Anonymous Referee #1

This work compiled a comprehensive anthropogenic chlorine emission inventory for China. It improved the authors' previous ACEIC inventory by including the sources from cooking, usage of disinfectant, and pesticide. The paper provides valuable data for chlorine emissions in China. Some parts of the calculation and discussion are not very clear and need improvement.

Response: We express our gratitude to the referee for offering a reflective and comprehensive evaluation of our paper. The referee's suggestions have significantly contributed to the enhancement of this manuscript. Below, we present point-by-point responses to the referee's remarks and summarize the modifications that have been implemented in the revised manuscript.

Major comments:

[Comment]: 1. I would suggest to rearrange the introduction to make it clearer. Paragraph 2-5 have some overlaps and inconsistent, making the logic confusing.

Response: Thanks for your valuable suggestion. We agreed and have rearranged the introduction to provide a clearer presentation. Please see Section 1 (Introduction) in the revised manuscript.

[Comment]: 2. The most significant finding of this work that differs from previous studies may be a large source of HOCl and Cl_2 from the usage of chlorine-containing disinfectants. It seems that the estimate of this part of emissions (section 2.2.6) assumes that chlorine gases volatilized from the water will be directly released to the atmosphere. However, many water treatment plants, hospitals, and swimming pools are indoors, some of the waste gases are also treated. This needs more discussion.

Response: Thanks for pointing out this important issue. Currently, there are no specific requirements or applicable standards for the management of waste gas from disinfection

in China. Consequently, a majority of waste gases are released without systematic regulation. Cl_2 and HOCl released from the usages of chlorine-containing disinfectants are known for their pungent odors, which can pose risks to human health. As a result, indoor disinfection procedures, such as those in hospitals and indoor swimming pools, require meticulous attention to ventilation (Huang, 2012; Tang, 2003; the standard GB 15982-2012). During the ventilation process, there is a rapid exchange of indoor and outdoor air, resulting in the release of chlorine gases into the atmosphere. We have added this discussion in Section 2.2.6 as follows: "Cl_2 and HOCl released from the usages of chlorine-containing disinfectant are known for their pungent odors, which can pose risks to human health. Consequently, indoor disinfection generally requires meticulous attention to ventilation, such as in hospitals and indoor swimming pools (Huang, 2012; Tang, 2003; the standard GB 15982-2012)".

Reference:

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).

[Comment]: 3. For the part of comparing with other works, the authors frequently attributed the difference to the use of different methods without other explanation. Please provide a clearer discussion. For instance, why different methods are used? which one is better? suggestions to reduce the uncertainties of the methodology ....

Response: Thanks for your valuable suggestion. We agreed and have made a major revision for discussing the comparison with other studies. Please see Section 4.1 in the revised manuscript.

[Comment]: 4. Please make sure all the numbers used in the calculations have proper references.

Response: Your suggestion is greatly appreciated. We have included proper references for all the numbers used for emission calculations throughout the revised manuscript.

Specific comments:

[Comment]: 5. For the name RCEI and statement such as in line 46, it is hard to say whether HCl and pCl can be grouped into reactive chlorine species as they are not that reactive and fast producing Cl radicals in the atmosphere.

Response: Thanks for your valuable suggestion. We agree with the perspective put forth by the reviewer. Consequently, we have substituted "reactive chlorine species" with "chlorine species" throughout the revised manuscript.

[Comment]: 6. Paragraph 2, you said research on anthropogenic chlorine emission in China is very limited and rarely considered in air quality simulations, but later you provide a series of examples in paragraph 3 and 4. I would suggest removing those statements and merging them with paragraph 4.

Response: Thanks for your valuable suggestion. To provide a logical presentation, we have removed these statements in paragraph 2, carried out the necessary modifications and restructured the introduction section. Please see Section 1 in the revised manuscript.

[Comment]: 7. Paragraph 3, this part starts with saying emission inventory in foreign countries, but no related information are introduced.

Response: Thanks for pointing out this issue. To avoid confusing, we have revised this sentence in Section 1 as follows: "The study of estimating the anthropogenic chlorine emission in China began by Mcculloch et al. (1999), who established a global

anthropogenic chlorine emission inventory called Reactive Chlorine Emissions Inventory (RCEI) based on a relatively rough statistical dataset in 1990".

[Comment]: 8. line 87, which sources were overlooked, such as?

Response: Thanks for your question. The overlooked sources in previous studies include environmental disinfection, tap water utilization, pesticide application, etc. We have added this information in Section 1 as follows: "Such overlooked emissions include those from environmental disinfection, tap water utilization, and pesticide application, and etc.".

[Comment]: 9. line 89, you may want to summarize the pros and cons of these studies at least briefly before this statement. Why did the previous estimates differ so largely? What are the uncertainties?

Response: We appreciate your valuable suggestions. We have provided a specific discussion on the previous studies in Section 1 as follows: "Despite these studies, the anthropogenic chlorine emission in China remains uncertain and further investigation is still warranted. Firstly, the estimated chlorine emissions in China varied in different studies due to the different applications of emission factors and estimation methods. Some studies have utilized emission factors derived from foreign sources or standards and guidelines that may not accurately reflect the specific local conditions in China. Some calculation methods are rudimentary and lack the granularity needed to effectively capture variations among provinces or different sources. Secondly, some modeling studies (Choi et al., 2020; Li et al., 2021) have used the anthropogenic chlorine emission as inputs and found that the simulated concentrations of chlorine species (HCl and pCl) were underestimated against the observation, which suggests that there are large uncertainties or missing sources for the current emission estimation. Lastly, chlorine emissions from anthropogenic activities were reported in the recent literature, but they have not been considered in the developed emission inventory for

China. Such overlooked emissions include those from environmental disinfection, tap water utilization, pesticide application, and etc. The neglect of these sources can lead to an underestimation of the total chlorine emission. As a result, the development of a comprehensive anthropogenic chlorine emission inventory that addresses the above issues is of great significance in reducing the uncertainty of the emission estimation".

[Comment]: 10. line 92, what is basic data? Do you mean activity data?

Response: Thanks for this valuable question. Here the basic data means the activity data. To clarify the description, we have replaced "basic data" with "activity data" in Section 1.

[Comment]: 11. line 116, please provide references for the release ratio.

Response: Thank you for your valuable suggestion. In the revised manuscript, we mentioned the value of release ratios for usages of chlorine-containing disinfectants and pesticides with references in Section 2.2.6 and 2.2.7, respectively. Here, we removed this introduction in Section 2.1.

[Comment]: 12. line 144, based on your statement, MM should equal to 0.5 for Cl_2 as you defined it as the ratios of the molar mass of chlorine atom to the molecular weight of chlorine species.

Response: Thanks for pointing out this issue. The term "MM" is the chlorine content in the chlorine species. As a result, it is 35.5/36:5, 1, 1 for HCl, fine particulate Cl-, and Cl_2, respectively. We have revised the text in Section 2.2.1 as follows: "MM refers to the chlorine content in chlorine species (35.5/36:5, 1, 1 for HCl, pCl, and Cl_2)".

[Comment]: 13. line 177-178, please provide references for the chlorine removal efficiencies.

Response: Thank you for your suggestion. We have provided the reference (Liu et al., 2018) for the chlorine removal efficiencies in the revised manuscript in Section 2.2.3.

Reference:

Liu, Y. M., Fan, Q., Chen, X. Y., Zhao, J., Ling, Z. H., Hong, Y. Y., Li, W. B., Chen, X. L., Wang, M. J., and Wei, X. L.: Modeling the impact of chlorine emissions from coal combustion and prescribed waste incineration on tropospheric ozone formation in China, Atmos Chem Phys, 18, 2709-2724, 10.5194/acp-18-2709-2018, 2018.

[Comment]: 14. Section 2.2.6, the whole part involves assumptions that the water in the facilities (water treatment plant, hospital, swimming pool, etc.) are open to the atmosphere and the waste gases are released without any treatment. This doesn't sound very true.

Response: Thanks for your valuable comment. Please see our response to the Comment 2.

[Comment]: 15. Section 2.2.6b, it is not clear how the calculation was conducted.

Response: Thank you for your suggestion. The emissions are estimated using Formula 12. We have specified the calculation method in Section 2.2.6b as follows: "The emission of water treatment is estimated using formula 12. The water amount that needs disinfection (W_j) is the quantity of tap water supplied in each province, which can be obtained from the China Urban and Rural Construction Statistical Yearbook 2019 (National Bureau of Statistics, 2019a). The emission factors can be found in Table S5."

[Comment]: 16. line 496, why are the emission factors and control technologies different? Are your estimations better? Same for the entire section 4.1, when you said different results were due to different calculation methods, could you please elaborate more and maybe demonstrate that your methods are more appropriate?

Response: Thank you for your valuable comment. Regarding the emission factors for coal combustion, this study relied on the local measured and survey data from the literature. Compared with Fu et al. (2018), who relied on control technology application ratios assumed from national policies, Yin et al. (2022), who used foreign application ratios, and Zhang et al. (2022), who employed an overestimation of application ratios using an S-curve formula, this study's selection of control technology application ratios is based on the domestic research literature, rendering it a more reasonable choice. For open waste incineration, this study estimates the open burning ratio for each province based on the proportion of open waste burning from the statistical yearbooks. Relative to Yin et al. (2022), this study offers a more refined differentiation for each province, aligning it more closely with real conditions. In the case of $PM_{2.5}$ emission factors for biomass combustion, Fu et al. (2018) and Yin et al. (2022) relied on data from the Guidelines for Compilation of Atmospheric Pollutant Emission Inventories for Biomass Combustion, while this study referenced literature from field observations, thereby making the estimations more reasonable. For restaurant emissions, this study adheres to lower flue gas flow rates, shorter cooking durations, and a lower Cl^- proportion in $PM_{2.5}$, which was obtained through local measurements and is in accordance with national standards and actual circumstances. Consequently, this approach reduces cooking emissions compared with Qiu et al. (2019) and Yin et al. (2022). In the case of swimming pools, while Yin et al. (2022) used data from the standard, this study drew upon data from the experimental research literature. Furthermore, this study has provided a more detailed breakdown of indoor and outdoor swimming pool operating hours and dosages. We have made specific modifications and explanations in Section 4.1.

[Comment]: 17. Figure 2 provides the same information as Figure 1d. Why put it as an individual figure? Also, it looks like the proportion of chlorine emissions from different sources of disinfection, not the proportion of actives as the figure title described.

Response: Thanks for your careful review. Figures 1d and 2 are two different figures. Figure 1d is to show the contribution of different source categories to the total HOCl emission, including emissions from usages of chlorine-containing disinfectants and pesticide. Figure 2 is to show the contribution of different usages of chlorine-containing disinfectant to the total emission of disinfection process, in which the emission from pesticide usages is excluded. To avoid confusion, we have revised the caption of Figure 2 as follows: "Figure 2 Proportion of chlorine emissions from different usages of chlorine-containing disinfectant in China".

[Comment]: 18. Figure 4: please introduce the green line in the label or figure captain.

Response: Thanks for this comment. The green line in Figure 4 represents the emissions for each province. We have included this information in the caption of Figure 4 as follows: "Figure 4 Emissions (green line) and contribution proportions of HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) by province in 2018."

[Comment]: 19. Figure 5: it is quite strange to use the unit of Mg/grid/yr, especially when no grid information is provided in figure captain. I would suggest using a unit of Mg/m2/yr or something similar.

Response: Thanks for this valuable comment. We have incorporated information about the grid resolution (0.1° × 0.1°) into the caption of Figure 5 as follows: "Figure 5 Spatial distribution of anthropogenic HCl, fine particulate Cl^-, Cl_2, and HOCl emissions in 2018 at a 0.1° × 0.1° resolution."

[Comment]: 20. Table 6: could you please also include emission numbers or ranges? I am not clear what useful information can be provided by comparing uncertainties with different studies.

Response: Thank you for your suggestion, which we agree. Instead of Table 6, we drew Figure 7 which included the emission and its range of uncertainty for different emission inventories. Generally, the percentage of uncertainty has slightly reduced in this study. We have revised the text in Section 4.2 as follow: "The uncertainties for HCl, pCl, Cl_2, and HOCl emissions were estimated at a 95% confidence interval, resulting in percentage ranges of -48% to 45%, -59% to 89%, -44% to 58%, and -44% to 79%, respectively (Figure 7). It can be seen that the estimated emissions of HCl and pCl are within the uncertainty ranges of other studies. Due to the additional sources of Cl_2 and HOCl in this study, the emissions are relatively higher compared with Yin et al. (2022). However, the percentage of uncertainty for all chlorine species generally reduces in this study compared with the other studies."

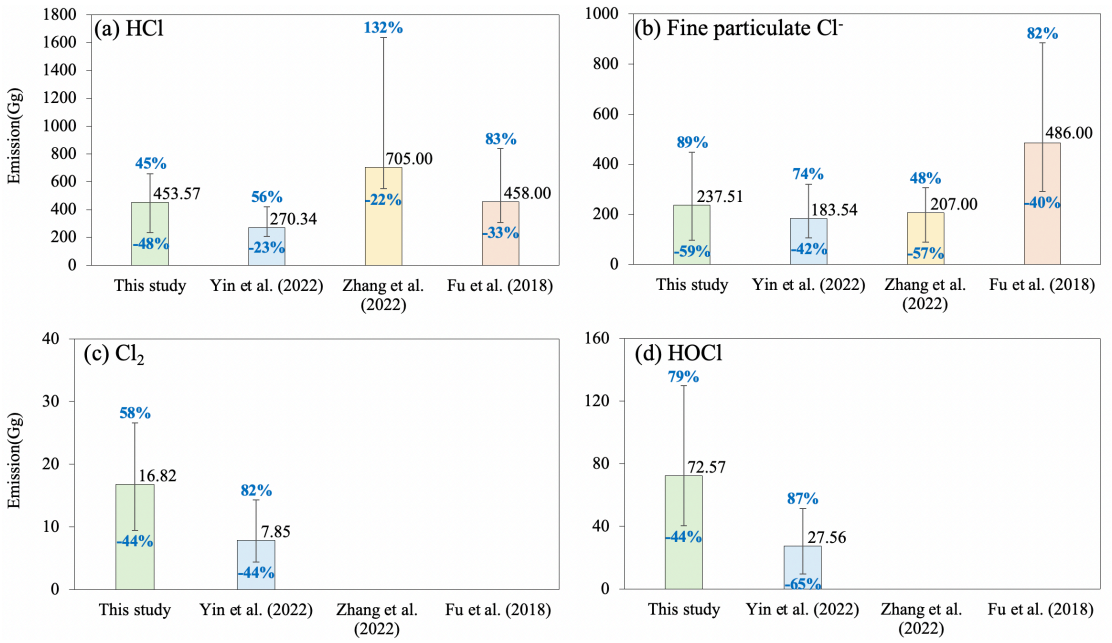

Figure7 Comparison of anthropogenic chlorine emissions and uncertainty ranges (blue text) with other studies.