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.

General Comments:

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 (SO₂, 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-NH₃ was further used to replace MEIC emissions for NH₃ over China, given that PKU-NH₃ was developed with a process-based model that represented the spatio-temporal variations in NH₃ 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.

Specific comments

1. Topic: MIX: a mosaic Asian anthropogenic emission inventory for the MICS-Asia and the HTAP project \rightarrow 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" \rightarrow "The MIX inventory"

Response: Revised as suggested.

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

Response: Revised as suggested.

c) P34815-Line 6: "30 countries and regions in Asia" \rightarrow 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." \rightarrow 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 SO₂, +19 % for NO_x, +4 % for CO," \rightarrow Why only for SO₂ accurate to the first decimal behind the comma and not for all other substances (in particular for CO₂, 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" \rightarrow 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 0.25x0.255 are developed" \rightarrow 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 \times lat \times month) by species, sectors and years from those .nc files.

3. Section 1 Introduction

a) P34815-Line 26: "Wang et al., 2008" \rightarrow Please include also some more recent publications, such as Kulkarni et al. (2014)Atmos. Chem. Phys., 15, 1683-1705, 2015, <u>http://www.atmos-chem-phys.net/15/1683/2015/</u>, 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" \rightarrow Please explain how you then go to $0.25 \text{deg} \times 0.25 \text{deg}$.

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" \rightarrow 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," \rightarrow 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" \rightarrow 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" \rightarrow 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:" \rightarrow delete the first "species"

Response: Revised.

g) P34818-Line 29: "NMVOC emissions are speciated into model-ready inputs for two chemical mechanisms" \rightarrow 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." \rightarrow 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:" \rightarrow 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 NH₃ 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 (SO₂, 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-NH₃ was further used to replace MEIC emissions for NH₃ 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." \rightarrow 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" \rightarrow 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." \rightarrow 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)" \rightarrow 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," \rightarrow 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."

 \rightarrow 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" \rightarrow 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" \rightarrow Does MEIC include the NH_3 of non-agricultural sectors (e.g. from catalysts in road transport)?

Response: MEIC only includes NH₃ emissions for agriculture sector. Actually the PKU-NH₃ includes both agriculture and non-agricultural emissions for NH₃ and we incorporated these emissions in the MIX inventory. We have removed "agriculture" from the sentence.

6. Section 2.2.3 PKU-NH₃ for China

a) P34823-Line 20: "Open biomass burning was considered as a natural emission source and excluded in the MIX inventory." \rightarrow 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" \rightarrow When extrapolating in time, why also not extrapolating in space? Why is it not used for neighbouring countries?

Response: PKU-NH₃ 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" \rightarrow 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." \rightarrow 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" \rightarrow 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 $0.25^{\circ} \times 0.25^{\circ}$ 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." \rightarrow 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" \rightarrow how are CO₂ 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." \rightarrow 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-NH₃. 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 \rightarrow 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." \rightarrow 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 SO₂, NO_x, BC, and OC emissions and used REAS for CO, NMVOC, PM_{2.5}, PM₁₀, and CO₂ 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 $(I,k,m) \rightarrow$ 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" \rightarrow 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 $2 \rightarrow$ Except for the MEIC inventory, the data source for the CO₂ 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%" \rightarrow 28%

Response: Revised as suggested.

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

Response: Revised as suggested.

c) P34829-Line3: "contributing 59 % of the total SO₂ emissions" \rightarrow insert "Indian" between "total" and "SO₂ 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" \rightarrow 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" \rightarrow 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" \rightarrow 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)." \rightarrow 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 \rightarrow 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" \rightarrow 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" \rightarrow 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" \rightarrow 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." \rightarrow 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" \rightarrow 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" \rightarrow 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 SO₂ emissions (while we see only very small difference between SO₂ 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 SO₂ emissions. SO₂ 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." \rightarrow 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." \rightarrow 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" \rightarrow 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." \rightarrow 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" \rightarrow 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" \rightarrow 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" \rightarrow 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." \rightarrow 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," \rightarrow 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 \rightarrow 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 SO₂ 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 SO₂ and CO₂, respectively" \rightarrow 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 SO₂ and CO₂ (3% differences for SO₂, and 8% for CO₂) in 2008, implying similar estimates in energy consumption and emission factors in two inventories. Compared to MIX, REAS2 estimates lower emissions of NO_x, PM₁₀, 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" \rightarrow 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)," \rightarrow 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" \rightarrow 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." \rightarrow 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" \rightarrow 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)" \rightarrow 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" \rightarrow 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." \rightarrow 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." \rightarrow 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.

 \rightarrow this is definitely too vague. Please specify the 30 countries.

Response: Revised as suggested.

 \rightarrow 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.

 \rightarrow add a column with the year of data availability

Response: Revised as suggested.

 \rightarrow 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.

 \rightarrow 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

 \rightarrow 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

 \rightarrow 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.

 \rightarrow 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."

 \rightarrow 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: NH₃ agriculture emission estimates for China

 \rightarrow Please include for EDGARv4.2 also 2005, 2006, 2007 .

Response: Only EDGAR v4.2 estimates for 2008 were used for comparison. MASAGE_NH₃ 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) Figure 1: Domain and component of the MIX emission inventory

 \rightarrow replace the legend title with: MIX emission inventory components

Response: Revised as suggested.

b) Figure 3: NMVOC speciation scheme used in the MIX inventory development

 \rightarrow 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).

 \rightarrow please add here or in the supplement the NMVOC species list

Response: NMVOC species list was added in the supplement.

c) Figure 4: Emission distributions among sectors in Asia in 2010

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

Response: Corrected.

d) Figure 5: Emissions distributions by Asian regions in 2010

 \rightarrow 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 SO₂ (a) and CO (b)

 \rightarrow 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.

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

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

 \rightarrow add in each graph the y-x labels (Lat, Lon)

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

 \rightarrow try to use the same color scale for most of the pollutants

Response: In Fig. 10(a), we use the same color scale for SO₂, NO_x and NH₃. 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.

 \rightarrow 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.

 \rightarrow 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) Figure 11: Inter-comparisons in Asia

 \rightarrow use grey shaded area in order to avoid confusion with values lower than -10

Response: Revised as suggested.

 \rightarrow grey shaded grids

Response: Revised as suggested.

 \rightarrow Please specify EDGARv4.2

Response: Revised as suggested.

 \rightarrow please add "in", so that the text is "as in Fig.5"

Response: Revised as suggested.

 \rightarrow 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) Figure 12: Inter-comparisons in China, power plant sector

 \rightarrow 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.

 \rightarrow please put the legend of CO₂ emissions in the upper part of the graph, while add a new color scale for the SO₂ to CO₂ 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.

 \rightarrow please specify the x axis label of the bottom figure (CO₂ emissions?)

Response: The x-axis label of the bottom panel is CO₂ emissions, which is added now.

k) Figure 13: Inter-comparisons in China, NH₃ emissions

 \rightarrow what is this square?

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

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

Response: Revised as suggested.

 \rightarrow "Provinces that included in the tropical zones are", delete "the"

Response: Revised as suggested.