Response to the comment from Anonymous referee #4

This study aims to provide a comprehensive inventory of chlorine emissions by updating the previous inventory to include data from a more recent year and expanding the range of species considered, as well as the number of anthropogenic sources. This paper provides valuable data on chlorine emissions in China. Despite the considerable effort involved, I suggest that the authors enhance the foundational support for the new additions and provide more evidence to demonstrate significant advancements in emission inventory calculation methods. Strengthening these aspects will greatly improve the reliability and impact of the study. My main suggestions for improving innovation and reducing uncertainty are as follows:

[Comment 1]: A significant portion of the chlorine emission sources in the study are indoors. However, the exchange of gases between indoor and outdoor environments is influenced by various factors, such as building structures, climate conditions, and building usage across different regions of China. It is recommended that the authors fully consider the impact of these factors.

Response: Thank you for your valuable suggestion. Many previous studies demonstrated that the indoor air pollutants can significantly affect outdoor air quality through indoor-outdoor exchange, but quantifying this impact is challenging and remains uncertainties (Santiago et al., 2022; Alsamrai et al., 2024). These uncertainties arise from various factors, including the complexity of emission sources, variability in building ventilation systems, and differences in geographic and climatic conditions. Here, to simplify, we assumed that all the indoor chlorine gases (e.g. environmental disinfection, indoor swimming pool) released into the atmosphere due to the rapid air exchange between indoor and outdoor during the ventilation process (Huang, 2012; Tang, 2003). Although this treatment may overestimate their impact on ambient concentrations, our modeling results show general agreement with the observation. We have discussed this limitation in the manuscript.

Revision in the updated manuscript:

1) Line 671-676: "(2) Complexity of indoor-outdoor air exchange: The study simplifies the treatment of some indoor chlorine emissions (e.g., environmental disinfection) by assuming rapid air exchange between indoor and outdoor environments. However, the actual exchange rate can vary significantly due to differences in building structures, ventilation systems, and climatic conditions. This variability can lead to uncertainties in estimating the impact of indoor emissions on outdoor air quality. Future research should incorporate more detailed assessments of these factors to improve the accuracy of emission estimates. "

Reference:

Alsamrai, O., Redel-Macias, M. D., Pinzi, S., and Dorado, M. P.: A Systematic Review for Indoor and Outdoor Air Pollution Monitoring Systems Based on Internet of Things, Sustainability, 16, 4353, 2024.

Huang, Y.: Study of the Natural Ventilation Strategy of Hospital Clinic Waiting in Lingnan Regions, M.S. thesis, South China University of Technology, China, 125 pp., 2012.

Tang, J.: Design of the Air-Conditioner for Chamber Indoor Swimming Pool, Mechanical and Electrical Equipment, 5, 17-20, 2003 (in Chinese).

Santiago, J. L., Rivas, E., Buccolieri, R., Martilli, A., Vivanco, M. G., Borge, R., Carlo, O. S., and Martín, F.: Indoor-outdoor pollutant concentration modelling: a comprehensive urban air quality and exposure assessment, Air Qual Atmos Health, 15, 1583-1608, 10.1007/s11869-022-01204-0, 2022.

[Comment 2]: Adding new emission sources or further subdividing the emission sources does not necessarily lead to better results. Inaccurate descriptions of the spatial and temporal distribution of sources can increase uncertainty. Given chlorine's high atmospheric reactivity, spatial and temporal distribution is crucial. For example, point

sources and area sources of chlorine emissions have significantly different atmospheric chemical impacts. The study's simplistic allocation of some point source emissions to entire areas may be inappropriate; additionally, the temporal distribution of pesticide use is influenced by crop types, climate, and geographical conditions, which the study does not detail.

Response: Thanks for your valuable suggestion. Firstly, regarding the spatial allocation of some point sources, such as residential coal combustion and biomass household burning, we treated them as areas sources and used the total/urban/rural population as a proxy for disaggregating emissions. Although this treatment may induce uncertainties, it is commonly adopted for those point sources without available specific location information in the development of national emission inventories in previous studies. As summarized in Table S4 by Li et al. (2017), for the well-known emission inventories of MEIC (http://meicmodel.org.cn), REAS2 (Kurokawa et al., 2013), PKU-NH_3 (Huang et al., 2012), ANL-India (Lu et al., 2011), CAPSS (Lee et al., 2011), total/urban/rural population served as a proxy for spatial allocations of some point sources (residential, industrial, solvent use, agriculture, etc.).

Secondly, regarding the temporal distribution of pesticides, we used the monthly pesticide production as the temporal allocation factors. The temporal variation of pesticide production can somewhat reflect the monthly emission characteristics of pesticide usage. We have provided this detailed information in Section 2.4.

Overall, there is a long way to go to reduce the uncertainties of air pollutant emission inventories currently. Further analysis and research are needed to reduce these uncertainties if specific sources information are available and novel estimation methods are proposed in the future. We have discussed the limitation of this inventory in the manuscript.

Revision in the updated manuscript:

1) Line 676-682: "(3) Spatial and temporal distribution of emissions: The study employs generalized proxies, such as population distribution, for spatial allocation of emissions from point sources like residential coal combustion and biomass burning,

This approach may not accurately capture the localized nature of emissions and their impacts. Additionally, the temporal allocation of pesticide emissions based on monthly production does not fully account for regional variations in crop types and climate. More precise data on spatial and temporal allocation factors, such as detailed information on the operating scales of point sources, are needed to reduce uncertainties and enhance the accuracy of the inventory. "

Reference:

Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., and Zhang, H.: A high‐resolution ammonia emission inventory in China, Global Biogeochem Cy, 26, 2012.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, Atmos Chem Phys, 13, 11019-11058, 2013.

Lee, D., Lee, Y.-M., Jang, K.-W., Yoo, C., Kang, K.-H., Lee, J.-H., Jung, S.-W., Park, J.-M., Lee, S.-B., and Han, J.-S.: Korean national emissions inventory system and 2007 air pollutant emissions, Asian Journal of atmospheric environment, 5, 278-291, 2011.

Li, M., Zhang, Q., Kurokawa, J., Woo, J. H., He, K. B., Lu, Z. F., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y. F., Hong, C. P., Huo, H., Jiang, X. J., Kang, S. C., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos Chem Phys, 17, 935-963, 10.5194/acp-17-935-2017, 2017.

Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, Atmos Chem Phys, 11, 9839-9864, 2011.

[Comment 3]: The authors should provide stronger evidence to show that this more

detailed method significantly reduces uncertainty. Some of these more detailed activity amounts and emission factors have limited sources. For example, the quantity of disinfectant utilized, which is cited from only one hospital, may not be sufficiently convincing.

Response: We appreciate your valuable suggestion. Firstly, the quantity of disinfectant utilized in a hospital to estimate emissions in this study is based on the survey results of Sun et al. (2007), with adjustments for annual variations. It is reported that the amount of disinfectant used in five hospitals (not only one hospital) in Taizhou ranged from 1997 to 2704 L, with an average of 2329.2L. The small deviation among these five hospitals shows that this average value is generally representative for the emission estimation in this study. Nevertheless, specific dataset about the quantify of disinfectant utilized in each hospital in China is warranted to estimate chlorine emissions accurately, but corresponding survey data are scarce currently. We have discussed this limitation in the manuscript.

Revision in the updated manuscript:

1) Line 666-671: "The potential limitations of the ACEIC emission inventory that require further refinement are summarized as follows. (1) Limited sources for some activity data and emission factors: For example, for estimating chlorine emissions from hospital disinfection, the average disinfectant usage derived from five hospitals may not accurately reflect the variability in usage across different regions and facility types. There is a need for further localized investigation on activity data and experimental studies on emission factors. This is particularly important for sectors such as waste treatment and usage of chlorine-containing disinfectant, where more localized observation data is needed to improve the accuracy of emission estimates. "

Reference:

Sun, Y., Tian, F., Sun, Z., Jiang, W., Wu, B., Xu, Z., and Gu, J.: Investigation and Analysis of the Current Condition of Applying Disinfectants in City Hospitals of Taizhou, Modern Preventive Medicine (in Chinese), 4738-4741, 2007.

Overall, my view aligns with previous reviewers that the methodologies and details of this study could benefit from further refinement. I encourage the authors to pursue additional investigation and experimental support.

Response: Thanks for your valuable comment. We recognize that this chlorine emission inventory still has some limitations which may induce uncertainties. In the future, we will continue to refine and update this inventory with further investigation and experimental support.