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 SO_2 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 (CO_2) 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, SO_2, and CO_2 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 SO_2, NO_x, and CO_2 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 SO_2, NO_x, and CO_2, 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: %)

Regions, Anthropogenic or Open Biomass	NO_x	SO_2	CO	NMVOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC	CO_2	Year	Reference
China, Anthropogenic						±91	±107	±187	±229		2005	Lei et al. (2011)
	-13~37	-14~13				-14~45	-17~54	-25~136	-40~121		2005	Zhao et al. (2011)
			-20~45								2005	Zhao et al. (2012)
	±31	±12	±70	±68		±132	±130	±208	±258		2006	Zhang et al. (2009)
		-16~17						-41~80	-44~92		2010	Lu et al. (2011)
	-15~35	-15~26	-18~42			-15~54	-15~63	-28~126	-42~114		2010	Zhao et al. (2013)
	±35	±40	±73	±76	±82	±83	±94	±111	±193	±19	2015	Kurokawa et al. (2020)
	-26~34	-22~25	-31~41	-32~56							2015	Sun et al. (2018)
					-14~13						2015	Zhang et al. (2017)
										-15~30	2017	Shan et al. (2020)*
India, Anthropogenic		-15~16						-41~87	-44~92		2010	Lu et al. (2011)
	±35	±41	±136	±115	±111	±120	±151	±133	±233	±27	2015	Kurokawa et al. (2020)
								±33			2011	Paliwal et al. (2016)
Japan, Anthropogenic	±32	±34	±45	±63	±103	±68	±74	±58	±100	±13	2015	Kurokawa et al. (2020)
OEA, Anthropogenic	±60	±38	±67	±63	±94	±69	±85	±82	±168	±19	2015	Kurokawa et al. (2020)
OSA, Anthropogenic	±34	±40	±87	±73	±93	±96	±112	±124	±211	±19	2015	Kurokawa et al. (2020)
SEA, Anthropogenic	±38	±46	±124	±86	±115	±125	±155	±161	±232	±25	2015	Kurokawa et al. (2020)
China, Biomass	-37~37	-54~54	-4~4	-9~9	-49~48	-7~6	-13~1	-61~61	-20~19	-3~3	2012	Zhou et al. (2017)
SEA, Biomass	±23	±30	±20	±18	±10			±20	±31	±15	2010	Shi and Yamaguchi (2014)

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.