Response to comments of "MIXv2: a long-term mosaic emission inventory for Asia (2010-2017)" - #1

The work of Li et al. developed a new emission inventory, MIXv2, for 2010-2017, following an emission mosaic approach. By incorporating the best available regional emission inventories, MIXv2 now represents the state-of-the-art long-term emissions over Asia. Seven sectors and ten chemical species are included, providing the essential emissions input for both atmospheric and climate models. Gridded emissions at a high spatial resolution (0.1 degree) are publicly available to the community. This updated inventory data are critical to improve our understanding of the sources and atmospheric chemistry over Asia. The manuscript is in general well organized, with solid analyses and nice figures. My comments are mainly technical, as listed below.

Response: Thanks for the reviewer's positive and constructive comments. Below we have addressed the comment one by one and revised the manuscript accordingly.

Line 47 - 49: Can you provide 1-2 sentences on how the data can contribute to the community in the abstract?

Response: Thanks for the comment. We add the following statement by the end of the abstract:

This updated long-term emission mosaic inventory is ready to facilitate air quality and climate model simulations, as well as policy-making and associated analyses.

Line 126: not only regional emission inventories, but also two global inventories are used in the mosaic process. Please revise the sentence to "seven regional and two global emission inventories". Also, please revise accordingly in Line 139.

Response: We thank the reviewer's comment. We revise the sentence as suggested.

Line 153: can you elaborate more on how to develop the gridded CO2 emissions here?

Response: Thanks for the comment. We clarified the CO₂ emissions processing procedure as follows:

In detail, we firstly calculated the total CO_2 emissions of Japan by multiplying the CO_2 to NO_x ratios and JPN's NO_x emission estimates by sectors, then we developed the spatial proxies based on the NOx gridded emissions of JPN. Lastly, the calculated CO_2 emissions were allocated to each grid month by month.

Line 161: please specify the vegetation types in detail.

Response: We specified the vegetation types as below:

Wildfires of various vegetation types (including savanna, forest, peatland)

Line 369 – 370: To make it easier for readers to follow, can you put a short summary for Sect. 4 here?

Response: We thank the reviewer's comment. The following summary is added at the end of Sect. 2.5:

In short summary, generally consistent emission estimates and trends over Asia are found based on bottom-up and top-down comparisons in Sect. 4. Discrepancies persist, especially in regions like South Asia and SEA, as well as among species like BC and NMVOC.

Line 452 – 454: how about OEA, OSA and SEA? You only mentioned China and India in this paragraph.

Response: Thanks for the comment. Relatively small emission changes are estimated for OEA (-0.2 Tg) and OSA (+0.6 Tg). Significant emissions growth from power plants drives the total anthropogenic increase by 44%, and a 41% rise when considering additional open biomass burning for SEA. We add the analyses in the revised manuscript.

Line 586: Fig. S2 is for open biomass burning. Please add it to make the sentence clearer. **Response:** Revised to *Fig. S2 (featuring open biomass burning)*.

Line 649: The title of Figure 11 is ambiguous. Please change "Emissions (right columns)" to "2010 and 2017 emissions (right columns)".

Response: Revised.

Line 697: do you mean different data sources of inventories? Please rephrase this sentence.

Response: We revise the sentence as suggested:

More validations and revisions are needed to identify the reasons of the discrepancies and narrow down the gaps.

Line 724: temporal resolution to high temporal resolution, chemical speciation to detailed chemical speciation.

Response: Revised.

Response to comments of "MIXv2: a long-term mosaic emission inventory for Asia (2010-2017)" - #2

This article has developed the MIXv2 Asian emission inventory under the framework of the Model Inter- Comparison Study for Asia (MICS-Asia) Phase IV, the emissions for anthropogenic and biomass burning sources covering 23 countries and regions has been estimated. It is a important database. However, the formal and the organizational structure of the paper needs to be reconsidered. It reads more like a database usage instruction. I suggest the authors can resubmit this paper after a major revision.

Response: We thank the reviewer's comments. We agree with the reviewer that as an emission inventory paper, it's important to describe in detail how the data is developed and what's the big implications to the community. In our paper, we have performed comprehensive analyses along with data description in Sect. 2 - Sect. 4. In Sect. 2, the mosaic methodology and component emission inventories are detailed described. Besides, we dig into the data and have made comprehensive analyses on the emission changes during the investigated period and their driving forces in Sect. 3. Uncertainties are analyzed in Sect. 4.

- The most up-to-date emissions in 2017 by Asian regions are investigated in Sect. 3.1. For each chemical species (including CO_2 and 9 air pollutants), we analyzed their emission changes from 2010 - 2017 by sectors and regions in Sect. 3.2. Rapid emission reductions for SO₂ and CO over Asia are found as a combined result of decreasing shares of China, and increasing contributions from India and Southeast Asia, also showing distinct sectoral variations. The policies driving the emission changes are analyzed.

- In Sect. 3.3 – Sect. 3.4, the seasonality and spatial distribution of emissions by regions and sectors are investigated. We included the open biomass sector in the analyses to have a more comprehensive picture to support in-field measurements.

- In Sect. 3.5, we addressed the distribution of Volatile Organic Compounds (VOCs) chemical species in 2010 and 2017, and their emission changes. These analyses can be important to support air quality research and policy assessment on ozone abatement.

- In Sect. 4, we compared our estimates with both other inventories and top-down estimates and pointed out the future directions of inventory development over Asia.

We believe the above analyses not only describe how to use the data, but also provide insights on important scientific questions on the historical emission trends and future air pollution control stemming from the inventory data.

Below, we have responded to all the reviewer's comments, addressed the reviewer's questions, and revised the manuscript accordingly. All responses and manuscript changes are highlighted in blue color.

1. Line 48-49: It may be inappropriate to place this address in the abstract.

Response: Thanks for the comment. We remove the data download address in the abstract in the revised manuscript.

2. The introduction should be further optimized. It is more like a briefly introduction for the MIX Asian inventory in a project report.

Response: Thanks to the reviewer's comment. We re-organize the introduction section as below, emphasizing the background and scientific questions to resolve in developing a long-term mosaic emission inventory over Asia. Revised parts are shown in *Italic*.

Air pollutants emitted from both anthropogenic and natural activities have caused severe impacts on human health, ecosystems, and climate over Asia (Adam et al., 2021; Geng et al., 2021; Takahashi et al., 2020; Wong et al., 2008; Xie et al., 2018). Over the last two decades, the emerging ozone pollution and haze events across Asia have got extensive attention from the government (Anwar et al., 2021; Feng et al., 2022; Zheng et al., 2018). Tremendous efforts have been made since 2010 continuously to improve air quality and protect human health. The effects of these policies on emission abatement need to be updated in inventories, to address the regional and global issues of air quality and climate change. Therefore, a long-term emission inventory plays key roles in historical policy assessment, and future air quality and climate mitigation.

Consistent greenhouse gas emissions are crucial for climate-air quality nexus research and policymaking (Fiore et al., 2015). Carbon dioxide (CO₂) is co-emitted with many air pollutants which are contributors of ozone and particulate matter, further changing climate through forcings of Earth's radiation budget (Fiore et al., 2015). Previous studies have emphasized the importance of air pollution mitigation and climate change (Jacob and Winner, 2009; Saari et al., 2015), as recently summarized by the Synthesis Report of the IPCC Sixth Assessment Report (IPCC: Intergovernmental Panel on Climate Change, report available at https://www.ipcc.ch/report/sixth-assessment-report-cycle/). Given the common sources of CO2 and air pollutants, it's important to quantify their emissions distribution in a self-consistent way to assess the co-benefits and pathways to cleaner air and carbon neutrality (Klausbruckner et al., 2016; Phillips, 2022; von Schneidemesser and Monks, 2013).

Emissions over Asia since 2010 are quantified in recent studies. Kurokawa et al. (2020) developed an anthropogenic emission inventory over Asia for 1950-2015, REAS (the Regional Emission inventory in ASia), covering power plants, industry, residential, transportation and agricultural sources. Emissions of both air pollutants and CO2 are estimated in REAS. Based on the Community Emissions Data System (CEDS), McDuffie et al. (2020) developed a global anthropogenic emission inventory covering major air pollutants over 1970-2017. Global emissions for air pollutants are estimated under the HTAPv3 (Task Force on Hemispheric Transport of Air Pollution) project for 2000-2018 for air pollutants by integrating official inventories over specific areas including Asia (Crippa et al., 2023). These regional / global emissions are estimated with limited updates of country-specific or even localized information. Following a mosaic approach, the first version of MIX Asian inventory (MIXv1) was developed to support the Model Inter-Comparison Study for Asia (MICS-Asia) Phase III projects, by incorporating five regional emission inventories for all major anthropogenic sources over Asia, providing a gridded emission dataset at a spatial resolution of 0.25 degree for 2008 and 2010. The mosaic approach has been proved to increase the emission accuracy and model performance significantly by including more local information (Li et al., 2017c). A profile-based speciation scheme for Non-Methane Volatile Organic Compounds (NMVOCs) was applied to develop model-ready emissions by chemical mechanisms, which reduced the uncertainties arising from inaccurate mapping between inventory and model species (Li et al., 2014; Li et al., 2019b). Specifically, MIXv1 advances our understanding of emissions and spatial distributions from power plants and agricultural activities through a mosaic of unit-based information and a process-based model.

However, it's difficult to develop consistent emissions over Asia for a long period using the mosaic approach because of the lack of available regional inventory data. Within the MICS-Asia community, developers of regional inventories have been endeavoring to extend their emission inventories to the present day since Phase IV. Through intensive collaboration and community efforts, we now have a complete list of available regional emission inventories covering major parts of Asia, and are able to combine them to produce a new version of MIX for 2010-2017. MICS-Asia is currently in its fourth phase, MICS-Asia IV, which aims to advance our understanding of the discrepancies and relative uncertainties present in the simulations of air quality and climate models (Chen et al., 2019; Gao et al., 2018; Itahashi et al., 2020; Li et al., 2017c). A critical component of the project is ensuring that emission inventories remain consistent across various atmospheric and climate models. In support of MICS-Asia IV research activities and related policy-making endeavors, we developed MIXv2, the second version of our mosaic Asian inventory. MIXv2 combines the best available state-of-the-art regional emission inventories from across Asia using a mosaic approach. This inventory is expected to enhance our capabilities to assess emission changes and their driving forces, and their impact on air quality and climate change, thus providing valuable insights for decision-makers and stakeholders. CO2 emissions are estimated based on the same emission inventory framework as the short-lived air pollutants, and further integrated into MIXv2 following the mosaic methodology.

MIXv1 has been widely applied to support scientific research activities from regional to local scales (Geng et al., 2021; Hammer et al., 2020; Li et al., 2019a; Li et al., 2017c). Compared to MIXv1, MIXv2 has the following updates to better feed the needs of atmospheric modelling activities:

- advances the horizontal resolution of the gridded maps from 0.25 to 0.1 degree
- incorporates up-to-date regional inventories from 2010-2017
- provides emissions of open biomass burning and shipping, in addition to anthropogenic sources
- develops model-ready emissions of SAPRC99, SAPRC07 and CB05

Methods and input data are described in Sect. 2. Emissions evolution and their driving forces, seasonality, spatial distribution, NMVOC speciation and inventory limitations are analyzed and

discussed in Sect. 3. Sect. 4 compares the MIX data with other bottom-up and top-down emission estimates. Concluding remarks are provided in Sect. 5.

3. How to ensure the data consistency when multiple emission inventories were used?

Response: Thanks for the comment. We agree with reviewer that it's important to ensure the consistency when multiple emission inventories are used. The consistency of emissions data in MIXv2 is ensured in three aspects: source aggregation, spatial distribution, and VOC speciation.

Firstly, we implement a consistent source definition system to aggregate sources from regional emission inventories into the final emission mosaic. The source mapping matrix, outlined in Table 2, establishes the correlation between IPCC codes and subsectors/sectors of MIX.

Secondly, the consistency of emissions spatial distribution during emissions mosaic between different inventories are ensured carefully. In India, we integrated the ANL-India emissions for NO_x, SO₂, and CO₂ for pointed power plants, and emissions from REAS for other species. To keep the consistency of spatial distribution, we developed spatial proxies based on the CO₂ emissions from ANL-India, and re-located REAS emissions for other species.

Thirdly, a uniform VOC speciation framework is employed across all component emission inventories, encompassing both anthropogenic and open biomass burning sources. A composite profile database is established by combining local source profiles with the SPECIATE v4.5 database. For each source category in regional emission inventories, we assign corresponding source profiles to speciate total VOCs into individual species. Furthermore, all individual species are grouped into three chemical mechanisms.

We add the above illustration on how to ensure the data consistency in emissions mosaic in revised manuscript (Sect. 2.2).

4. Line 366: The uncertainties of MIXv2 can not be evaluated, can we find a better way to solve this problem?

Response: We thank the reviewer's constructive comment. It's always difficult to quantify the uncertainties for a mosaic emission inventory such as MIXv2. To better address the uncertainties by Asian regions, we add a new table illustrating the uncertainty estimates by Asian regions in previous studies. The uncertainty ranges are quantified based on propagation of uncertainty or Monte Carlo simulations. In the main text, we add Table 3 and following analyses on uncertainty estimates by species in the revised manuscript.

In regard of anthropogenic sectors, the precision of emission estimates for SO₂, NO_x, and CO₂ is higher than that of other pollutants, owing to the minimal uncertainties associated with power plants and large industrial facilities. This is particularly notable in the case of MIXv2, where uncertainties are even lower due to the integration of unit-based power plant information for both China and India. While uncertainties for CO and NMVOC are comparable, they are higher than

those for SO₂, NO_x, and CO₂, because of substantial emission contributions from biofuel combustion. Emissions for particulate matter (especially BC and OC) tend to be more uncertain compared to trace gases, primarily due to the low data reliability of activity rates and emission factors related to residential biofuel combustion. The need for more detailed information at the technology or facility level in regions, such as India, OSA, and SEA, is crucial to narrow down the overall uncertainties in Asia in the future. For open biomass burning, previous investigations have estimated low uncertainty ranges for species like CO, NMVOC, and OC, while more further analyses are in urgent need. In this work, we conducted uncertainty analyses qualitatively by comparing the MIXv2 estimates with other bottom-up inventories and those derived from satellite retrievals in Sect. 4 (Li et al., 2018).

Table 3. Uncertainties in emission estimates by Asian regions (95% confidence intervals if not noted; unit: %)

9

5. Line 432-433: Power plants are the major driving factor for emissions reduction of NOx. So how about on-road transportation sources? As you mentioned the anthropogenic emissions in OSA and SEA was also mainly driven by the vehicle growth.

Response: Thanks for the comment. We clarified the emission changes, and their driving forces as follows:

NO_x emissions show increasing-decreasing-increasing trend for 2010-2017, with a peak in 2012. This trend is a combination of significant power plant emissions reduction (-22% from 2010-2017), and emissions increase from industry (+4%) and transportation (+6%). As estimated, China's emissions for all anthropogenic sectors dropped by 4.6 Tg (-17%) from 2010-2017, along with 2.6 Tg (+38%) emissions growth from India and 0.9 Tg (+19%) from SEA. As a result, China's contribution decreased from 63% to 54%, and Indian share grew from 16% to 22% (anthropogenic, Fig. 5a).

6. Line 568-569: Can you explain why the small monthly variations were found in Indian and OSA?

Response: Thanks for the reviewer's comment. Indian and OSA emissions show small monthly variations compared to other Asian regions. This pattern is attributed to the predominant role of the residential sector on emissions for the investigated species, as depicted in Fig. 7. The minimal seasonal variations in surface temperature within the tropical climate of India and OSA contribute to the overall stability in monthly residential emission patterns. We clarified the reason in the revised manuscript.

Response to comments of "MIXv2: a long-term mosaic emission inventory for Asia (2010-2017)" - #3

This paper is a welcomed update of the MIX emission inventories developed within the MICS-Asia framework. This is a great example of an inclusive and collaborative effort that brings together emission experts from various countries in Asia and combines best available country level information to produce an Asia wide consistent set of emissions. This current work extends this inventory in time from 2010 to 2017 and increases the spatial resolution (from 25 km to 10 km). These are significant advancements. This new inventory will support many modeling and analysis studies.

The previous inventory has been widely used and been a great contribution to air quality studies in Asia. However, there was a clear need to update in time the inventory to support further studies as emissions in this region continue to change rapidly over time.

The paper is well written and presents sufficient details to understand the methodologies used. It also presents useful information regarding the trends in species and sectors in various parts of Asia.

A significant contribution.

Response: We greatly appreciate the recognition of the reviewer. We hope the updated emission inventory can support more air quality studies in Asia. Thanks for the comment.

Response to community comments of "MIXv2: a long-term mosaic emission inventory for Asia (2010-2017)" by Gabriele Pfister (TOAR II Steering Committee Member)

This manuscript was submitted to ACP as part of the TOAR-II Community Special Issue, https://doi.org/10.5194/egusphere-2023-2283, 2023 Preprint. Discussion started December 8, 2023; discussion closes January 19, 2024. I, Gabriele Pfister (National Center for Atmospheric Research, Boulder CO) am providing this comment as a member of the TOAR II Scientific steering committee.

Owen Cooper, TOAR Scientific Coordinator of the TOAR-II Community Special Issue or a member of the TOAR-II Steering Committee, will post comments on all papers submitted to the TOAR-II Community Special Issue, which is an inter-journal special issue accommodating submissions to six Copernicus journals: ACP (lead journal), AMT, GMD, ESSD, ASCMO and BG. The primary purpose of these reviews is to identify any discrepancies across the TOAR-II submissions, and to allow the author teams time to address the discrepancies. Additional comments may be included with the reviews. While O. Cooper and members of the TOAR Steering Committee may post open comments on papers submitted to the TOAR-II Community Special Issue, they are not involved with the decision to accept or reject a paper for publication, which is entirely handled by the journal's editorial team.

General Comments

This paper presents an update to the publicly available MIXv1 Asian emissions inventory as described in Li et al., (2017) and which has been widely used for scientific research. The MIXv2 effort has combined seven available regional Asian inventories and two global inventories into one consistent data set, which is a significant effort. Amongst other updates, MIXv2 combines up-to-date regional emission inventories, provides higher spatial resolution (from 0.25 degrees to 0.1 degrees), extends the time period from 2008-2010 in v1 to 2010-2017 and also includes open biomass burning and shipping emissions. MIXv2 is also being provided in different chemical specifications which will help modelers to map them more easily to chemical speciations of their choice.

There is no question about the need for improved, state-of-the art emission inventories to characterize changes over time and space and reduce uncertainties in simulating air quality and climate. TOAR studies heavily rely on both observations but also modeling studies and specifically for the latter accurate emission inventories and their trends in time and space are essential input. Having a consistent multi-year emission inventory will also provide a context for the analysis of ozone observations and trends. As such this paper and the provided emission data set make a significant contribution to TOAR, to atmospheric chemistry research in general but also to policy making. The inclusion of CO2 emissions in addition to air pollutants will be a great benefit to studying the combined effects of air quality and climate change.

The paper presents a comprehensive description of the underlying emission inventories and the methods used to generate the final MIXv2. In addition, it provides a detailed analysis of the derived MIXv2 emission drivers and trends including analysis of individual sectors, as well as a

comparison to other inventories. The paper shows that Asian emissions are a large part of global anthropogenic emissions. Changes in emissions over 2010-2017 vary by region and sector and the analysis shows a general latitudinal shift in emissions but with different trends for different species. This will lead to significant changes in ozone production over Asia and further impacts the global tropospheric ozone budget. It will be important to have follow-up modeling studies exploring these impacts and placing them into relation to observed changes.

The paper includes a ton of information, potentially too much for the casual reader but no question important information for anyone using MIXv2. However, the authors might want to consider moving some of the Tables and Figures to the Supplement to make it easier to navigate through the paper.

Response: We greatly appreciate the positive comments and recognition from Dr. Gabriele Pfister, on behalf of the TOAR II steering committee. We believe the updated emission inventory and our analyses on emission trends will make a significant contribute to TOAR, as well as atmospheric science community and policy making.

We have addressed all the detailed comments below and revised the manuscript accordingly.

Other comments that I hope the authors find helpful

Line 75: It might be helpful to be more specific what type of process-based model has been used

Response: Thanks to the reviewer's comment. We have specified the process-based model as follows:

Specifically, MIXv1 advances our understanding of emissions and spatial distributions from power plants through a mosaic of unit-based information, and agricultural activities based on a process-based model which parameterized the spatial and temporal variations of emission factors for NH₃.

Section 2.2: Maybe state right upfront that seven regional and two global inventories have been combined

Response: We thank the reviewer's comment. The sentence has been revised to:

In brief, nine regional and two global emission inventories were collected and integrated into a uniform format.

Line 163: I suggest to mention in the introduction already that anthropogenic sectors are reported on a monthly basis while open biomass emissions are available with daily resolution

Response: Thanks to the reviewer's comment. We add the following statement in Sect. 2.1 in the revised manuscript:

Monthly emissions between 2010 - 2017 are allocated to grids at 0.1×0.1 degree. Open biomass burning emissions are also available with daily resolution upon request.

Section 2.5: I understand what the authors try to say about inconsistencies at borders, but I would

acknowledge that there is a general inconsistency between the different countries given that the regional inventories rely on different reporting and approaches.

Response: We appreciate the reviewer's comment. We acknowledge that it's a general inconsistency of accuracy between countries as a mosaic emission inventory. In Sect. 2.5, we add the following statement in the revised manuscript:

We also acknowledge the general inconsistency of uncertainty levels between countries where different inventories are used following various data sources and approaches.

Section 2.5: The authors provide brief discussions on the limitations and potential uncertainties of MIXv2 but I would add that one other limitation, specifically for air quality studies, is also the lack of a diurnal cycle as well as stack height information for point sources. I suggest adding this to the discussions as potential future development needs.

Response: Thanks for the comment. This is really a good point. We add the following analyses in Sect. 2.5:

For air quality simulation purposes, the lack of diurnal variations and vertical distribution is another limitation when applying MIXv2 data directly. Development of Asia-specific temporal and vertical profiles is important to improve the model simulation performance in the future.

Line 404: While listed in the caption for S1 I would also include in the text what inventories are used for global reference.

Response: We clarified the data sources in the revised manuscript:

Emissions over Asia are derived from MIXv2, and emissions over the rest of the world are estimated by EDGARv6.

Section 3.2: I would be cautious defining the emission changes in open biomass burning as "trend" given the large year-to-year variability in this sector.

Response: Thanks for the comment. We agree with the reviewer that it's not appropriate to use "emissions trend" for open biomass burning. We have reviewed the whole paper and corrected such statement carefully.

Line 554: Maybe rephrase to: *Monthly variations of emissions, which are highly sector dependent, are estimated by MIXv2.*

Response: Revised.

Line 623-625: This sentence reads confusing.

Response: Thanks for the comment. The sentence has been revised as below:

Alkenes and Alkynes show a stable trend, reflecting the combined results of emission reduction in residential and growth in industry and transportation sectors.

Section 4: Please be specific if the comparisons include open biomass burning or not. E.g. I assume Figure 12 only looks at anthropogenic sectors?

Response: We have specified the source types as anthropogenic sectors in the caption of Figure 12.

Line 673: To provide **a** potential ... **Response:** Revised.

Line 682: I assume with MIX the authors mean MIXv2? **Response:** Revised to MIXv2.