-NH_3 was further used to replace MEIC emissions for NH_3 over China, given that PKU-NH_3 was developed with a process-based model that represented the spatio-temporal variations in NH_3 emissions. Table 2 lists the information of each inventory used in MIX.

Figure 2 illustrates the mosaic process for the MIX inventory development. Each dataset was reprocessed to 0.25° × 0.25° resolution with monthly variations when necessary. We used monthly gridded emissions from each component inventory where available, and assumed no monthly variation in emissions when the component inventory only provided annual emissions. The monthly profiles and spatial proxies used in each component emission inventories are summarized in Table S3 and Table S4.

For each regional emission inventory, emissions were acquired with sub-sector information and then aggregated into five sectors: power, industry, residential, transportation, and agriculture. Table S5 presented the sectoral mapping tables from sub-sectors to the five MIX sectors for each regional inventory. For each sub-sector, the corresponding IPCC sectors are also provided in Table S5. For agriculture sector, only NH_3 emissions are provided in the MIX inventory given that soil NO_x emissions and agriculture PM emissions are not available in the regional inventories used for compiling MIX. Emissions from open-biomass burning, fugitive dust, aviation, and international shipping were excluded in the MIX inventory because those emissions were only available in a few inventories.

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 monthly emission grid maps over Asia with a uniform spatial resolution of 0.25° × 0.25°.

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 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 CO_2 emissions from the Carbon Monitoring for Action database (CARMA, Wheeler and Ummel, 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 the REAS2 to the year 2010, following the same approach documented in Kurokawa et al. (2013).

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, b, 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 for both air pollutants and CO_2 by sectors at 0.25 × 0.25 degree resolution. We aggregated the 11 REAS2 sectors to five sectors provided in the MIX inventory. Emissions from open-biomass burning, aviation, and international shipping were excluded from the REAS2 before incorporating into MIX. 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 areal sources and gridded at 0.25 × 0.25 degree resolution using maps of rural, urban and total populations and road networks (See Table S4).

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 emission inventories developed by the same group (Streets et al., 2006; Zhang et al., 2007a, 2007b, 2009; Lei et al., 2011) and integrated them into a uniform framework. The major improvements include a unit-based power plant emission database (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 0.25º × 0.25º 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. NH_3 emissions in MEIC were replaced by PKU-NH_3, which will be discussed in the next section.

2.2.3 PKU-NH_3 for China

We used a high-resolution NH_3 emission inventory in China compiled by Peking University (PKU-NH_3, Huang et al., 2012) to replace China's NH_3 emissions in MEIC. MEIC used annual and regional average NH_3 emission factors to calculate emissions from each source category, while PKU-NH_3 used a process-based model to estimate NH_3 emissions which parameterized the spatial and temporal variations of emission factors with consideration of ambient temperature, soil property, and other factors. For 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-NH_3 estimated 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 excluded from the MIX inventory aggregation since the MICS-Asia III project uses GFED dataset for biomass burning. PKU-NH_3 is available at 1km × 1km resolution with monthly variation. We then regridded PKU-NH_3 monthly emissions to 0.25º × 0.25º. In the MIX inventory, 2006 emissions from PKU-NH_3 are used for both 2008 and 2010 since 2006 is the most recent year for emissions in PKU-NH_3 when the MIX inventory was developed. As the major drivers of 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.

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 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 (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 (~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 SO_2, NO_x, BC, and OC.

ANL-India is available for the period of 1990-2010 at 0.1º × 0.1º 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 (SO_2, BC, and OC for all sectors, and NO_x for power plants), monthly emissions by sector (excluding open biomass burning) from ANL-India were first regridded to 0.25º × 0.25º and then merged with REAS2 before being implemented in MIX to cover all species. The merge process is presented in Sect. 2.3.

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 estimated 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 CO_2 and five regulated air pollutants, SO_2, NO_x, CO, NMVOC, and PM_{10}. We derived sector-specific emission ratios between PM_{10} and the other aerosol components from Lei et al. (2011) and applied those ratios to estimate $PM_{2.5}$, 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 0.25º × 0.25º 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

ANL-India is available for SO_2, BC, and OC for all sectors as well as NO_x for power plants. 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 steps.

First, 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 could introduce inconsistency due to a mismatch of unit information in the two databases. In this work, we directly used ANL-India for SO_2, NO_x, BC, and OC emissions and used REAS for CO, NMVOC, $PM_{2.5}$, PM_{10}, and CO_2 but redistributed the total magnitudes of REAS2 power plant emissions by using the spatial distribution of power plants in the ANL-India inventory. We generated the spatial proxies of fuel consumption for each fuel type (coal, oil and gas) at 0.25×0.25 degree by aggregating fuel consumptions of each unit in the ANL-India inventory. We then applied the spatial proxy to the REAS2 estimates by fuel type for species that not included in ANL-India.

Second, we used BC and OC emissions from ANL-India but used $PM_{2.5}$ and PM_{10} emissions from REAS2. In certain grids, the sum of BC and OC emissions may exceed $PM_{2.5}$ emissions because the two inventories may use different activity data, emission factors and spatial proxies. The so-called "PMfine" species in chemical transport models are usually calculated by subtracting BC and OC emissions from total $PM_{2.5}$ emissions, leading to negative emissions of "PMfine" in those grids. In this case, we adjusted the emissions of $PM_{2.5}$ 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:

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

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). $Mol(j)$ is the mole weight of species j; and $C(j,m)$ is the conversion factor between j and m obtained from the mapping tables in 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.

2.5 Monthly profiles

We directly used monthly emissions from each regional emission inventory when compiling the MIX inventory. We assume no monthly variation in emissions when monthly profiles are absent from the regional emission inventories. Table S3 presents the monthly profiles used in each component emission inventory for MIX. In summary, monthly profiles for power plant emissions usually developed based on monthly statistics of power generation. Monthly profiles of industrial emissions are derived from monthly output of industrial products or industrial GDP. Residential monthly profiles are estimated from stove operation time based on ambient temperatures by regions (Streets et al., 2003).

2.6 Spatial proxies

We used gridded emissions from each regional emission inventory to compile the gridmaps of emissions. Locations of emitting facilities were used to derive gridded emission for large sources, while spatial proxies such as population density, road networks, and land use information are used to allocate emissions of areal sources. Table S4 summarized spatial proxies used in developing gridded emissions for each regional inventory.

3. Results

3.1 Asian anthropogenic emissions in 2010

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_2, 52.1 Tg NO_x, 336.6 Tg CO, 67.0 Tg NMVOC, 28.8 Tg NH_3, 31.7 Tg PM_{10}, 22.7 Tg $PM_{2.5}$, 3.5 Tg BC, 8.3 Tg OC and 17.3 Pg CO_2. 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_2 (50% of total), NMVOC (38%), PM_{10} (48%), and CO_2 (40%) emissions. Power plants have significant contributions for SO_2 (38% of total), NO_x (29%), and CO_2 (34%) emissions.

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_3, with more than 50% contribution for SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$ and CO_2 emissions. Following China, India is the largest contributor for NH_3 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_3, 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.

Table 4 presents Asian 2010 emissions by region and by sector. Emissions by country and by sector can be downloaded from the MIX website (http://www.meicmodel.org/dataset-mix.html). China's anthropogenic emissions in 2010 are estimated as follows: 28.7 Tg SO_2, 29.1 Tg NO_x, 170.9 Tg CO, 23.6 Tg NMVOC, 9.8 Tg NH_3, 16.6 Tg PM_{10}, 12.2 Tg $PM_{2.5}$, 1.8 Tg BC, 3.4 Tg OC and 10.1 Pg CO_2. Overall, industry is the largest emitter of China's anthropogenic emissions, contributing 49% of the total CO_2 emissions and 59%, 39%, 61%, and 50% of SO_2, 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 SO_2 emissions in China in 2010 surpassed SO_2 emissions from the U.S. and Europe combined. Power plants contributed 32% of the total CO_2 emissions and 28%, 33%, and 7% of SO_2, NO_x, and $PM_{2.5}$ emissions, respectively. Emission ratios of SO_2/CO_2 and $PM_{2.5}/CO_2$ are lower in power plants than in the industrial sector, reflecting 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_2 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.

In the MIX inventory, Indian emissions in 2010 are estimated as follows: 9.3 Tg SO_2, 9.6 Tg NO_x, 67.4 Tg CO, 16.9 Tg NMVOC, 9.9 Tg NH_3, 7.1 Tg PM_{10}, 5.2 Tg $PM_{2.5}$, 1.0 Tg BC, 2.5 Tg OC and 2.3 Pg CO_2. 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_2 emissions and the largest contributor for CO, NMVOC, $PM_{2.5}$, BC, and OC emissions, in which more than 70% of those emissions are contributed by biofuel combustion. With the rapid growth of coal-fired generation units, SO_2 emissions from Indian power plants are estimated to be 5.5 Tg in 2010, contributing 59% of the total Indian SO_2 emissions. The SO_2/CO_2 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).

Figure 6 compared per capita emissions by sector and by species in 2010 for each country. Emissions are ranked by GDP per capita of each country. The correlations between emission intensity (per capita emissions) and economic development (GDP per capita) at country level are not always significant because emission intensities are affected by not only economic level but also by other factors such as industrial structure and dominant fuel type. Nevertheless, the changes in emission intensities in general follow the pattern of Kuznets curve for most species except NH_3, BC, and OC. Emission intensities tend to increase following the GDP growth first and then tend to decrease for high-income countries. For BC and OC, per capita emissions are higher in developing countries than in developed countries because low-income countries with low incomes tend to use biofuels in which emitted more BC and OC than other fuel types.

Ratios of different species were widely used to inform emission characteristics. For example, SO_2/CO_2 ratio was used as an indicator of coal combustion and emission control levels (Li et al., 2007), and ratios of CO/CO_2 were used to inform combustion efficiency (Wang et al., 2010). Figure 7 compares regional emission ratios of SO_2/CO_2 and CO/CO_2 estimated by the MIX inventory. Emission ratios of SO_2/CO_2 are lowest in Other East Asia among different regions, which could be attributed to small share of coal use and high penetration of emission control facilities. While high emissions ratios of SO_2/CO_2 were found in Russia Asia and Central Asia due to high fraction of coal use and less emission controls. Other East Asia also has the lowest emission ratios of CO/CO_2

among different regions, owing to high contribution from industrial and transportation emissions. In contrast, high emissions from small residential combustions led to low combustion efficiencies and high emission ratio over India and Southeast Asia.

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 and emission ratios of 2010 to 2016 by country. For the whole of Asia, emission growth rates from 2006 to 2010 are estimated as follows: -8.1% for SO_2, +19.2% for NO_x, +3.9% for CO, +15.5% for NMVOC, +1.7% for NH_3, -3.4% for PM_{10}, -1.6% for $PM_{2.5}$, +5.5% for BC, +1.8% for OC and +19.9% for CO_2. Growth in CO_2 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).

During 2006-2010, CO_2 emissions were increased for China (+29.4%), India (+20.4%), Other South Asia (+15.2%), and Southeast Asia (+12.3%), and relatively stable for other regions. The increases in CO_2 emissions are driven by energy consumption growth stimulated by economic development over Asian regions, especially for China and India. As reported by IEA (International Energy Agency), the total primary energy consumption of Asia has increased by 20.6% during the period of 2005 and 2010 (IEA, 2013). During the same period, SO_2 emissions were decreased for China (-17.2%), Other East Asia (-9.5%), and Central Asia (-32.8%) and due to effective emission control, while SO_2 emissions were increased for India (+23.9%), Other South Asia (+23.9%), and Southeast Asia (+7.8%) due to growth in coal use and absence of desulfurization devices. The decrease in SO_2 emissions changes in Asian are dominated by changes in China and India. Figure 7(a) demonstrates the changes in SO_2 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_2 emission changes over Asia. SO_2 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_2 emission reduction over Asia. In contrast, SO_2 emissions in India increased by 27% during 2006-2010, owing to dramatic construction of new power plants and lack of emission control facilities (Garg et al., 2001, 2006). As a consequence, the Indian share of the total Asian SO_2 emissions increased from 13% in 2006 to 18% in 2010. NO_x and NMVOCs emissions were increased in all Asian regions except Other East Asia (-17.3% for NO_x and -13.0% for NMVOCs respectively), indicating lack of effective control measures for those two species over Asia. Increases of NO_x and NMVOC emissions are mainly driven by growth in industrial activities and vehicle population. For NO_x, remarkable emission increases are observed for China (+22.6%), India (+27.8%), Other South Asia (+20.5%), and Southeast Asia (+23.4%) during 2006-2010. For NMVOC, emissions were increased by 14.0%, 15.0%, 12.3%, 24.9%, 23.6%, and 9.3% for China, India, Other South Asia, Southeast Asia, Central Asia and Russia Asia respectively. Emission changes of other species are relatively small (i.e., within 6%) during 2006-2010. For CO, PM_{10}, 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. 8b), 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. While in India, Other South Asia and Southeast Asia, CO emissions increased by 21%, 11%, and 16%, respectively, between 2006 and 2010.

Satellite observations have shown promising capabilities in detect trends in surface emissions (Streets et al., 2013). The increases in NO_x emissions over China and India were confirmed by satellite-based inversions and the growth rates in satellite-based NO_x emission trends during 2006-2010 are generally comparable to our estimates in emission inventories (Table 6). For SO_2 emissions, the downward trend over China and upward trend over India were also observed by satellite remote sensing, while higher growth rates were detected by OMI than the bottom-up emission inventory (Krotkov et al., 2016). The downward trend of CO emissions over China in recent years 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). The decreasing rate of CO emissions over China is estimated to be -1.2% yr^{-1} from 2006 to 2010 in in the MIX inventory, consistent with the rates observed by multiple satellites in range of -1.0% yr^{-1} to -3.1% yr^{-1} during 2000-2012 (Table 6).

3.3 Speciated NMVOC emissions

Figure 9 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 varies significantly by different chemical groups. Over Asia, the industrial sector is the major source of emissions of alkanes and aromatics. Alkanes emissions from industrial sector are mainly contributed by gas production and distribution (19.8% of total industrial emissions), coal combustion (17.1%), and oil refinery (15.0%), and aromatics emissions are mainly contributed by architectural paint use (21.0% of total industrial emissions), other industrial paint use (16.6%), and gas production and distribution (10.6%). The residential sector has a high contribution of OVOCs, alkynes, and alkenes, among which mainly contributed by biofuel combustions. 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. The high emissions of alkenes in India (and Other South Asia) are mainly from contributions of biofuel combustions and motorcycles, and OVOC emissions in India are dominant by biofuel combustions. Southeast Asia shares around 20% of the emissions of alkanes, alkenes, aromatics and OVOCs.

3.4 Seasonality

Monthly emissions by sector and by Asian region are provided in Table S6 – Table S14. Monthly profiles in emissions are highly sector-dependent, given that monthly activity rates vary among different sectors. Figure 10 illustrates the monthly variations of Asian SO_2, CO, $PM_{2.5}$, and CO_2 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_2 emissions in winter are even higher than other species, because SO_2 emissions from China dominate residential emissions in Asia (70% of total), of which 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 11 presents monthly variations of SO_2, CO, $PM_{2.5}$, and CO_2 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 degree resolution. Emission maps of all species in 2010 are shown in Fig. 12. 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 degree are available from the website, including SO_2, NO_x, CO, NH_3, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC, and CO_2. Speciated NMVOC Emissions for CB05 and SAPRC-99 chemical mechanisms are provided at the same spatial and temporal resolution. The MIX inventory has been regridded to 0.1 × 0.1 degree 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).

4 Comparison with other inventories

4.1 MIX, REAS2 and EDGAR v4.2 over Asia

A comprehensive inter-comparison among different emission inventories over Asia was conducted by Kurokawa et al. (2013). In this work, we compare the MIX inventory with REAS2 and EDGAR v4.2 (EC-JRC/PBL, 2011), 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 v4.2. Russian Asia was excluded from comparison. Asian anthropogenic emissions of MIX, REAS2 and EDGAR v4.2 in 2008 are tabulated in Table 7. Over Asia, MIX and REAS 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 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.

Remarkable differences are observed between MIX and EDGAR v4.2. Compared to MIX, 2008 Asian emissions in EDGAR v4.2 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 with differences within 3.2%. Figure 13 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 7. Discrepancies are relatively large at the sector level compared to total emissions. EDGAR's estimates for SO_2 emissions from power plants are 60% higher than estimates in MIX. For China, 70% of power generation capacities were equipped with FGD and the average SO_2 removal efficiency was 78% (Liu et al., 2015). The high estimates in EDGAR v4.2 most likely due to underestimation of FGD penetration or SO_2 removal efficiencies of FGD (Kurokawa et al., 2013). Remarkable differences for the residential and transportation sectors are found for NO_x, CO, and NMVOC estimates in the two inventories. For instance, EDGAR v4.2 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 v4.2 are about a factor of 1.5 lower than in MIX, leading to 35% lower estimates of CO emissions in EDGAR v4.2 compared to MIX. Underestimates of CO emissions in EDGAR v4.2 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 discrepancy by sector could only be attributed to differences in the raw emission factors and abatement measures. Although a point-by-point comparison of emission factors between EDGAR v4.2 and MIX is not feasible, we can still speculate that EDGAR v4.2 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 v4.2 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 v4.2 estimates. Differences in PM_{10} emissions at the sector level are also large; similar estimates of PM_{10} emissions in the two datasets are rather a coincidence than real agreements.

For CO_2 emissions, good agreements are found among MIX, REAS2 and EDGAR v4.2 inventories in Asia with differences in total emissions less than 1%. CO_2 emissions from biofuel combustion are included in the inter-comparison. CO_2 emission estimates in MIX and REAS2 only differ in China because REAS2 are used in MIX for regions other than China. Total CO_2 emissions in MIX and REAS2 are quite similar (1.5% higher in REAS2), while REAS2 estimated higher emissions for power sector (+386 Tg or +13.6% compared to MIX) but lower emissions for industry (-293 Tg, -6.8%) emissions. EDGAR v4.2 estimates lower CO_2 emissions for China (-308 Tg, -3.4%) and higher emissions for other regions: +102 Tg (+5.6%) for Other East Asia, +83.9 Tg (+5.6%) for Southeast Asia, +204 Tg (+9.7%) for India, +29.5 Tg (+7.5%) for Other South Asia, and +25.3 Tg (+6.4%) for Central Asia. At sector level, EDGAR v4.2 estimates are 29% (+833 Tg) higher for power, 22% (-944 Tg) lower for industry, and 31% (-192 Tg) lower for transportation over China compared to MIX. For Other East Asia, differences between EDGAR v4.2 and MIX are mainly contributed by power sector, with 20% higher emissions (+130 Tg) in EDGAR v4.2. Residential sector is the main contributor to the differences between EDGAR v4.2 and MIX over India, with 28% (+177 Tg) higher emissions estimated in EDGAR v4.2. The relatively large discrepancy at sector level can be attributed to differences in energy statistics and emission factors (Guan et al., 2012; Liu et al., 2015) as well as differences in sector definitions. In particularly, EDGAR v4.2 used fuel consumption data from IEA statistics while MIX and REAS2 used provincial level data from Chinese Energy Statistics, which can differ by 20% at sector level (Hong et al., 2016). Emissions from heating plants are aggregated to the industrial sector in the MIX inventory, while in EDGAR v4.2, heating plants are aggregated to the energy sector and then compared to the power sector in the MIX inventory. In the future, harmonizing the sector (sub-sector) definition among global and regional inventories would help to reduce the discrepancy of emission estimates at sector level.

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 SO_2 and CO_2 (3% differences for SO_2, and 8% for CO_2) in 2008, implying similar estimates in energy consumption and emission factors in two inventories. Compared to MIX, REAS2 estimates lower emissions of NO_x, PM_{10}, $PM_{2.5}$ by more than 20%, mainly due to the differences in the emission factors used in compiling China's emissions. 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 plants to large ones. REAS2 included 380 power plants for China, compared to 2411 plants in MIX. While power plants in REAS2 are large ones which contributed 72% of CO_2 emissions in China.

Figure 14(a) compares CO_2 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. 14(a) 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 CO_2 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_2 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).

Figure 14(b) further compares the emission ratios of SO_2/CO_2 in the three inventories for individual power plants over Shanxi. Large deviations of SO_2/CO_2 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_2/CO_2 emission ratio with increase of plant size (corresponding to higher CO_2 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 SO_2/CO_2 emission ratios are found in REAS2, because power plant SO_2 emissions in REAS2 were estimated by using the average FGD penetration rates at provincial level (Kurokawa et al., 2013). The constant ratios for all power plants in EDGAR indicate that (a) the emission factors are not varied within China and (b) the spatial distribution treats all power plants equal, which doesn't take the variations among power plants into consideration.

4.2.2 Agriculture

The agriculture sector is a dominant source of NH_3 emissions, mainly contributed by fertilizer applications and manure managements. MIX incorporated the PKU-NH_3 inventory for China, which estimated agricultural NH_3 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 8 compares agricultural NH_3 emissions in China estimated in different emission inventories. Compared to other work, PKU-NH_3 yields lower estimates for fertilizer application but higher estimates for manure management. The differences are mainly because PKU-NH_3 used local correction factors for fertilizer volatilization and manure loss rate (Huang et al., 2012). Top-down inversion of NH_3 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, Figure 15 compares NH_3 agricultural emissions for MEIC and the PKU-NH_3 inventory for different climate zones. MEIC agrees well with PKU-NH_3 in temperate zones but is significantly higher than PKH-NH_3 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 NH_3 (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_3 estimated 9% average NH_3 loss rate for urea for tropical zones and 14% for temperate zone.

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 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). Emission standards implemented during 2005-2010 are summarized in Table S15. 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_x and PM (dominated by diesel vehicles).

4.3 India

For India, MIX used ANL-India for SO_2, BC, and OC emissions and REAS2 for other species. Here we compare ANL-India and REAS2 for SO_2, BC, and OC emissions, to evaluate the impact of using ANL-India. Both ANL-India and REAS2 used energy consumption data from 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_2 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, 2002b). 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 $0.25^{\circ} \times 0.25^{\circ}$ 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_x 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 Uncertainties and limitations

The MIX emission inventory subjects to uncertainties and several limitations. Emission estimates from bottom-up inventories are uncertain due to lack of complete knowledge of human activities and emission from different sources. Uncertainty ranges of an emission inventory could be estimated using propagation of error or Monte Carlo approaches (e.g., Streets et al., 2003; Zhao et al., 2011). However, in a mosaic emission inventory like MIX, a normalized quantitative assessment of uncertainty ranges is difficult because detailed information for emission inventory development is not collected. Table 9 summarized the uncertainty range estimates for China, India, and other Asian regions in different regional emission inventories. It should be noted that those ranges are not directly comparable due to differences in methods (propagation-of-error or Monte Carlo simulation). However, those numbers might roughly represent the uncertainty ranges in the MIX inventory as it was compiled from several inventories listed in Table 9. In general, uncertainty ranges are relatively small for species which emissions are dominated my large-scale combustion sources (e.g., SO_2, NO_x, and CO_2) but lager for species which emissions are mainly from small-scale and scattered sources (e.g., CO, NMVOC, and carbonaceous aerosols). More detailed discussions on the uncertainty sources of Asian emission inventories can be found in previous literatures (e.g., Lu et al., 2011; Zhao et al., 2011; Kurokawa et al., 2013).

As indicated by Janssens-Maenhout et al. (2015), the mosaic process could introduce additional and undesired uncertainties when compiling a gridded emission inventory from different datasets. The uncertainties may arise from inconsistencies among datasets, including missing species in specific datasets, closure of mass balances for aerosols, and inconsistency on the country boarders.

When species in a specific inventory were missing, alternative estimates or datasets were used to fill the gap in which may involve additional uncertainties. In the MIX inventory, $PM_{2.5}$, BC, and OC emissions for Republic of Korea were roughly estimates from PM_{10} emissions in the CAPSS inventory and sector-specific emission ratios between PM_{10} and other aerosol components from Lei et al. (2011). For India, we used ANL-India for SO_2, BC, and OC for all sectors and NO_x for power plants. REAS2 was used to fill the gap where emissions from ANL-India were absent (See Sect. 2.3 for detailed process procedure). For above cases where estimates of different species in the same country were obtained from various sources, the ratios between species may less reliable and should be used with caution.

When using different datasets for different types of aerosols in the same country, additional uncertainties for aerosol emissions might be introduced from the inconsistency in mass balance closure due to differences in spatial proxies (Janssens-Maenhout et al., 2015), which is the case for Indian emissions in this study. In the MIX inventory, BC and OC emissions were obtained from ANL-India while $PM_{2.5}$ and PM_{10} emissions were taken from REAS2. During the mosaic process of Indian emissions, additional check was performed by grid for each sector and emissions of $PM_{2.5}$ were adjusted to the sum of BC and OC emissions for the grids which the sum of BC and OC emissions exceeds $PM_{2.5}$ emissions.

For a mosaic emission inventory, inconsistencies could occur at country boarders when emissions of the two adjacent countries were obtained from different datasets (Janssens-Maenhout et al., 2015). In the MIX inventory, the inconsistencies are expected at the country boarder of China and India. However, low populations and emissions are observed along the border of China, reducing the impact of cross-border grids on the accuracy of emissions. Also deriving country totals from the gridded emissions is not appropriate for small countries due to the impact from cross-board grids, especially for those grids with large point source emissions (Janssens-Maenhout et al., 2015).

The current MIX inventory also has several limitations. Firstly, it provides emissions with aggregated sectoral information, which may be sufficient for the base case model but insufficient for targeted policy cases. Secondly, the MIX inventory is provided with moderate spatial resolution (i.e., 0.25º×0.25 º), which could support global and regional models but still too coarse for urban models. Finally yet importantly, gridded emissions are only available for 2008 and 2010 to support base years modelling activities in MICS-Asia and HTAP. For other years, modelers could use available global/regional inventories with more complete year coverage (e.g., EDGAR v4.2, REAS2) or extrapolate the gridded MIX inventory to the neighboring years. Developing a complete time series of gridded emission dataset with best available local inventories is a challenging task because it requires extensive international collaboration to coordinate various resources. Continuous efforts under international collaboration frameworks (e.g., MICS-Asia, HTAP) could help to deliver improved and updated emission inventories over Asia continuously.

6 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_3, 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 0.25º × 0.25º. 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. 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. The inter-comparison between MIX and other inventories indicated that significant differences in methodology and input data were used in different emission inventories. Harmonizing the efforts among different regions and research groups through international collaborations 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	29 countries and regions in Asia
Countries and regions	China, Japan, Democratic People's Republic of Korea, Republic of Korea, Mongolia, India, Afghanistan, Bangladesh, Bhutan, Maldives, Nepal, Pakistan, Sri Lanka, Brunei, Cambodia, Indonesia, Laos, Malaysia, Myanmar, Philippines, Singapore, Thailand, Vietnam, Kazakhstan, Kyrgyzstan, Tajikistan, Turkmenistan, Uzbekistan, Russia (East Siberia, Far East, Ural, West Siberia)
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

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
Year	1990-2010	2006	2008, 2010	2008, 2010	1996-2010	2008, 2010	2000-2010
Region	China	China	Republic of Korea	Japan	India	India	Asia
Seasonality	Monthly	Monthly	Annual	Monthly	Monthly	Annual	Monthly
Resolution	0.25 degree[a]	1 km	0.25 degree	1 km	0.1 degree	0.25 degree[a]	0.25 degree
SO_2	X		X	X	X		X
NO_x	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

[a]power plant emissions are developed with specific geophysical locations and allocated into 0.25° × 0.25° grids.

Table 3. National anthropogenic emissions in the MIX emission inventory in 2010 (Units: Tg for CO_2 and Gg for other species).

Countries	SO_2	NO_x	CO	NMVOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC	CO_2
China[a]	**28,663**	**29,071**	**170,874**	**23,619**	**9,804**	**16,615**	**12,200**	**1,765**	**3,386**	**10,124**
Japan	708	1,914	4,278	1,178	479	114	81	20	8	1,107
Korea, DPR	211	238	4,488	138	111	264	115	14	17	71
Korea, Rep of	418	1,062	838	851	190	124	87	24	8	541
Mongolia	99	62	735	47	97	109	46	2	4	15
Other East Asia[b]	**1,437**	**3,275**	**10,339**	**2,215**	**875**	**610**	**328**	**60**	**37**	**1,735**
India	**9,259**	**9,565**	**67,423**	**16,892**	**9,871**	**7,093**	**5,216**	**1,019**	**2,530**	**2,277**
Afghanistan	3	178	456	141	143	21	20	8	10	2
Bangladesh	133	368	2,575	788	1,016	342	234	33	121	84
Bhutan	5	13	302	50	41	26	21	4	14	5
Maldives	3	8	151	9	1	0	0	0	0	2
Nepal	30	83	2,109	443	254	150	139	27	105	34
Pakistan	1,397	946	9,279	2,112	1,859	600	558	114	390	263
Sri Lanka	133	116	1,321	367	122	152	111	15	59	31
Other South Asia[b]	**1,704**	**1,712**	**16,194**	**3,910**	**3,435**	**1,290**	**1,082**	**200**	**699**	**421**
Brunei	11	12	6	32	8	1	0	0	0	9
Cambodia	26	47	1,025	211	134	59	56	11	44	17
Indonesia	1,964	2,570	23,749	7,970	1,945	1,182	947	178	692	554
Laos	150	41	397	85	87	24	22	4	16	6
Malaysia	365	631	3,731	1,765	255	216	132	16	35	201
Myanmar	67	91	2,705	814	425	165	156	31	125	49
Philippines	503	361	2,347	869	413	193	123	15	68	119
Singapore	175	116	162	334	10	7	5	1	1	43
Thailand	614	809	8,572	2,327	649	495	275	35	149	297
Vietnam	575	442	8,231	2,234	665	710	562	87	322	232
Southeast Asia[b]	**4,449**	**5,120**	**50,925**	**16,640**	**4,592**	**3,051**	**2,278**	**378**	**1,452**	**1,527**
Kazakhstan	1,050	559	3,348	544	41	442	222	13	28	204
Kyrgyzstan	27	35	371	40	12	62	28	2	3	6
Tajikistan	14	25	192	30	15	22	13	1	1	4
Turkmenistan	64	124	417	238	14	64	30	2	3	52
Uzbekistan	493	228	899	310	50	373	165	3	11	121
Central Asia[b]	**1,648**	**971**	**5,227**	**1,162**	**133**	**963**	**458**	**21**	**46**	**387**
East Siberia	1,649	534	2,874	394	23	368	198	14	20	184
Far East	358	489	2,681	303	18	223	123	22	25	120
Ural	1,480	456	4,005	591	22	1,047	598	19	75	186
West Siberia	677	926	6,045	1,310	42	465	269	32	52	340
Russia Asia[b]	**4,164**	**2,405**	**15,605**	**2,597**	**105**	**2,103**	**1,188**	**87**	**172**	**830**

Asia	**51,324**	**52,118**	**336,588**	**67,034**	**28,816**	**31,726**	**22,749**	**3,530**	**8,322**	**17,301**

[a] Hong Kong, Macao, and Taiwan are included.
[b] The Asian region includes the set of countries listed in the section.

Table 4. Asian emissions by sector in 2010 for each region (Units: Tg for CO_2 and Gg for other species).

Regions	SO_2	NO_x	CO	NMVOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC	CO_2
China	**28,663**	**29,071**	**170,874**	**23,619**	**9,804**	**16,615**	**12,200**	**1,765**	**3,386**	**10,124**
Power	8,166	9,455	2,077	255	0	1,389	893	2	0	3,245
Industry	16,775	11,218	71,276	14,461	239	9,451	6,061	575	530	4,928
Residential	3,489	1,140	76,579	6,349	450	5,246	4,737	908	2,752	1,266
Transportation	234	7,257	20,942	2,553	76	529	509	281	104	684
Agriculture					9,040					
Other East Asia	**1,437**	**3,275**	**10,339**	**2,215**	**875**	**610**	**328**	**60**	**37**	**1,735**
Power	427	460	70	13	3	162	64	0	0	617
Industry	644	649	3,329	1,475	26	285	142	20	6	584
Residential	189	361	1,402	259	143	77	47	8	18	233
Transportation	176	1,805	5,538	467	23	87	74	32	13	300
Agriculture					680					
India	**9,259**	**9,565**	**67,423**	**16,892**	**9,871**	**7,093**	**5,216**	**1,019**	**2,530**	**2,277**
Power	5,476	2391	2,676	125	8	2,029	842	1	3	886
Industry	2,959	973	18,164	3,372	172	1,284	931	217	200	585
Residential	685	968	34,317	7,311	2,185	2,946	2,640	709	2,275	659
Transportation	139	5,233	12,267	6,085	9	834	802	92	52	147
Agriculture					7,496					
Other South Asia	**1,704**	**1,712**	**16,193**	**3,910**	**3,436**	**1,290**	**1,082**	**200**	**699**	**421**
Power	774	290	51	6	0	24	12	0	1	79
Industry	546	165	2,536	635	38	400	231	18	75	93
Residential	153	395	12,148	2,600	724	777	753	147	594	199
Transportation	231	863	1,459	669	1	90	87	34	29	49
Agriculture					2,673					
Southeast Asia	**4,449**	**5,120**	**50,925**	**16,640**	**4,592**	**3,051**	**2,278**	**378**	**1,452**	**1,527**
Power	1,596	1,136	580	110	10	540	157	2	0	393
Industry	2,101	748	4,309	3,182	133	902	566	28	245	450
Residential	364	396	26,804	5,792	1,139	1,478	1,430	286	1,140	424
Transportation	388	2,840	19,233	7,556	15	131	126	63	66	260
Agriculture					3,296					
Central Asia	**1,648**	**971**	**5,227**	**1,162**	**133**	**963**	**458**	**21**	**46**	**387**
Power	1,066	379	40	7	1	4	1	0	0	155
Industry	407	134	354	623	7	930	428	6	32	105
Residential	154	131	576	159	85	3	3	1	2	98
Transportation	20	327	4,257	373	1	26	26	15	12	29
Agriculture					39					
Russia Asia	**4,164**	**2,405**	**15,605**	**2,597**	**105**	**2,103**	**1,188**	**87**	**172**	**830**
Power	1,981	1,149	478	26	6	27	15	2	9	517
Industry	1,996	179	2,727	1,520	14	1,949	1,050	21	103	180

Residential	101	87	1,056	236	58	23	20	4	16	63
Transportation	86	990	11,344	815	2	105	103	59	44	71
Agriculture					25					
Asia	**51,324**	**52,118**	**336,588**	**67,034**	**28,816**	**31,726**	**22,749**	**3,530**	**8,322**	**17,301**
Power	19,487	15,260	5,972	543	28	4,175	1,984	7	13	5,893
Industry	25,429	14,065	102,695	25,267	629	15,200	9,409	884	1,192	6,925
Residential	5,134	3,478	152,882	22,707	4,784	10,551	9,630	2,063	6,798	2,944
Transportation	1,274	19,316	75,040	18,517	126	1,800	1,727	576	320	1,540
Agriculture					23,249					

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Table 5. Asian emissions in 2006 and trends between 2006 and 2010 (Units for emissions: Tg for CO_2 and Gg for other species)[a].

Regions	**SO_2**	**NO_x**	**CO**	**NMVOC**	**NH_3**	**PM_{10}**	**$PM_{2.5}$**	**BC**	**OC**	**CO_2**
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)	2,352 (0.81)	5,888 (0.73)	1,538 (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	**1,376 (1.24)**	**1,421 (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	1,676 (1.17)	1,999 (1.29)	19,379 (1.23)	6,134 (1.30)	1,634 (1.19)	1,237 (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)	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)**	**1,360 (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 (1.62)	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	**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)**	**13,878 (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)**

[a]numbers in the parentheses represent emission ratios of 2010 to 2006.

Table 6. Comparison of emission trends of NO_x, CO and SO_2 over Asia with satellite observations.

Species	Regions	Study	Method	Period	AGR (%/yr)[a]
NO_x	China	Berezin et al., 2013	Inverse modeling	2001-2008	11.5
	China	Gu et al.,2013	Inverse modeling	2005-2010	4.0
	China	Miyazaki et al., 2016	Inverse modeling	2005-2010	3.7
	East China	Mijling et al., 2013	Inverse modeling	2007-2011	9.0
	East China	Krotkov et al., 2016	Satellite	2005-2010	5.4
	Central East China	Itahashi et al., 2014	Satellite	2000-2010	11.0
	China	This work	Inventory	2006-2010	5.2
	India	Krotkov et al., 2016	Satellite	2005-2010	4.6
	India	Miyazaki et al., 2016	Inverse modeling	2005-2010	3.2
	India	This work	Inventory	2006-2010	6.3
SO_2	East China	Krotkov et al., 2016	Satellite	2005-2010	-6.9
	China	This work	Inventory	2006-2010	-4.6
	India	Krotkov et al., 2016	Satellite	2005-2010[c]	16.5
	India	This work	Inventory	2006-2010	5.5
CO	China	Yumimoto et al., 2014	Inverse modeling	2005-2010	-3.1
	China	Yin et al., 2015	Inverse modeling	2002-2011	-1.1
	East China	Worden et al., 2013	Satellite	2000-2012	-1.6, -1.0[b]
	China	This work	Inventory	2006-2010	-1.2

[a]AGR = annual growth rate.

[b]results are developed using MOPITT, AIRS respectively.

Table 7. Inter-comparisons of total anthropogenic emissions[a] among MIX, REAS2 and EDGAR v4.2 for 2008.

Unit: Tg yr^{-1}	SO_2	NO_x	CO	NMVOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC	CO_2
					Asia[b]					
MIX	49.00	46.38	317.11	60.26	27.66	30.16	21.71	3.40	8.04	1514
REAS2	52.82	46.17	344.20	65.94	32.74	34.21	23.51	2.95	7.55	1527
EDGAR v4.2	63.26	36.73	212.16	53.43	20.08	31.08				1528
					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

[a]including power, industry, residential, transportation, and agriculture.
[b]"Asia" refers to all Asian regions excluding the Russia Asia in MIX.

Table 8. NH_3 agriculture emission estimates for China.

Unit: Tg-NH_3 yr^{-1}	PKU-NH_3	MEIC v.1.0	REAS2	EDGAR v4.2	MASAGE_NH_3
Year	2006	2008	2008	2008	2005-2008[a]
Fertilizer application	3.20	4.40	9.40	8.26	3.64
Live stock	5.30	5.30	2.80	2.31	5.83

[a]averaged estimates during 2005-2008.

Table 9. Uncertainty in emission estimates by Asian regions in 2010 (95% Confidence Intervals, unit: %).

Regions	SO_2	NO_x	CO	NMVOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC	CO_2	References
China	±12	±31	±70	±68		±132	±130	±208	±258		Zhang et al. (2009)
						±91	±107	±187	±229		Lei et al. (2011)
	-14-13	-13-37				-14-45	-17-54	-25-136	-40-121		Zhao et al. (2011)
	-16-17							-43-93	-43-80		Lu et al. (2011)
	±31	±37	±86	±78	±153	±114	±133	±176	±271	±31	Kurokawa et al. (2013)
India	-15-16							-41-87	-44-92		Lu et al. (2011)
	±32	±49	±114	±137	±144	±120	±145	±178	±233	±49	Kurokawa et al. (2013)
Others	±35	±47	±131	±111	±148	±194	±208	±257	±286	±44	Kurokawa et al. (2013)

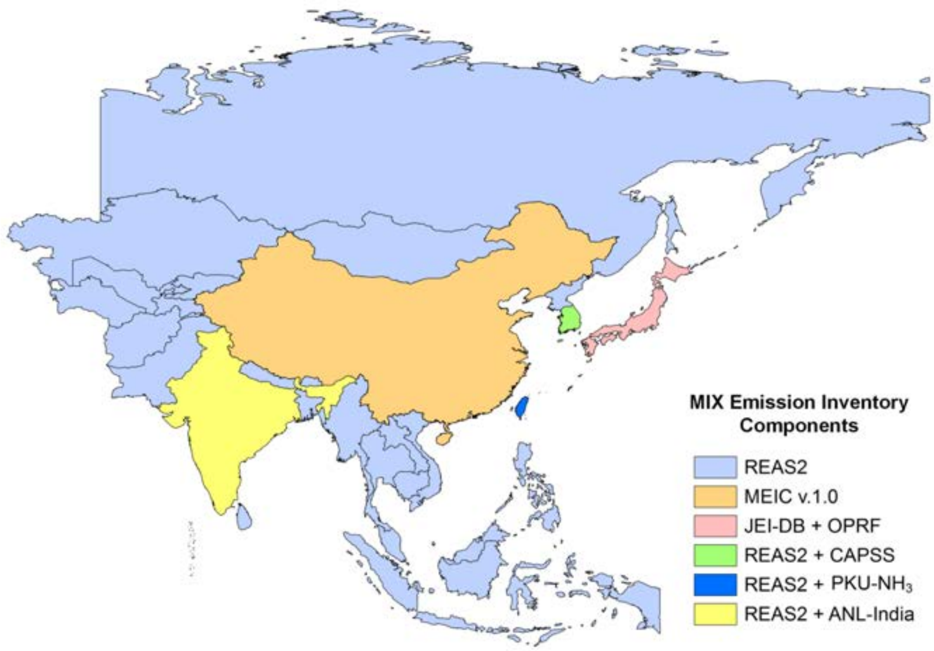


Figure 1. Domain and component of the MIX emission inventory.

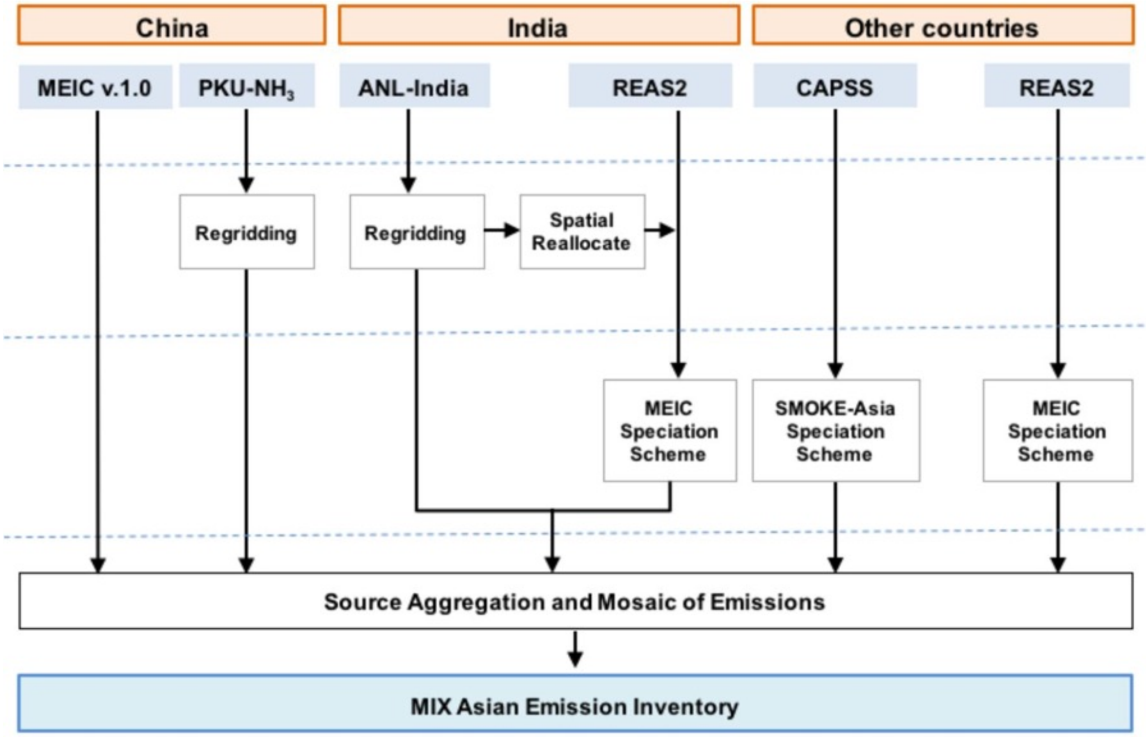


Figure 2. Schematic methodology of the MIX emission inventory development.

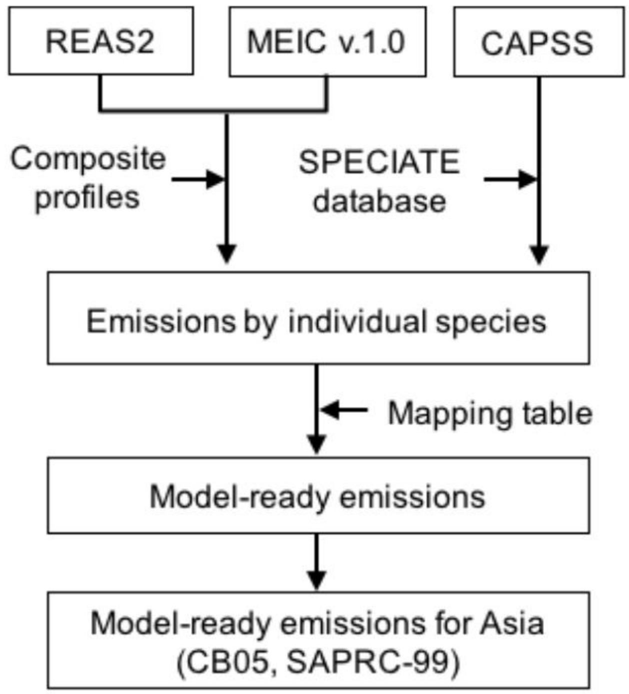


Figure 3. NMVOC speciation scheme used in the MIX inventory development. The mapping table is derived from Carter et al. (2013).

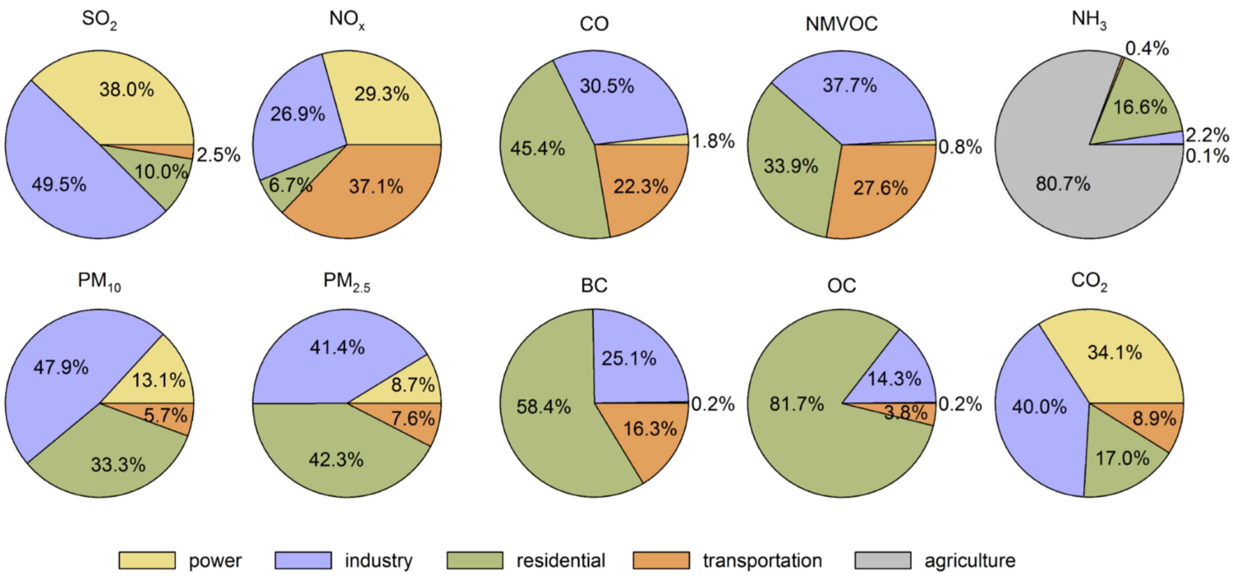


Figure 4. Emission distributions among sectors in Asia in 2010.

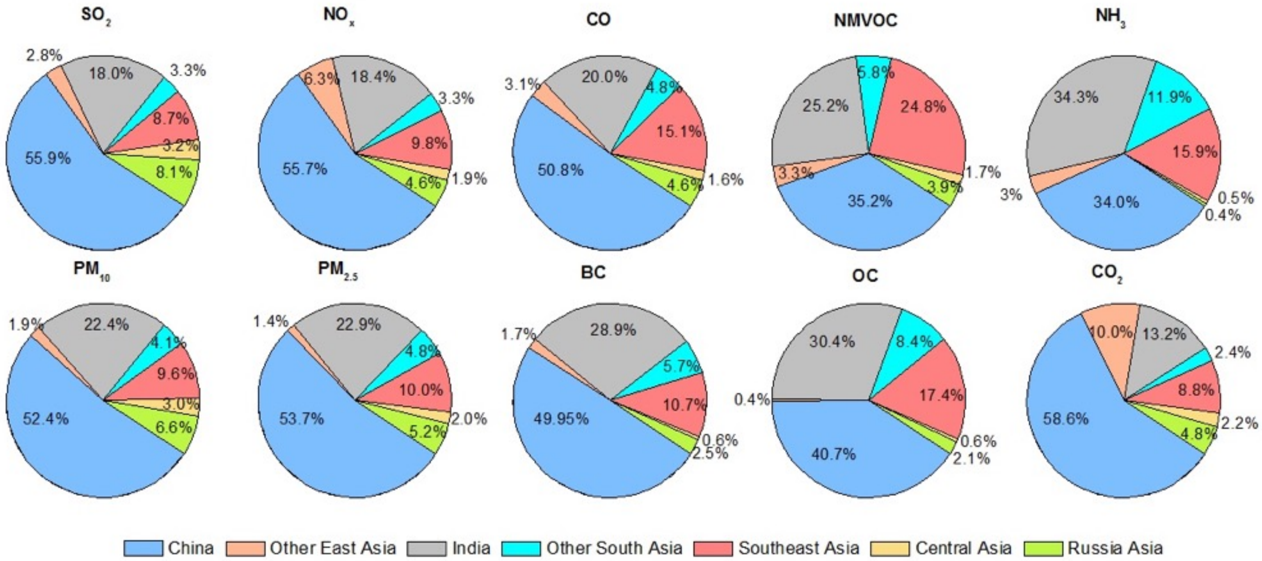


Figure 5. Emissions distributions by Asian regions in 2010.

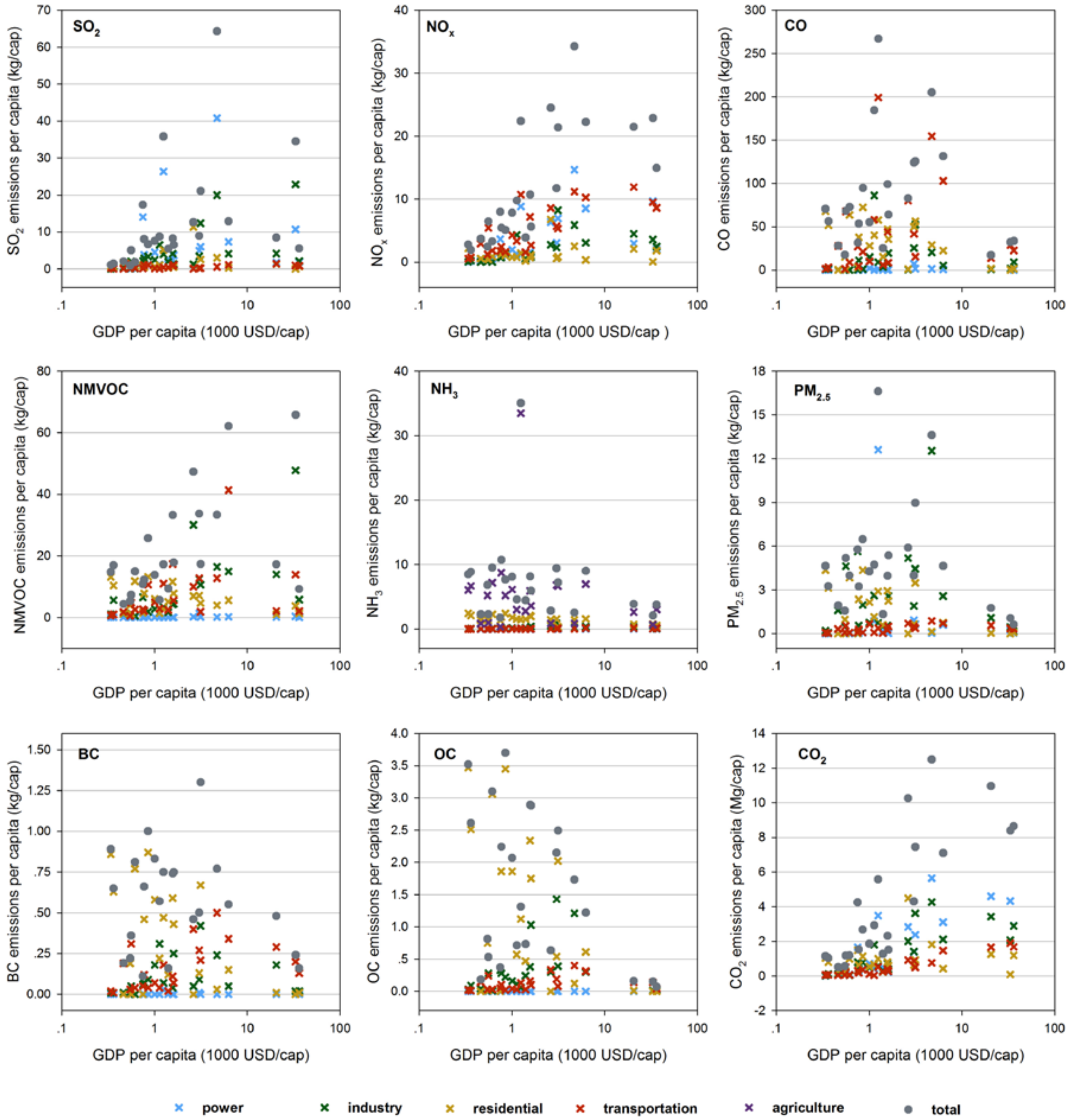


Figure 6. Per capita emissions by sector for 2010 in MIX, ranked by GDP per capita for each country.

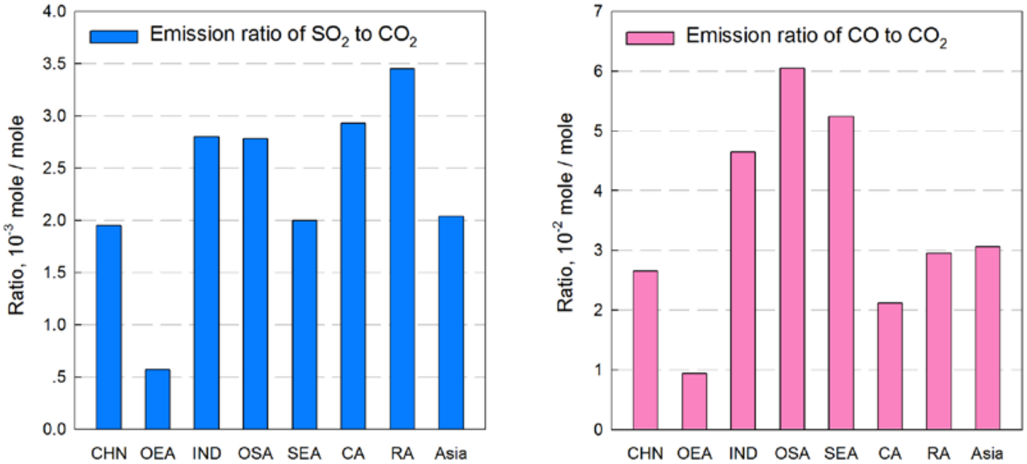
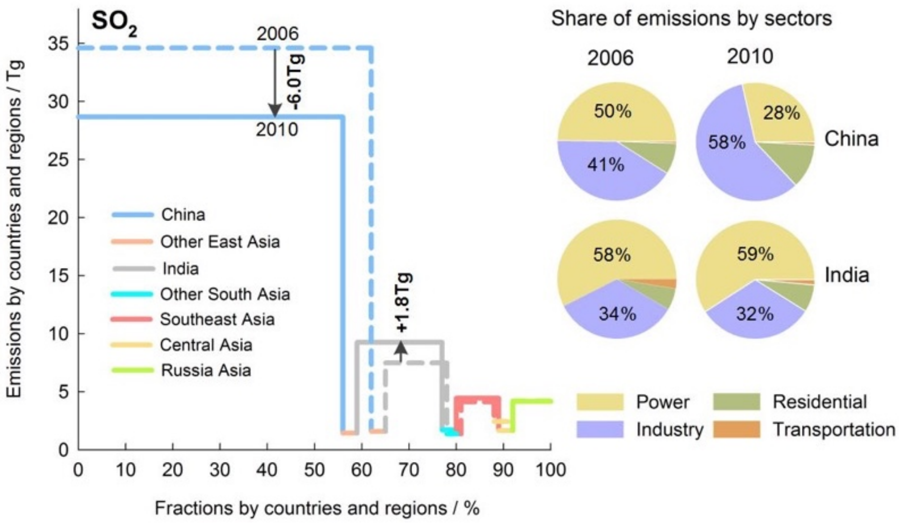
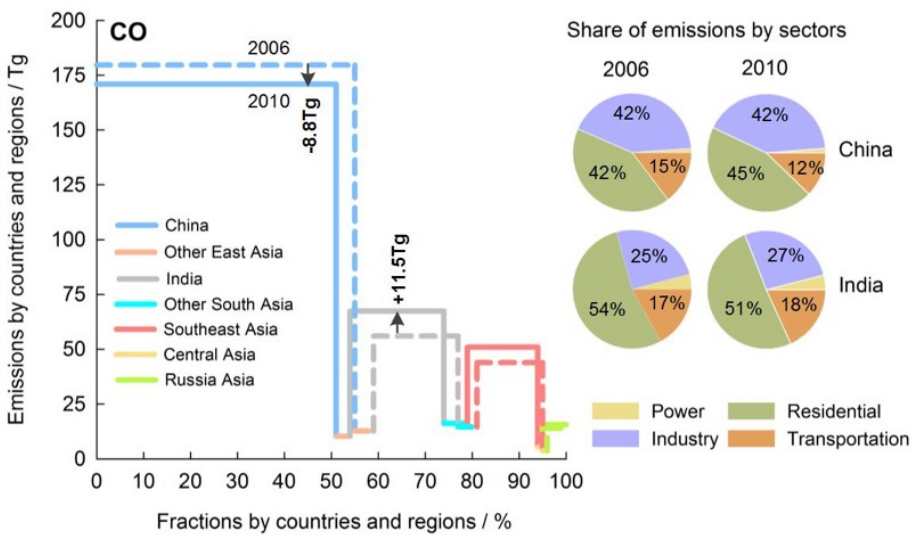


Figure 7. Emission ratios of SO_2 to CO_2, and CO to CO_2 by Asian regions. "CHN": China, "OEA": Other East Asia, "IND": India, "OSA": Other South Asia, "SEA": Southeast Asia, "CA": Central Asia, "RA": Russia Asia.



(a)



(b)

Figure 8. Emission changes from 2006 to 2010 by Asian regions for SO_2 (a) and CO (b). Left panel: Emissoins in 2006 and 2010 by region. *Y*-axis represents emissions by region. X-axis represents accumulative emission contribution of regions. The dotted and solid lines represent emissions in 2006 and 2010 respectively. Right panel: the shares of emissions by sectors over China and India in 2006 and 2010.

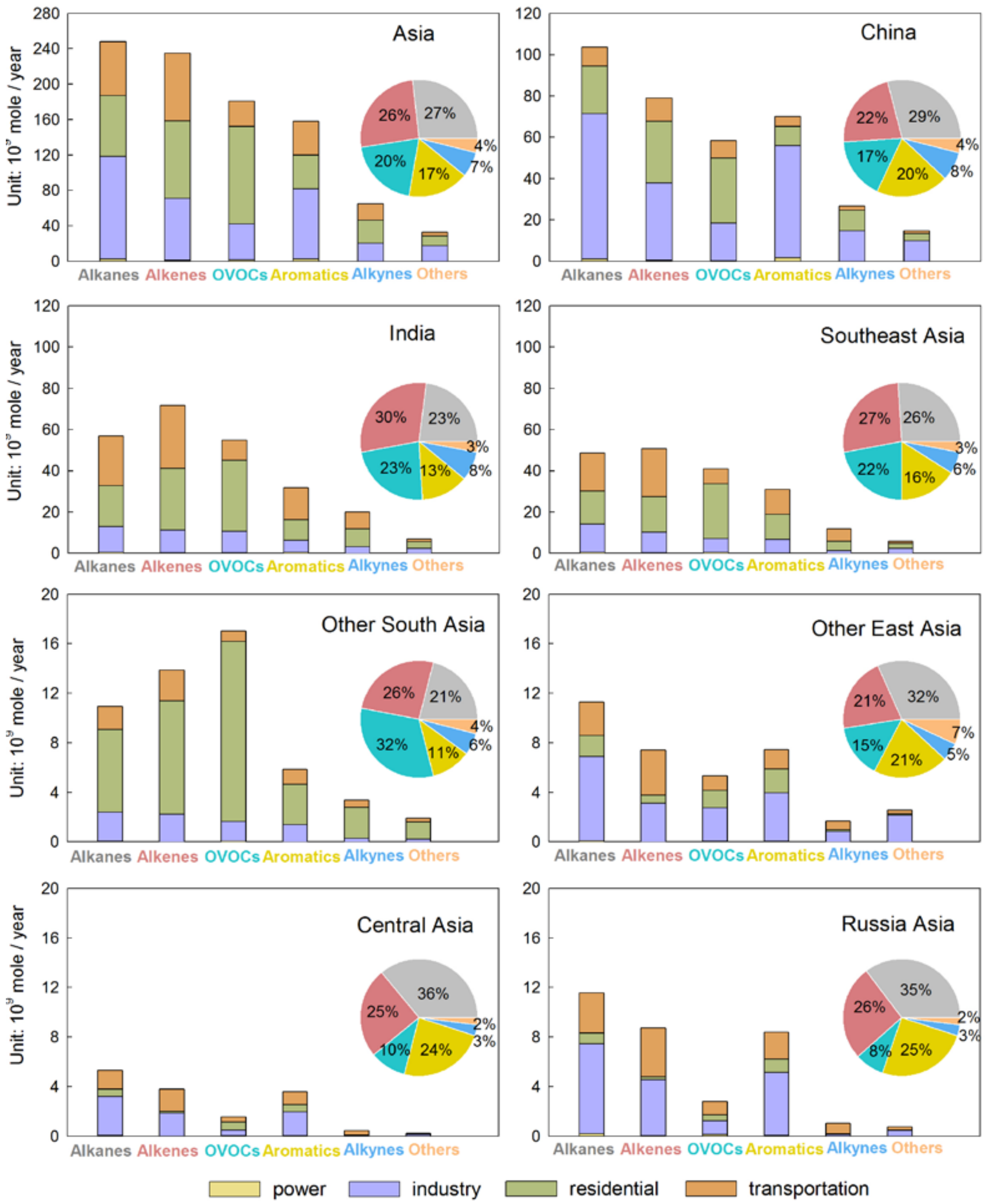


Figure 9. Speciated NMVOC Emissions for the year 2010 by chemical group and by Asian regions. Alkanes: ethane, propane, butanes, pentanes, hexanes, higher alkanes and their isomers; Alkenes: ethane, propene, isoprene, terpenes, higher alkenes and their isomers; Alkynes: ethyne and other alkynes; Aromatics: benzene, toluene, xylene, trimethyl-benzene, other aromatics and their isomers; OVOCs: aldehydes (formaldehyde, acetaldehyde, and higher aldehydes), ketones (acetone and higher ketones), alcohols (methanol, ethanol and higher alcohols), ethers and acids; "Others": halogenated hydrocarbons, unidentified species, etc.

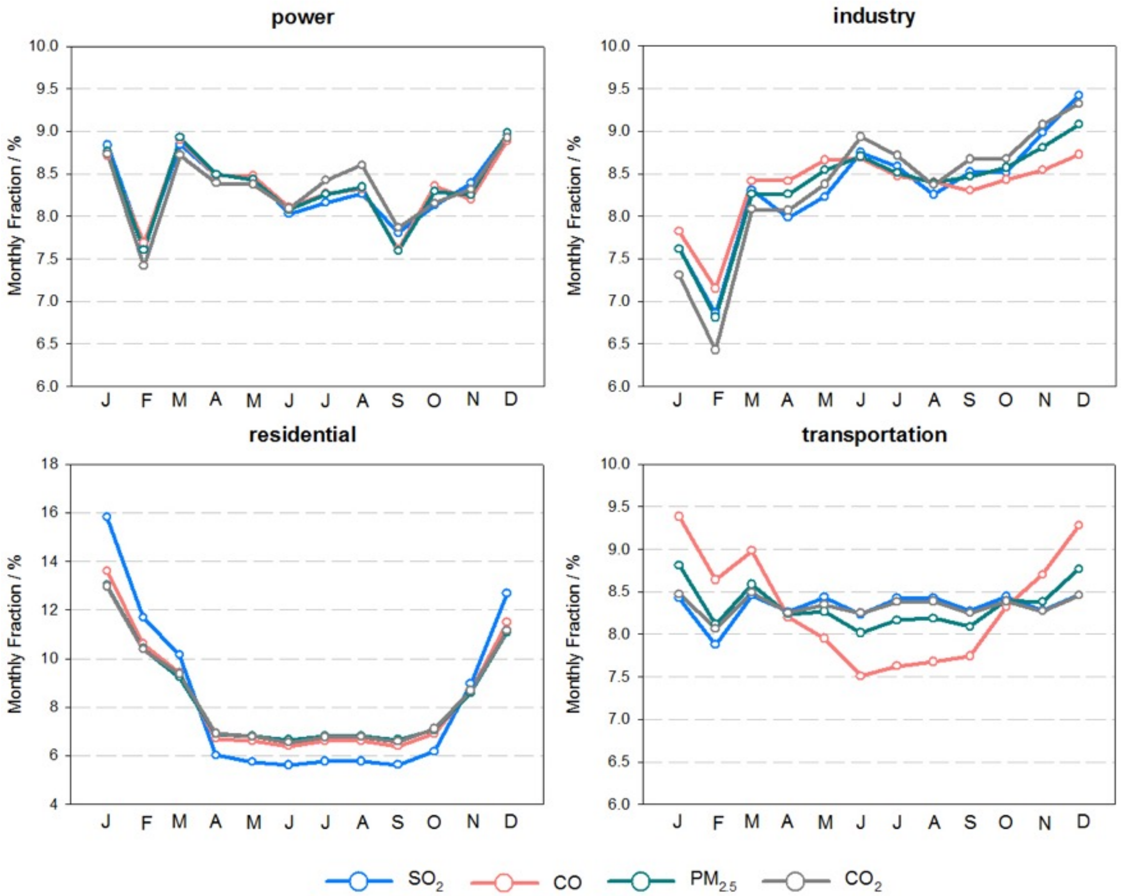


Figure 10. Monthly variations of Asian SO2, CO, PM2.5, and CO2 emissions by sector for the year 2010.

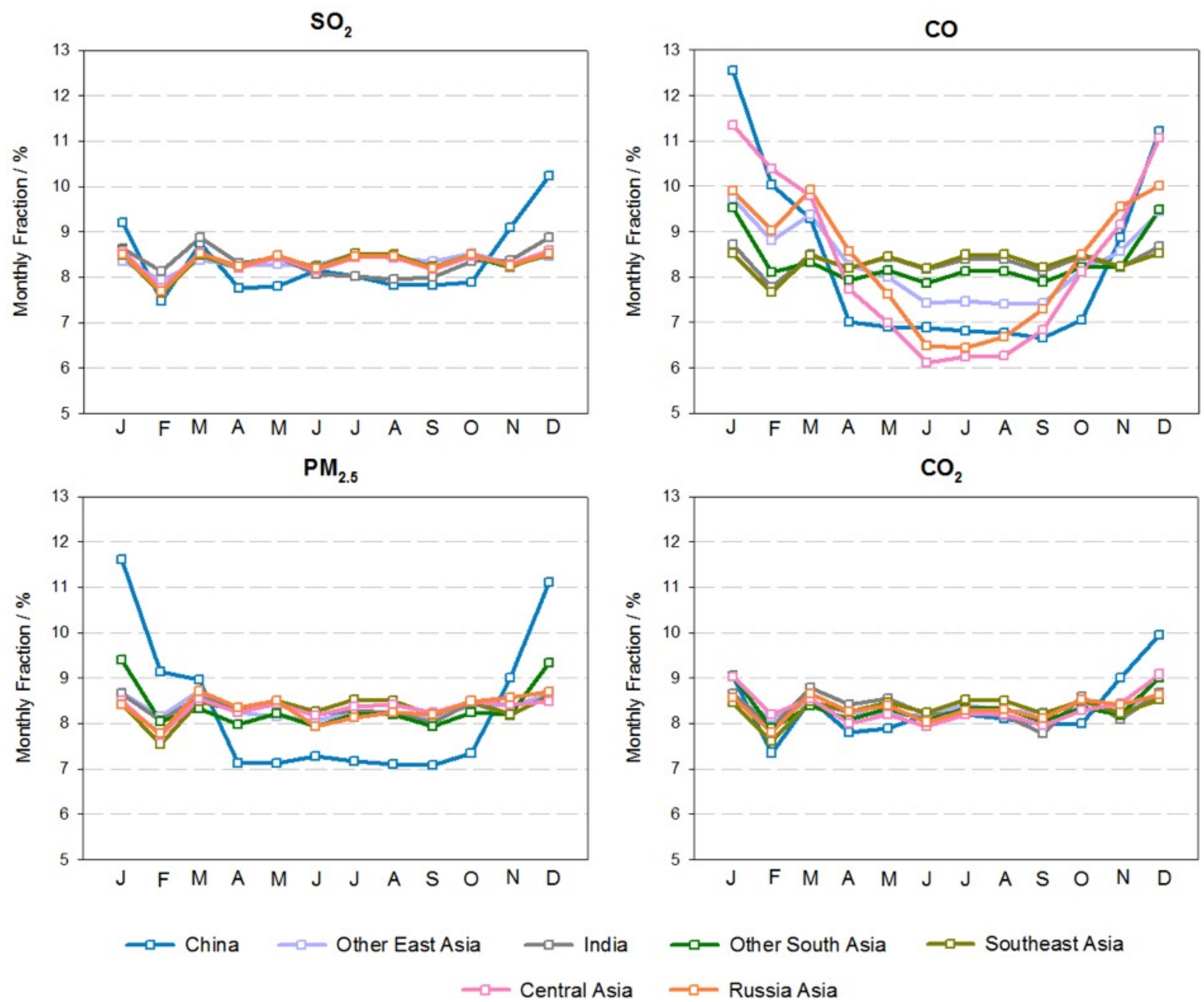
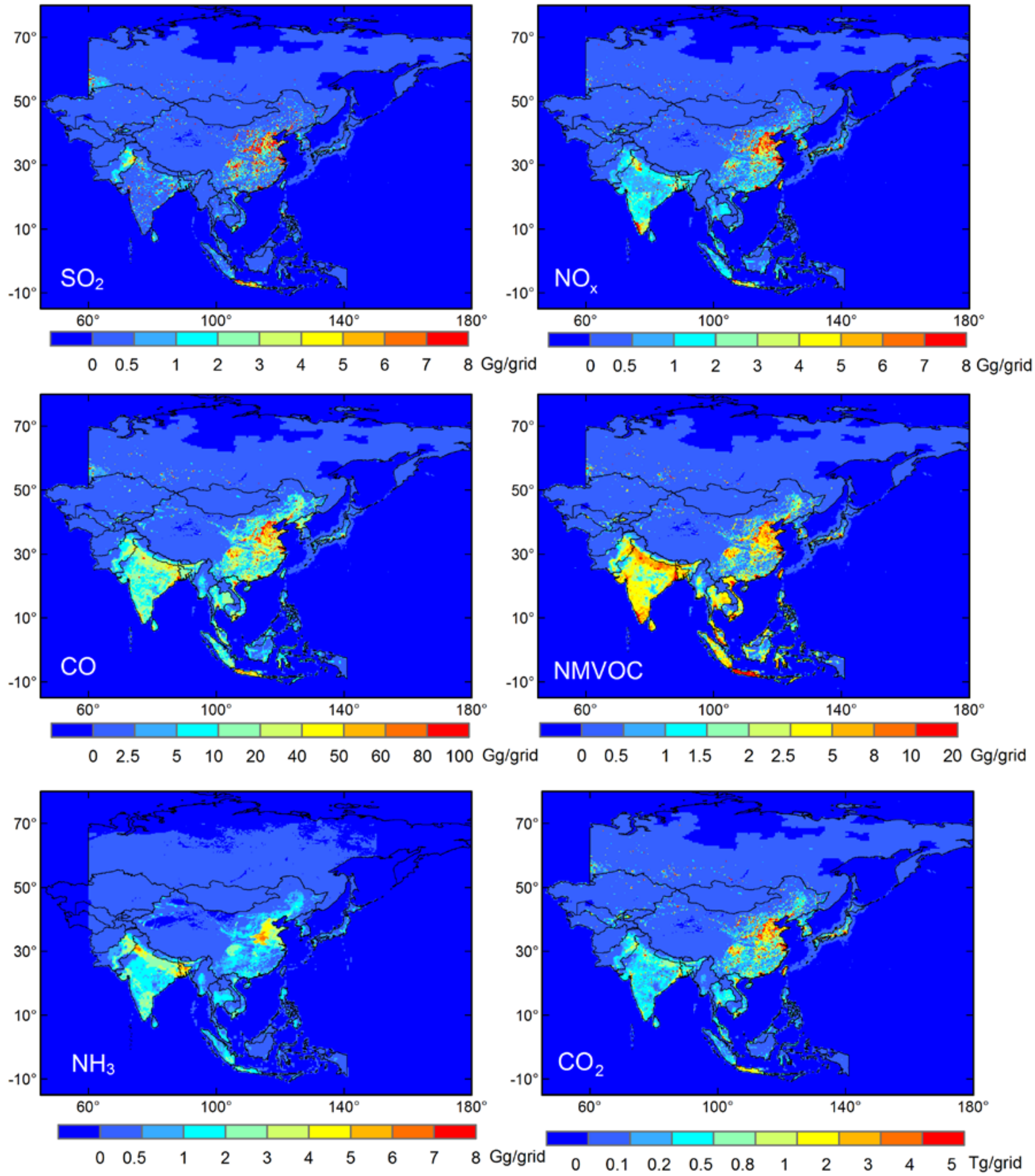
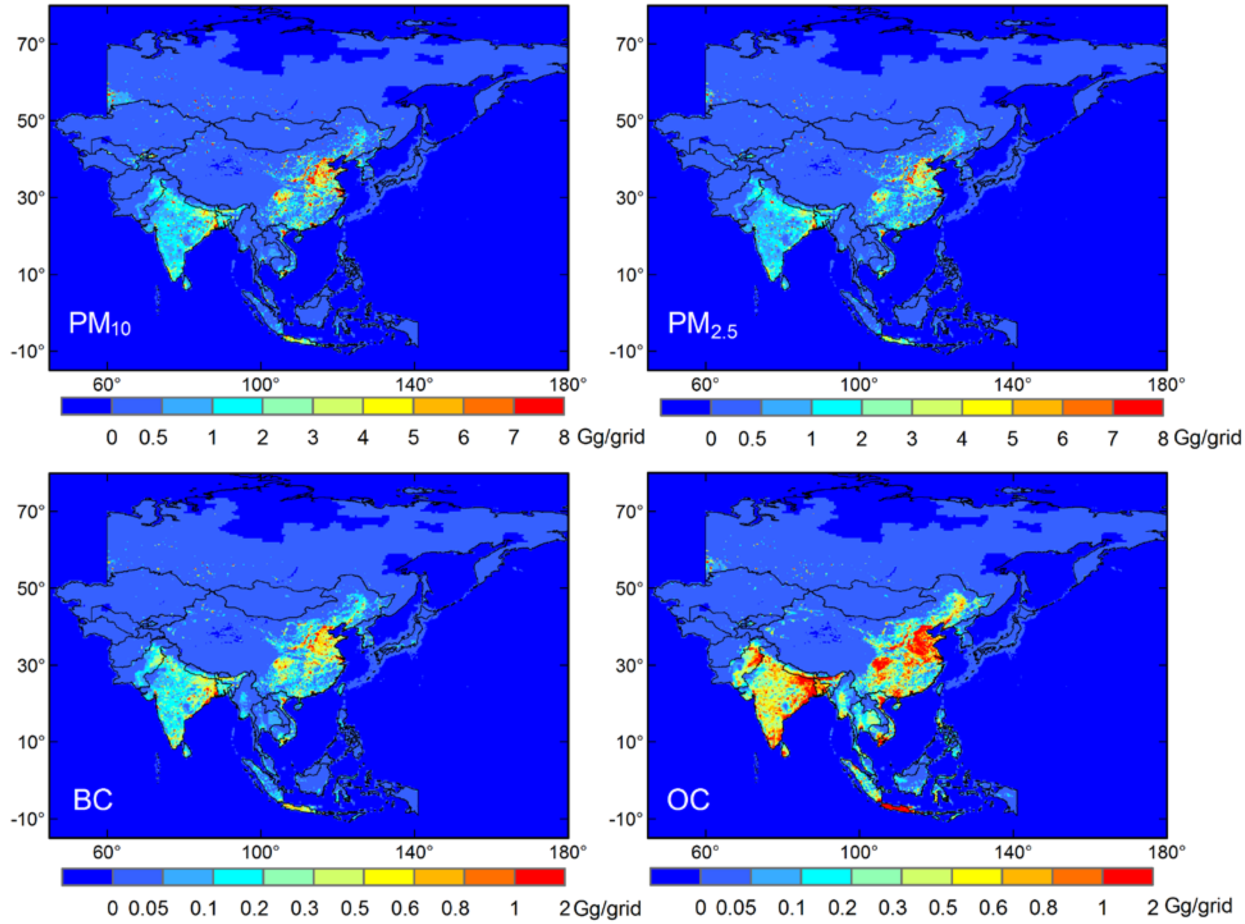


Figure 11. Monthly variations of SO2, CO, PM2.5, and CO2 emissions by Asian region for the year 2010.



(a) Gaseous species



(b) aerosol species

Figure 12. Grid maps for gaseous (a) and aerosol (b) species in the MIX Asian emission inventory, 2010.

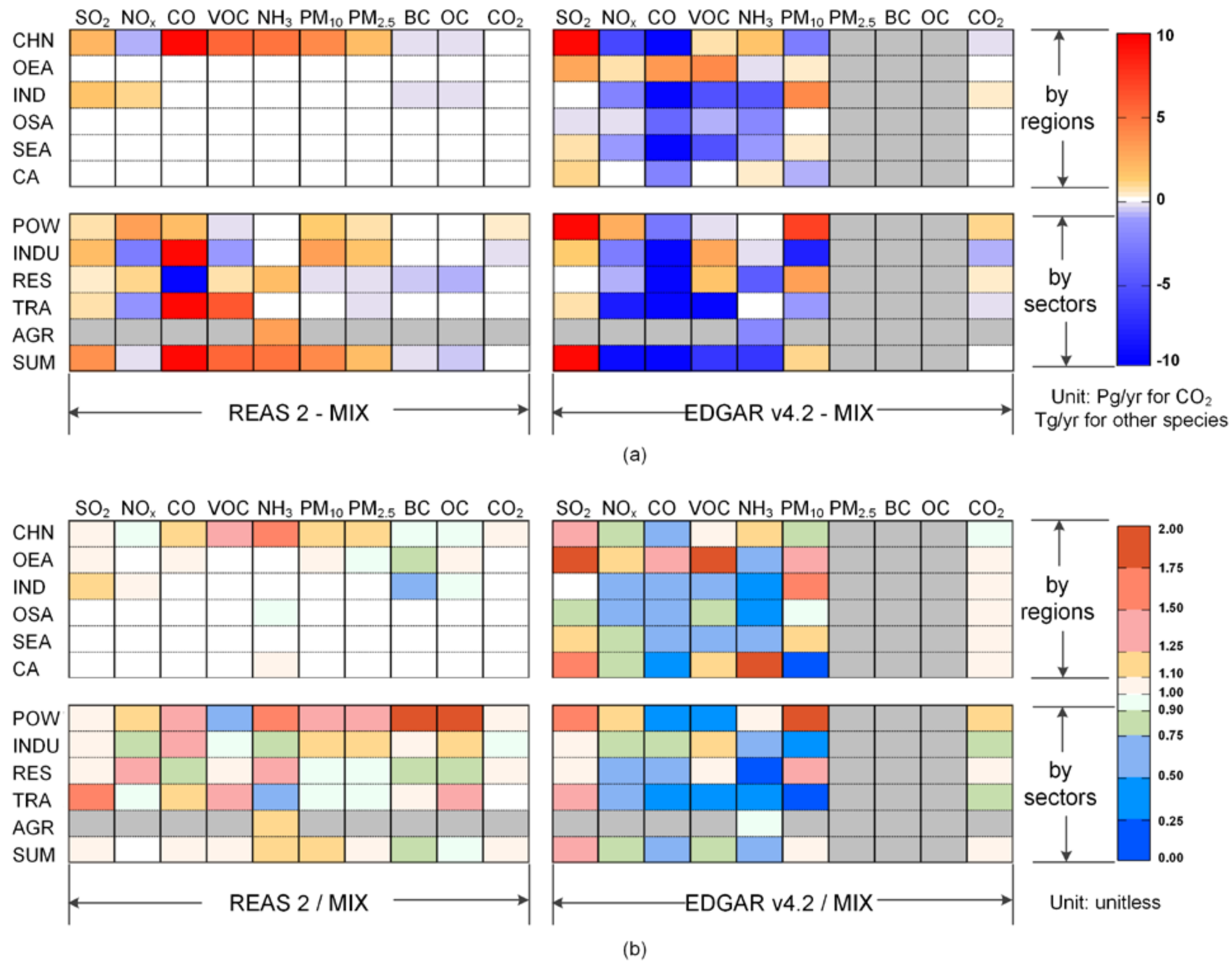


Figure 13. Inter-comparisons of emission estimates between MIX, REAS2 and EDGAR v4.2 by Asian regions and sectors. (a): absolute differences of emission estimates. (b): ratio of emission estimates. Grey shaded grids indicate that the comparison is not available due to absence of emission estimates in EDGAR v4.2. Abbreviations of Asian countries and regions are the same as in Fig. 6. 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.

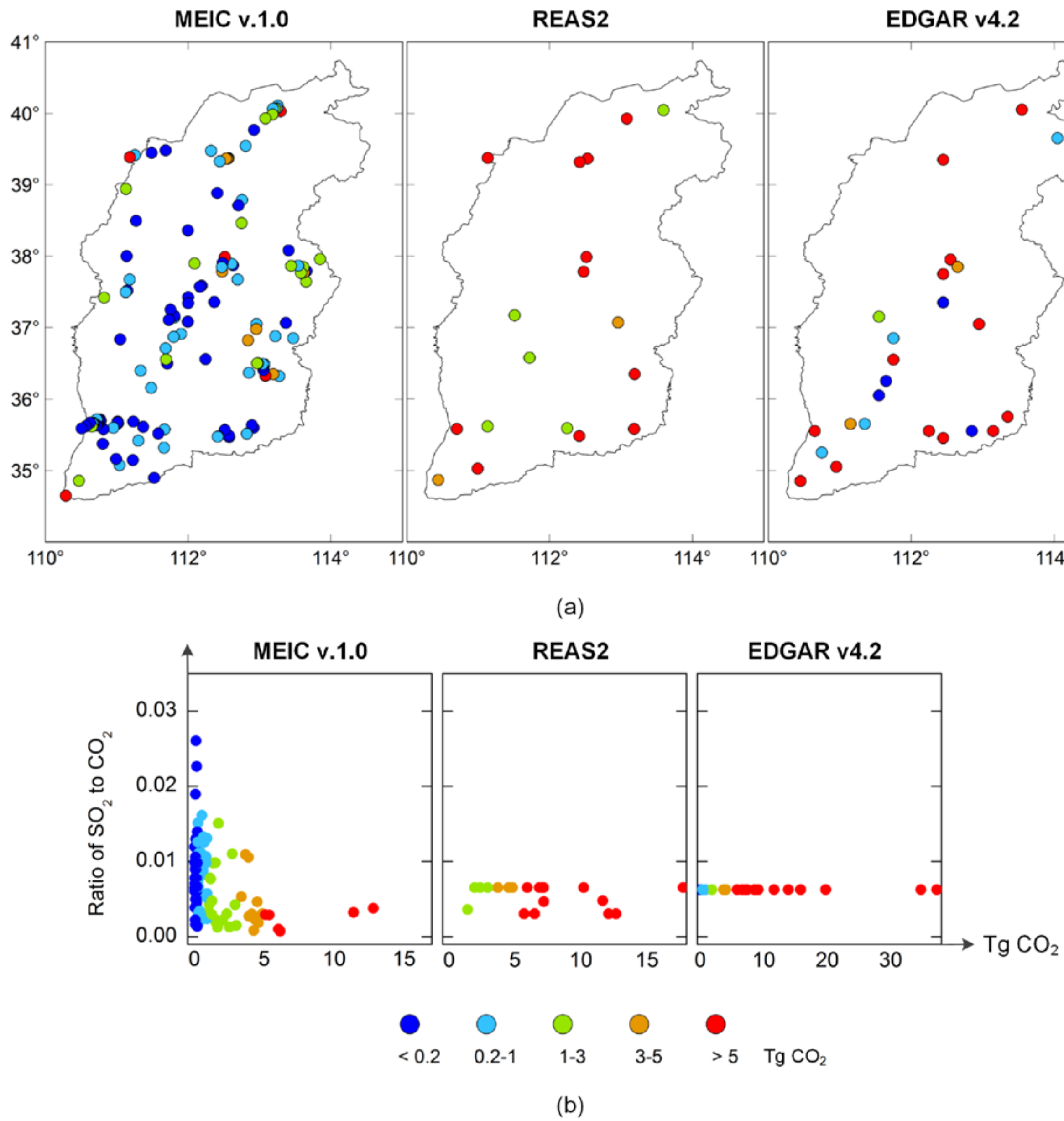


Figure 14. Comparison of 2008 power plants emission estimates between MEIC v.1.0, REAS2 and EDGAR v4.2 for Shanxi province, China. (a) Spatial distribution of CO_2 emissions, and (b) emission ratios of SO_2 to CO_2. CO_2 emissions are grouped by colors.

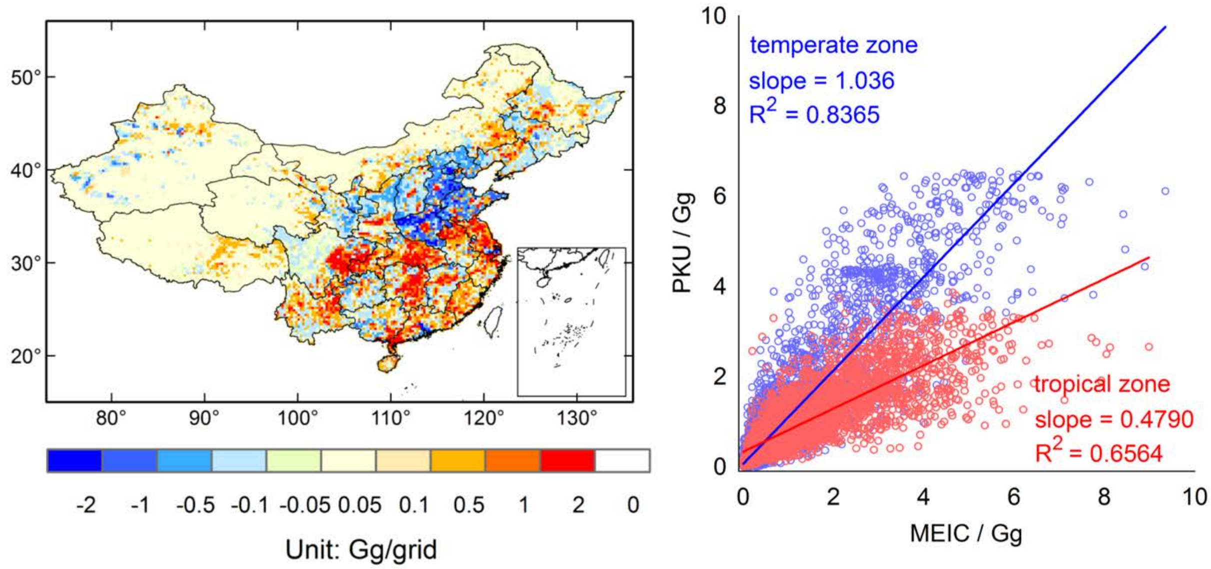


Figure 15. Comparisons of spatial distribution of NH_3 agricultural emissions between MEIC v.1.0 and PKU-NH_3. Provinces that included in tropical zones are: Fujian, Guangdong, Hainan, Guangxi, Guizhou, Hubei, Hunan, Yunnan, Sichuan, Jiangxi, Anhui, Zhejiang and Jiangsu. Other provinces are treated as temperate ones.