Review comments on the manuscript titled “ACEIC: a comprehensive anthropogenic chlorine emission inventory for China”

The chlorine radical assumes a pivotal role in atmospheric chemistry, exerting a significant influence on atmospheric oxidation capacity, thereby contributing to secondary air pollution. Chlorine emanates from diverse sources, encompassing both anthropogenic and sea salt aerosol. While preceding studies have partially addressed certain aspects of chlorine emissions, a more comprehensive and reliable dataset consisting complete chlorine species and their respective source contributions remains a pressing necessity. The present study developed an emission inventory detailing anthropogenic chlorine sources, including HCl, Cl_2, pCl and HOCl, across 7 distinct source sectors in China for the year 2018 by using the emission factor methodology. Although the topic is both interesting and significant, the manuscript appears to be hindered by a lack of novelty. Several existing papers have already reported chlorine emissions, spanning cities, regions and even the entire nation of China, encompassing a multitude of sources. The current study closely aligns with these prior works, employing nearly identical methods, activity data and reported emissions factors. Consequently, the findings and uncertainties presented within this study are largely consistent with those from earlier investigations. Therefore, the identification of genuinely novel insights or contributions to the scientific community proves to be a challenge.

Response: We thank the referee for providing a thoughtful and detailed review of our paper. We have carefully considered the valuable suggestions and major revisions of this study are shown as follows: 1) The abstract and introduction sections have been reorganized to highlight the novelty of this study. Compared with previous studies, this updated inventory considered more anthropogenic sources, used more localized emission factors, and adopted more refined estimation method. 2) We improve the emission inventory by using the fire data from the Himawari-8 satellite to allocate the provincial open biomass burning emissions spatially and temporally. 3) We provide a deeper discussion on the comparison of this updated inventory to the previous studies. The advantage of this inventory is highlighted. The referee’s comments have helped to

improve this manuscript. Below, we provide a point-by-point response to the referee's comments and summarize the changes that have been made in the revised manuscript.

Major concerns:

[Comment]: 1.The primary objective of this study is to establish an extensive inventory encompassing anthropogenic chlorine emissions in China. This inventory includes HCl, fine particle Cl^-, Cl_2, and HOCl from 7 anthropogenic sources for year 2018. Nonetheless, the clarity in communicating the novelty and distinctiveness of this study from its precursors is lacking. Several papers have already reported on China's chlorine emissions inventory for various years, such as 2014 (Fu et al., 2018), 2019 (Yin et al., 2022), and even the range spanning 1960 to 2014 (Zhang et al., 2022). Particularly, the work by Yin (2022) has extensively detailed emissions of HCl, pCl, Cl_2, and HOCl, with source categories covering 22 sectors. Notably, they employed a spatial resolution of $0.1° \times 0.1°$ and reported the temporal resolution as well. The outcomes of the present study remain consistent with those presented in previous investigations (Fu et al., 2018; Zhang et al., 2022; Yin et al., 2022), and the level of uncertainties closely mirrors that of earlier researches, as indicated in Table 6. It is pertinent to highlight that this study adopts a methodology akin to prior works, employing the emission factor method, which does raise concerns about potential duplication of prior research efforts.

Response: Thank you for this question. However, this study is not a repetitive work. Compared with previous studies, we have made innovations and improvements in the following three aspects:

1. Expanded emission sources: This study has augmented the range of emission sources. For instance, it has included Cl_2 emissions from flat glass production, emissions from urban open waste incineration, emissions from municipal tap water usage and wastewater treatment in county towns, emissions from tap water usage during disinfectant application, emissions during environmental disinfection processes, and emissions of chlorine-containing substances from pesticide use.

2. Localized emission factors: Certain emission factors have been localized in this

study. For example, for $PM_{2.5}$ emissions from biomass combustion, while Yin et al. (2022) and Fu et al. (2018) relied on emission factors from the Guidelines for Compilation of Atmospheric Pollutant Emission Inventories for Biomass Combustion, this study primarily drew upon local measured data from the literature. This localization makes the emission factors more aligned with real-world conditions, thus reducing uncertainty. For the emission of cooking, the study adopted lower flue gas flow rates, shorter cooking durations, and a lower Cl^- proportion in $PM_{2.5}$, based on national standards and actual circumstances, which results in reduced cooking emissions compared with Qiu et al. (2019) and Yin et al. (2022). Additionally, for chlorine dosing in swimming pools, this study referenced experimental research literature, whereas Yin et al. (2022) relied on national standards.

3. Refined estimation method: The estimation methodology has been refined, rendering the results more detailed and realistic. For instance, in the case of waste incineration, different provinces are assigned different open burning rates, enhancing the accuracy of the inventory for open waste incineration. For swimming pool emissions, the study distinguishes the opening days between indoor and outdoor pools, in contrast to Yin et al. (2022), who assumed all pools only open in summer, making this study more realistic and detailed.

Hence, this study does not duplicate previous efforts but rather innovates and improves upon prior research. It expands emission sources, localizes certain emission factors, and refines calculation methods, reducing errors and uncertainties in the inventory, thereby enhancing the completeness of the anthropogenic chlorine emissions inventory for China. Please see the revision in Section 1 and Section 4.

[Comment]: 2. The authors have highlighted that there exists uncertainty regarding previous anthropogenic sources, necessitating further investigation. However, the specific nature of these uncertainties and the areas requiring further inquiry remain unclear. Has the current research succeeded in mitigating any of these uncertainties? If so, through what means has this reduction been achieved? Have the authors

incorporated more precise activity data, or have they embraced empirically measured emission factor data? It appears that the authors predominantly relied on activity data from statistical yearbooks and incorporated emissions factor information gleaned from previous literature. Consequently, the uncertainties associated with HCl, pCl, Cl_2, and HOCl emissions spanned a range of -48% to 45%, -59% to 89%, -44% to 58%, and -44% to 79% respectively. These figures closely mirror those from earlier studies, as also evidenced in Table 6.

Response: Thank you for this question. To ameliorate the uncertainty associated with this inventory, our study has undertaken the following concerted efforts.

Firstly, with respect to emission sources, our research surpasses the scope of Yin et al. (2022) by considering Cl_2 emissions from flat glass production, emissions from urban open burning of waste, emissions resulting from municipal tap water usage and wastewater treatment in counties, emissions during the use of disinfectants that include tap water, and emissions during environmental disinfection and pesticide use. These additions rectify underestimations and reduce a certain degree of uncertainty stemming from insufficient consideration of sources.

Secondly, we have localized improvements in certain emission factors, thereby mitigating uncertainties. While Yin et al. (2022) and Fu et al. (2018) relied on $PM_{2.5}$ emission factors for biomass burning derived from the Guidelines for Compilation of Air Pollutant Emission Inventories for Biomass Burning, our study predominantly relies on emission factors sourced from field measurements, rendering them more congruent with actual conditions and reducing uncertainty. For the cooking emission, we have adopted lower smoke flow rates and shorter cooking durations in accordance with national standards and practical circumstances. Additionally, we have employed Cl^- in $PM_{2.5}$ ratios lower than those derived from local measurements compared with Qiu et al. (2019), thereby reducing cooking emissions. Moreover, regarding the emission factor of the swimming pool, while Yin et al. (2022) based their calculations on national standards, our study draws from experimental research literature, aligning more closely with actual chlorine addition practices.

Lastly, we have refined the estimation methodology to yield more detailed and

reasonable results, thus diminishing uncertainty. In the context of waste incineration, we have differentiated between provinces with varying rates of open incineration, augmenting the completeness and precision of the inventory for this aspect. Similarly, with respect to swimming pool emissions, we have considered the varying opening days of indoor and outdoor pools, in contrast to Yin et al. (2022) who assumed that all pools only open during the summer season.

In summary, the innovations in our study primarily aim to reduce errors and uncertainties in the inventory, thereby enhancing the comprehensiveness and meticulousness of the anthropogenic chlorine emission inventory. We have revised the text in Section 4.2.

[Comment]: 3. The authors emphasized that "it should be noted that some important sources of chlorine emissions have been overlooked, leading to large uncertainties in recent decade estimates (219-707 Gg for HCl emissions in China)." These findings pertain to diverse years and encompass various source sectors. The methodology employed in this study is the "emission factor" method, aligning precisely with the approaches adopted in previous works (Fu et al., 2018; Hong et al., 2020; Yi et al., 2021; Yin et al., 2022); however, the references have not been properly cited in describing all the methods. Activity data were sourced from various references including statistical yearbooks, government statistics, and Gaode's POI data (Line 120); whereas emission data were curated and chosen from the existing literature (Line 121).

Response: Thank you for this question. Firstly, all the activity data and emission factors and their sources can be found in the supplementary information. We have also added citations for the critical activity data and emission factors in the revised manuscript. We have revised the text in Section 2.2 as follows: "This emission inventory uses a large amount of activity data and advanced emission factors. Most of the activity data, emission factors, and related references can be found in Tables S3-S12. Generally, the activity data were obtained from the yearbook (e.g., China Energy Statistical Yearbook, China Industry Statistical Yearbook, and China Urban-Rural Construction Statistical

Yearbook), government statistics (e.g., National Bureau of Statistics, and General Administration of Sport of China), and Gaode's POI data. The emission factors were mainly based on the measured and survey data from the literature".

Secondly, even though we employed the same "emission factor" methodology, there are variations in the selection of emission factors and specific estimation methods in different studies. Some of them have been improved in this study compared with previous studies. For example, the estimation of the combustion ratio for the open waste burning and the estimation of opening hours for the swimming pool have seen innovative improvements in this study. Please see our response to Comment 1 and 2.

[Comment]: 4. The manuscript refers to "41 specific source categories" concerning anthropogenic chlorine emissions. However, it remains unclear what precisely these 41 specific source categories encompass? Figure1 illustrate 7 primary categories and 24 distinct sources; Figure 3 demonstrate 5 economic sectors and 13 sources. In Table S1, there are 33 sub-categories noted. Upon examination of Table 2, only 24 source categories are evident. The classifications appear to be rather confusing. Additionally, why do the authors aggregate the emission source sectors from 7 to 5? If the authors divide the 7 main categories into 5 economic sectors. For restaurant sources, swimming pools, water treatment, and wastewater sources, to which sector do they belong to? These are missing in Figure 3.

Response: Thank you for pointing out this issue. In the revised supplementary information, Tables S1 and S2 show the aggregation of 41 specific sources into 7 primary source categories and 5 economic sectors, respectively. We have added this information in Section 2.1 as follows: "In our study, we have compiled the emissions of chlorine species (HCl, pCl, Cl_2, HOCl) from 41 anthropogenic activities (Table S1 and S2) across the 31 provinces in mainland China. These emissions are categorized into seven major source categories (Table S1)", Section 3.2 as follows: "We aggregated the anthropogenic chlorine emissions from 41 specific sources into 5 economic sectors, including power, industry, residential, agricultural, and biomass burning (Table S2)",

the caption of Figure 1 as follows: "The aggregation of 7 primary source categories from 41 specific sources can be found in Table S1", and the caption of Figure 3 as follows: "The aggregation of 5 economic sectors from 41 specific sources can be found in Table S2". Previous studies (e.g., Yin et al., 2022) on chlorine emissions have classified specific sources into 7 primary source categories based on the energy consumption. In this study, besides this classification method, we aggregated the specific sources into economic sectors. This categorization is also adopted for the widely-used emission inventory in China (e.g., MEIC) and it can attract the interest of potential readers of government officials and economic experts. According to Table S2, restaurant sources, swimming pools, water treatment, and wastewater sources belong to the residential sector.

Reference:

Yin, S., Yi, X., Li, L., Huang, L., Ooi, M. C. G., Wang, Y., Allen, D. T., and Streets, D. G.: An Updated Anthropogenic Emission Inventory of Reactive Chlorine Precursors in China, ACS Earth and Space Chemistry, 6, 1846-1857, 10.1021/acsearthspacechem.2c00096, 2022.

[Comment]: 5. The primary contribution to chlorine emissions in the outcomes of this study arises from biomass burning. However, instead of relying on FINN/GFED/GFAS data, this research utilized the "percentage of biomass domestic burning and open burning by province." The data from Table S6 are referenced from Zhou (2017). It's noteworthy that this information may not accurately reflect the conditions in the year 2018. Also, only using statistical data to estimate emissions of biomass burning and allocate the emissions spatially and temporally will raise large uncertainties.

Response: Thank you for this question. The activity data for biomass burning emission estimation can be derived from statistical data or satellite data. Both methods have their advantages and disadvantages. Compared with the method using satellite data, the method based on statistical data can estimate the household burning, and avoid missing the small or short-term fire incidents that cannot be detected by the satellite. It's worth

noting that in recent years, many studies have employed this calculation method (Zhang et al., 2019; Li et al., 2019; Zhou et al., 2019; Wang et al., 2018; Yan et al., 2006), which underscores the advantages of this approach. For the spatial and temporal allocation, in the revised manuscript, we used the fire location and fire radiation power over cropland from the Himawari-8 satellite data in 2018, instead of rural population density and empirical statistic data, to allocate the provincial open biomass burning emission to reduce the uncertainty. The spatial distribution maps for chlorine emission have been revised in the study.

We have added this description in Section 2.3 as follows: "For the emission of biomass open burning, we allocated the provincial emissions spatially to the fire location according to its fire radiation power over the cropland. The fire location and its fire radiation power data were derived from the Himawari-8 satellite data (https://www.eorc.jaxa.jp/ptree/userguide.html, last access: 1 January 2023)", and Section 2.4 as follows: "Based on the fire location and its fire radiation power over the cropland from the Himawari-8 satellite data, we performed temporal allocation of chlorine emissions from biomass burning for each province".

Reference:

Li, L., Zhao, Q., Zhang, J., Li, H., Liu, Q., Li, C., Chen, F., Qiao, Y., Han, J.: Bottom-up emission inventories of multiple air pollutants from open straw burning: A case study of Jiangsu province, Eastern China, Atmospheric Pollution Research, 10, 501–507, 2019.

Wang, J., Xi, F., Liu, Z., Bing, L., Alsaedi, A., Hayat, T., Ahmad, B., Guan, D.: The spatiotemporal features of greenhouse gases emissions from biomass burning in China from 2000 to 2012, Journal of Cleaner Production, 181, 801-808, 2018.

Yan, X., Ohara, T. Akimoto, H.: Bottom-up estimate of biomass burning in mainland China, Atmospheric Environment, 40, 5262-5273, 2006.

Zhang, X., Lu, Y., Wang, Q., Qian, X.: A high-resolution inventory of air pollutant emissions from crop residue burning in China, Atmospheric Environment, 213, 207–21, 2019.

Zhou, Z., Tan, Q., Deng, Y., Wu, K., Yang, X., Zhou, X.: Emission inventory of

anthropogenic air pollutant sources and characteristics of VOCs species in Sichuan Province, China, Journal of Atmospheric Chemistry, 76, 21–58, 2019.

[Comment]: 6. Regarding the industrial production process, could you clarify how many industries are encompassed within this category? As it stands, it appears that only four specific types of industries are accounted for, namely cement, iron, steel and flat glass. However, there are additional industries, such as chemical industries, which are known to release chlorine. How have these industries been addressed by the authors? Furthermore, from my perspective, "iron" and "steel" could arguably be regarded as a single industry. What's the difference here by separating them into two specific industries? Regrettably, the provided information lacks details.

Response: Thank you for this question. In Section 2.2.2, we have mentioned that five specific types of industries have been taken into account in this inventory: cement, iron, steel, flat glass, and HCl production. It's worth noting that HCl production belongs to the chemical industries. In the future, we are committed to conducting further research to incorporate additional sources of chlorine emissions from various chemical industrial processes into the inventory.

Regarding the separation of iron and steel, we have maintained this distinction due to the differences in their emission factors (please see Table S6), as outlined in the study of Yi et al. (2020). This separation allows us to provide a more accurate and detailed estimation of chlorine emissions from these two distinct processes.

Reference:

Yi, X., Yin, S., Tan, X., Huang, L., Wang, Y., Chen, Y., and Li, L.: Preliminary study on the inventory of sources of hydrogen chloride and particulate chlorine in the atmosphere in Shanghai, Acta Scientiae Circumstantiae (in Chinese), 40, 469-478, 10.13671/j.hjkxxb.2019.0376, 2020.

[Comment]: 7. Some of the key parameters employed in this study are quite old. For

instance, in Line 175: “the value 2.2 g kg^{-1} reported by Emmel et al. (1989)”. “ηd is the chlorine removal efficiency of dust removal facilities (25.1%), and ηs is the chlorine removal efficiency of sulfate-removal facilities (95.5%)”. In Lines 138-140: “we adopted the data from the study of Fu et al. (2018), which is the value of consumed coal considering the coal transportation… The values of X, R, ηd, and ηs can be found in Table 2 of our previous study (Liu et al., 2018). ρ is the chlorine proportion of HCl (86.33%), fine particulate Cl^- (10.09%), and Cl_2 (3.58%) in emitted flue gases based on the local measurement (Deng et al., 2014).” Line 185: “η is the removal efficiency of $PM_{2.5}$ (99%) in the garbage incineration station (Nan, 2016)” In Line 190-195, the authors adopted data from Fu et al (2018), while those data are for year 2014… It's worth noting that these datasets hold the potential for inducing overestimations and might not accurately reflect the circumstances of the year 2018.

Response: Thank you for this question. The emission factors used in this study were derived from the relevant literature, with a preference for more recent and locally measured data whenever possible. However, it's acknowledged that due to the extensive scope of the work, some of the data might not be the most up-to-date, which can introduce uncertainties into the inventory. We are committed to continuously improving the emissions inventory. If more suitable data become available in the future, we will certainly update the inventory accordingly.

[Comment]: 8. “Water treatment” and “Tap water use”, are there any double counting? Water treatment should include “tap water use”.

Response: Thank you for this question. Water treatment and the usage of tap water are two different processes for chlorine emission. After undergoing water treatment, residual chlorine remains in the water, which is released during the process of using tap water. Hence, water treatment does not include the use of tap water, and there is no double counting.

[Comment]: 9. Spatial allocation: Addressing emissions from other point sources where detailed information is unavailable, a uniform distribution across each individual point within each province has been employed. This approach might be perceived as somewhat arbitrary. Could you confirm whether emissions from industries are also apportioned in an averaged manner? If the primary sources stem from biomass burning, could you please elaborate how to conduct the allocation of emissions from biomass burning? It appears that the authors did not incorporate the geographical information of fire spots for open biomass burning. If population density was employed, it could potentially introduce significant uncertainties.

Response: Thank you for your question. In the revised manuscript, we used the fire location and fire radiation power over cropland from the Himawari-8 satellite data in 2018, instead of rural population density and empirical statistic data, to allocate the provincial open biomass burning emission spatially and temporally. Such an allocation method can help reduce the uncertainty. Please see our response to Comment 5. Emissions from household biomass burning are still allocated based on rural population density.

For emissions from other point sources with unavailable installed capacity data, such as industrial production, we assumed a uniform emission for them and spatially allocated the provincial emissions evenly to each point. We acknowledge that this method may induce uncertainties. If we can obtain the production data from factories in the future, we will use this data as weighting factors for allocation.

[Comment]: 10. The discussions concerning temporal variations appear to be limited in depth. For instance, the substantial increase in HCl emissions in October compared to January, where the former is three times higher, and pCl emissions in October being five times that of January, lack sufficient justification. It's imperative to provide a reasoned explanation for these discrepancies.

Response: Thank you for your question. The temporal variation of HCl and pCl emission is mainly contributed by the biomass burning emission. In the revised

manuscript, we used the fire location and fire radiation power over cropland from the Himawari-8 satellite data to allocate the temporal variation of chlorine emissions from biomass open burning in various provinces. This modification can greatly reduce the uncertainty of the estimated temporal variation of the emission.

We have revised the text in Section 3.5 as follows: "Figure 6 shows the temporal variation of anthropogenic emissions for different chlorine species. For HCl and pCl, the emission in mainland China presents a bimodal variation. A remarkable peak is in early spring (February to April), and a small peak is in early autumn (August to October). The high emission in these months is attributed to the biomass burning emission with active agricultural activities. In contrast, emissions from other sectors remain relatively stable throughout the year. It's worth noting that the monthly variations vary across different regions because of the varied period of biomass burning, as shown in Fig. S5 and S6. For example, in Northeast China (Liaoning, Jilin, and Heilongjiang), where extensive straw burning occurs before crop planting, emissions are elevated in spring only."

[Comment]: 11. Comparison with previous studies: the discussions provided lack specificity. For instance, the statement "The total HCl emission in this study is comparable with those estimated in the study of Fu et al. (2018) but with different contributions from source categories" requires more detailed clarification. "The HCl produced by coal combustion in this study is ~2 times higher than their estimation, which is mainly due to the different emission factors and control technology". Given that Fu's study was conducted for year 2014 and this current study pertains to 2018, it's important to address the significant increase of HCl emissions, especially in light of the advancements in control technology over the intervening years. The contribution of emission factors and control technology to this twofold discrepancy needs to be explicitly outlined. The explanation for the lower HCl emissions from waste incineration being attributed to the use of more detailed and lower open-air combustion rates in various provinces could be elucidated further. Similarly, stating that higher HCl

emissions from biomass burning are due to different estimation methods of household combustion rate and open combustion should be expanded upon for better clarity. When comparing your study's HCl emissions estimation with Zhang et al. (2022), discussing how your lower estimations were achieved due to factors such as coal combustion and waste incineration estimation is a good start. However, it would be beneficial to explicitly mention whether these adjustments have led to reduced uncertainties and whether your results can be considered more accurate compared to Zhang et al. (2022). For Cl_2 emissions, explaining why you adopted relatively higher release ratios of chlorine for coal combustion and the factors contributing to the higher emissions from the usage of disinfectant is important.

Additionally, there are some papers reporting chlorine emission for different emission sectors for various regions, eg, Li et al (2020) for Shanghai; Yi et al (2021)for YRD; Qiu et al (2019) for Beijing. The results can also be compared and inserted to the table.

Response: Thank you for this question. The differences in HCl emissions from coal combustion in this study compared to previous research mainly stem from variations in emission factors and control technologies. Our study relies on emission factors sourced from field observations, in contrast to Fu et al. (2018), who utilized control technology application ratios based on national policies, Yin et al. (2022), who adopted foreign application ratios, and Zhang et al. (2022), who employed an overestimated application ratio derived from an S-curve formula. The control technology application ratios used in our study are based on domestic research literature, providing a more reasonable basis. Regarding open waste incineration, we estimated the open burning ratio for each province as 1 minus the treatment rate (sourced from the statistical yearbook), in contrast to Yin et al. (2022), who assumed a uniform open burning ratio of 0.05 for all provinces nationwide. Our approach allows for a more nuanced consideration of each province's unique circumstances and is better aligned with reality. In our study, higher HCl emissions from biomass combustion are primarily attributed to differing estimations of household combustion rates and outdoor combustion methods. Our household combustion rate is substantiated by literature with documented data sources. Additionally, our approach to open burning employs a bottom-up emission factor

method, which avoids underestimation resulting from the omission of small-scale or short-term fire incidents compared to satellite-based detection methods. Concerning the $PM_{2.5}$ emission factor for biomass combustion, Fu et al. (2018) and Yin et al. (2022) used data from the Biomass Burning Emissions Inventory Guidelines. In contrast, our study references literature with field observations, resulting in a more realistic estimation. For the restaurant sector, we adopted lower flue gas flow rates, shorter cooking times, and lower Cl^- proportions in $PM_{2.5}$, compared to Qiu et al. (2019), which led to reduced cooking emissions.

Regarding Cl_2, our chlorine release rate is based on a specific study (Deng et al., 2014) and derives from on-site experiments, lending it greater credibility. Both Fu et al. (2018) and Yin et al. (2022) referenced this article, but their chlorine release rates were incorrect, resulting in duplicated calculations. For swimming pools, Yin et al. (2022) used standard data, while our study incorporated data from the experimental research literature, further delineating the indoor and outdoor pool opening times and dosages. This addition resulted in higher disinfectant emissions than Yin et al. (2022). Furthermore, our study also includes emissions from environmental disinfection, tap water usage, and pesticide use, broadening the sources and thus reducing the uncertainty of the inventory.

In our discussion within the main text in Section 4.1, we have addressed these comparisons with other literature.

[Comment]: 12. “Only this study and Yin et al. (2022) considered the emissions from cooking. The emission from cooking in this study is lower due to lower flue gas flux and shorter cooking durations”. Qiu also considers cooking (Qiu, et al., Atmos. Chem. Phys. 2019, 19, 6737−6747).

Response: Thanks for your comment. Yin et al. (2022), Qiu et al. (2019), and this study all considered emissions from cooking. In this study, due to reference to national standards and actual conditions, lower smoke flow and shorter cooking time were used, and the proportion of Cl^- in $PM_{2.5}$ obtained through local measurements was lower than

that of Qiu et al. (2019), thereby reducing cooking emissions. We have revised the text in Section 4.1 as follows: "Yin et al. (2022), Qiu et al. (2019b) and this study all considered emissions from cooking.In this study, we employed lower flue gas flow rates and shorter cooking durations following national standards and actual conditions, and lower proportions of Cl^- in $PM_{2.5}$ based on the local measured data from the literature, which reduced the cooking emission compared with the other two studies."

Specific comments:

[Comment]: 13. Lines 35-40: The term "chlorine atoms" is defined as "Cl," which could lead to confusion with the "Cl free radical" mentioned in lines 45-50. To avoid any ambiguity, please consider clarifying this terminology or employing an alternate term for either "Cl free radical" or "chlorine atoms."

Response: Thank you very much for your suggestion. The entire text has been uniformly revised from "chlorine atoms" to "chlorine radials".

[Comment]: 14. Lines 53-54: The authors mentioned that "However, research on anthropogenic chlorine emission inventories in China is currently limited, and the temporal and spatial distribution of these emissions remains unclear". These descriptions are inaccurate. There are papers reporting temporal and spatial distribution of chlorine emissions (Fu et al., 2018; Yin et al., 2022).

Response: Thanks for pointing out this issue. We have removed this inaccurate description in the revised manuscript.

[Comment]: 15. Lines 54-57: The authors mentioned that "Consequently, anthropogenic chlorine emissions are rarely considered in numerical simulations of air quality, making it challenging to study the chemical mechanism of chlorine and quantify the contribution of anthropogenic chlorine emissions to ozone and other

pollutants using models". These descriptions are inaccurate. There are some papers that have already conducted the modeling study, but they are not properly cited (eg. Choi et al., Environ. Sci. Technol. 2020, 54, 13409−13418; Li et al., Environ. Sci. Technol. 2021, 55, 13625−13637; Wang et al., Atmos. Chem. Phys., 21, 13973 - 13996, 2021; Li et al., Journal of Geophysical Research: Atmospheres, 125, e2019JD032058. https://doi.org/10.1029/2019JD032058; Li et al., Journal of Geophysical Research: Atmospheres, 126, e2020JD034175. https://doi.org/10.1029/2020JD034175; Wang et al., Cite This: Environ. Sci. Technol. 2020, 54, 9908−9916; ⋯).

Response: Thanks for pointing out this issue. We have removed this inaccurate statement in Section 1. We have added the citations of these paper in Section 1 as follow: "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."

Reference:

Choi, M., Qiu, X., Zhang, J., Wang, S., Li, X., Sun, Y., Chen, J., Ying, Q.: Study of Secondary Organic Aerosol Formation from Chlorine Radical-Initiated Oxidation of Volatile Organic Compounds in a Polluted Atmosphere Using a 3D Chemical Transport Model, Environ Sci Technol., 54, 13409–13418, 2020.

Li, J., Zhang, N., Wang, P., Choi, M., Ying, Q., Guo, S., Lu, K., Qiu, X., Wang, S., Hu, M., Zhang, Y., Hu, J.: Impacts of chlorine chemistry and anthropogenic emissions on secondary pollutants in the Yangtze river delta region, Environmental Pollution, 287, 117624, 2021.

[Comment]: 16. Lines 75-80, lack punctuation: "and waste incineration The study pointed out."

Response: Thanks for pointing out this typo. We have added "." after "waste incineration" for a punctuation here in Section 1.

[Comment]: 17. Line 77, missing “.” after “waste incineration”.

Response: Thanks for pointing out this typo. We have added ”.” after “waste incineration” here in Section 1.

[Comment]: 18. Line 80: It defines "particulate Cl^-" as "(pCl)," which has already been defined in line 45. To avoid redundancy, please refrain from repeating this definition.

Response: Thank you for your suggestion. To avoid redundancy, we have removed the definition of pCl here in Section 1.

[Comment]: 19. Line 85: “chlorinecontaining” should be “chlorine-containing”.

Response: Thanks for your suggestion. The word “chlorinecontaining” has been changed to "chlorine-containing” in Section 1.

[Comment]: 20. Line 115: The provided emission ratios of 0.84 and 0.11 for disinfectant use sources HOCl and Cl_2, respectively, require an explanation regarding the origin of this data.

Response: Thanks for pointing out this issue. 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]: 21. Line 120: To ensure clarity, it's essential to delineate the distinction between "activity data" and "emission data." Additionally, providing a precise definition for "emission data" would enhance understanding.

Response: Thank you very much for your suggestion. The term “emission data” has

been revised to "emission factor" for clarification in the first paragraph of Section 2.2. We have thoroughly reviewed the entire article and made the necessary corrections.

[Comment]: 22. Lines 175-180, the numerical values for ηd and ηs need references.

Response: Thank you very much for your suggestion. The reference (Liu et al., 2018) has been added in Section 2.2.3. Besides, we have carefully checked the manuscript and provided references for all the numbers used for emission calculations.

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]: 23. Line 195 mentions that the proportion of open burning of solid waste varies by location. Are there specific values listed in the appendix or a reference for this information?

Response: Thank you very much for your suggestion. F represents the proportion of open burning of solid waste, which means the untreated portion. It is calculated using 1-f. f represents the treated proportion of solid waste, which is derived from the China Urban and Rural Construction Statistical Yearbook 2019 (National Bureau of Statistics, 2019). We have added the text in Section 2.2.3 as follows: "F represents the proportion of open burning of solid waste, which means the untreated portion (1-f). f represents the treated proportion of solid waste, which is derived from the China Urban and Rural Construction Statistical Yearbook 2019 (National Bureau of Statistics, 2019a). The F value varied in different provinces due to the imbalance of economy, urbanization, and garbage disposal technology popularization."

Reference:

National Bureau of Statistics: China Urban-Rural Construction Statistical Yearbook (2019), China Statistics Press, Beijing, China, 2019.

[Comment]: 24. Lines 285-305: It appears that formulas (15) and (16) might be repetitive. Considering that the study encompasses both semi-standard and non-standard swimming pools, totaling 72%, could these categories be combined for the purpose of calculation? The study initially classifies swimming pools into public and private, and subsequently differentiates them as indoor and outdoor pools. Is it accurate to assume that the ratios of indoor and outdoor pools are evenly distributed between public and private ones? Additionally, does the value assigned to outdoor pool openings throughout the year seem excessively high?

Response: Thanks for your comment. Formulas 15 and 16 are two different calculations for the volumes of public and private swimming pools, which are not repetitive. We agree that the semi-standard and non-standard swimming pools can be combined for calculation purposes. To avoid confusion, we have revised the text in Section 2.2.6d as follows: "Swimming pools include public swimming pools and private swimming pools, and they have different volumes. The volume of public swimming pools was calculated as follows:

$$V_i = n_i \times \sum_j (a_j \times b_j \times h_j \times r_j) \quad (15)$$

where i and j represent different provinces and size types. Swimming pool size types include standard, semi-standard/non-standard swimming pools. We assume that the sizes of semi-standard and non-standard swimming pools are the same. n is the number of swimming pools, and the provincial data comes from the State Sports General Administration (https://www.sport.gov.cn/, last access: 1 January 2023). a, b, and h are the length, width, and depth of the swimming pool with different size types, as shown in Table S11. r represents the proportion of different size types of swimming pools, with standard swimming pools accounting for 28%, and semi-standard/non-standard swimming pools accounting for 72% (Zhang, 2015).

The volume of private swimming pools was calculated as follows:

$V_i = n_i \times a \times b \times h$ (16)

where i represent different provinces. n is the number of swimming pools, and the provincial data are estimated based on the ratio of residents' income to the number of swimming pools following the method proposed by Li et al. (2020). a, b, and h are the length, width, and depth of the private swimming pool, as shown in Table S11."

Due to the lack of relevant literature research or statistical data, this study assumes that the proportion of indoor and outdoor swimming pools is evenly distributed between public and private swimming pools. This assumption may induce uncertainties, and if the data is available in the future, we will improve it accordingly.

As for the opening hours of outdoor swimming pools, we made a typo during the writing process of the article. We have corrected the text in Section 2.2.6d as follows: "D is the number of opening days for the swimming pool. The indoor swimming pool in this study is open all year round, and the outdoor swimming pool is only open in summer."

[Comment]: 25. Line 317: Considering the total health expenditure and the corresponding ratio, it is estimated to be 11,898.0 L in 2018. How is it derived?

Response: Thank you for your question. As reported, in 2007, Taizhou hospitals used an average of 2329.2 L of chlorine-containing disinfectants (Sun et al., 2007). Since there was no data available for the usage of chlorine-containing disinfectants in 2018, we assumed that its change is proportional to the total health cost in recent years. The amount of disinfectant usage is estimated using the formula $U_{2018}=U_{2007} \times C_{2018}/C_{2007}$, where U represents the amount of disinfectant usage, and C represents the total health cost. The total health cost can be obtained from the China Health Statistical Yearbook (National Bureau of Statistics, 2008, 2019). As a result, the estimated usage of disinfectants in 2018 is 11898.0 L. It's worth noting that the inflation and price fluctuations may induce the uncertainty of this estimation method. We have added the text in Section 2.2.6e as follows: "Due to the absence of data in 2018, we assumed that its change is proportional to the total health cost in recent years. The amount of

disinfectant usage is estimated using the formula $U_{2018}=U_{2007} \times C_{2018}/C_{2007}$, where U represents the amount of disinfectant usage, and C represents the total health cost. The total health cost can be obtained from the China Health Statistical Yearbook (National Health Commission of the People's Republic of China, 2019, 2008). As a result, the usage of disinfectants in 2018 is estimated to be 11898.0 L".

Reference:

National Health Commission of the People's Republic of China: China Health Statistical Yearbook (2008), Peking Union Medical College Press, Beijing, China, 2008.

National Health Commission of the People's Republic of China: China Health Statistical Yearbook (2019), Peking Union Medical College Press, Beijing, China, 2019.

[Comment]: 26. Line 325, formula (19) does not provide the chlorine disinfectant concentration for the aquaculture industry. What are the proportions of HOCl and Cl_2 in this case?

Response: Thanks for pointing out this issue. The release ratios of HOCl and Cl_2 are provided in this formula. They are 0.84 and 0.11 according to the study of Wong et al. (2017). We have added the text in Section 2.2.6e as follows: "R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017)".

Reference:

Wong, J. P. S., Carslaw, N., Zhao, R., Zhou, S., and Abbatt, J. P. D.: Observations and impacts of bleach washing on indoor chlorine chemistry, Indoor Air, 27, 1082-1090, 2017.

[Comment]: 27. Line 340: It's possible that chlorine disinfectant use in household toilets is lower compared to public restrooms. Could the assumption of a 2 times higher chlorine disinfectant use in public toilets potentially be an overestimation? I

recommend estimating the quantity of chlorine disinfectant utilized per household (e.g., per bottle of bleach) based on population, and subsequently comparing it against the emission estimate rooted in the 2 times higher value. This analysis can help identify any potential instances of overestimation.

Response: Thanks for your suggestion. Due to the absence of data pertaining to disinfectant usage at the household level, coupled with the inherent variability in disinfectant consumption among households, the results of our study are subject to considerable uncertainty. In light of these limitations, we have opted to adopt the approach outlined by Li et al. (2020), as a temporary solution to address this challenge.

[Comment]: 28. Line 375, please provide the emission ratios for HOCl and Cl_2 during pesticide application.

Response: Thank you for your question. The emission ratios for HOCl and Cl_2 are 0.84 and 0.11, respectively, during pesticide application. These values were adopted according to the study of Wong et al. (2017) and Yi et al. (2021). We have added this information in Section 2.2.7 as follows: "R is the release ratio of HOCl (0.84) and Cl_2 (0.11) during the pesticide application (Wong et al., 2017; Yi et al., 2021)".

Reference:

Wong, J. P. S., Carslaw, N., Zhao, R., Zhou, S., and Abbatt, J. P. D.: Observations and impacts of bleach washing on indoor chlorine chemistry, Indoor Air, 27, 1082-1090, 2017.

Yi, X., Yin, S., Huang, L., Li, H., Wang, Y., Wang, Q., Chan, A., Traoré, D., Ooi, M. C. G., Chen, Y., Allen, D. T., and Li, L.: Anthropogenic emissions of atomic chlorine precursors in the Yangtze River Delta region, China, Sci. Total Environ., 771, 144644, https://doi.org/10.1016/j.scitotenv.2020.144644, 2021.

[Comment]: 29. Lines 442-448, the authors presented per-unit-area and per-capita emissions. However, it remains unclear what reasons contribute to these results? Within

the discussion section, the paper predominantly showcases data results, yet falls short in delving into a comprehensive exploration of the underlying reasons.

Response: Thank you for your suggestion. We have provided the reasons contributing to the provincial variations of per-unit-area and per-capita emissions in the revised manuscript. We have added the discussion in Section 3.3 as follows: "For the per-unit-area emission intensity, Shandong is the province with the highest emission intensity of HCl (238.13 kg km^{-2}) and fine particulate Cl^- (132.05 kg km^{-2}), which is attributed to its relatively higher emission but smaller area. Shanghai has the highest emission intensity of Cl_2 (60.07 kg km^{-2}) and HOCl (419.48 kg km^{-2}), which is due to its small area. For the per-capita emission intensity, Heilongjiang has the highest emission intensity of HCl (1014.21g per people) and fine particulate Cl^- (720.31 g per people) due to its highest emission across the country. Ningxia is the province with the highest emission intensity of Cl_2 (39.01 g per people) due to its low population. Shanghai is the province with the highest emission intensity HOCl (109.72 g per people) due to its relatively higher emission but lower population".

[Comment]: 30. Line 473-475, it is stated that "The emissions of HCl and fine particulate Cl exhibit relatively higher levels during early summer and autumn, coinciding with the frequent occurrence of biomass burning". However, it's important to note that the time period of biomass burning varies across different regions.

Response: Thanks for your comment, which we agree. In the revised manuscript, we used the fire location and its fire radiation power data from the Himawari-8 satellite data to allocate the temporal distribution of HCl and pCl emissions from biomass open burning in various provinces. We have added this description in Section 2.4 as follows: "Based on the fire location and its fire radiation power over the cropland from the Himawari-8 satellite data, we performed temporal allocation of chlorine emissions from biomass burning for each province". We have created new temporal distribution maps (Figure 6) and provided the temporal distribution maps for the seven major regions in mainland China in the supplementary information (Figures S5 and S6). The results

show that the variation of open biomass burning emissions presents regional differences. We have added the discussion in Section 3.5 as follows: "Figure 6 shows the temporal variation of anthropogenic emissions for different chlorine species. For HCl and pCl, the emission in mainland China presents a bimodal variation. A remarkable peak is in early spring (February to April), and a small peak is in early autumn (August to October). The high emission in these months is attributed to the biomass burning emission with active agricultural activities. In contrast, emissions from other sectors remain relatively stable throughout the year. It's worth noting that the monthly variations vary across different regions because of the varied period of biomass burning, as shown in Fig. S5 and S6. For example, in Northeast China (Liaoning, Jilin, and Heilongjiang), where extensive straw burning occurs before crop planting, emissions are elevated in spring only".

[Comment]: 31. Line 544-545, "The inventory can be enhanced by including emissions from other anthropogenic activities that release chlorine. For example, the disposal and combustion of medical waste, which often contains high levels of plastic, can result in the release of significant amounts of active chlorine" isn't medical waste included in this study?

Response: Thank you for your question. This inventory currently includes the emissions from domestic waste treatment, in which medical waste is excluded. We have added the text in Section 2.2.3 for clarification as follows: "Currently, only the emissions from domestic waste are considered in this study". We acknowledge the value of your suggestion and plan to incorporate it into the inventory in the future for comprehensiveness.

[Comment]: 32. Line 554-555: "In this study, we developed a Chinese anthropogenic chlorine emissions inventory (ACEIC 2018) using emission factors mainly based on local measurements", this is inaccurate, as there are no measurements presented in this

paper.

Response: Thanks for pointing out this issue. The emission factors for emission calculation were mainly based on the local measured and survey data from the literature. We have clarified the text in Section 5 as follows: "In this study, we developed a Chinese anthropogenic chlorine emissions inventory (ACEIC 2018) using emission factors mainly based on local measured and survey data from the literature."

[Comment]: 33. In section 3.3, it is suggested to provide reasons for higher chlorine emissions in different provinces to enhance the results analysis.

Response: Thank you for your suggestion, which we accept. We have revised the text in Section 3.3 as follows: "Regarding HCl emissions, Heilongjiang (38.27 Gg), Shandong (38.10 Gg), Henan (36.05 Gg), Hebei (32.46 Gg), and Hunan (24.45 Gg) emerge as the top five contributing provinces. They account for 8.4%, 8.4%, 7.9%, 7.2%, and 5.4% of the total emissions, respectively. The elevated emissions in Heilongjiang, Shandong, and Henan are attributed to the major contributions of biomass burning with higher agricultural production, which accounts for 77%, 54%, and 58%, respectively. For Hebei and Hunan, the higher emissions from industrial production are the major contributors, accounting for 40% and 31%, respectively. The top five contributors to fine particulate Cl^- emissions are Heilongjiang (27.18 Gg), Henan (21.60 Gg), Shandong (21.13 Gg), Hebei (15.46 Gg), and Anhui (14.67 Gg). In these provinces, higher biomass burning emissions induced by active agricultural activities dominate the total emission. Cl_2 emissions are predominantly attributed to Guangdong (1.40 Gg), Shandong (1.22 Gg), Hebei (1.09 Gg), Jiangsu (1.00 Gg), and Hunan (0.91 Gg). The top five provinces contributing to HOCl emissions are Guangdong (8.61 Gg), Jiangsu (5.50 Gg), Shandong (4.86 Gg), Zhejiang (4.04 Gg), and Sichuan (3.39 Gg). Due to the large population and developed economy that stimulates the need for disinfection processes, provinces such as Guangdong, Shandong, and Jiangsu have relatively high emissions of Cl_2 and HOCl".

[Comment]: 34. Line 740 mentions missing information on the meaning of the green line in Figure 4.

Response: Thanks for pointing out this issue. The green line in Figure 4 represents the emissions for each province. We have added 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]: 35. Line 760 suggests changing "Power" in the "Subsector" in Table 3 to "coal combustion."

Response: Thank you for your suggestion. We have replaced "Power" under the "Subsector" category with "Coal combustion" in Table 3.

[Comment]: 36. Upon observing Figure S1, it raises the question of why the Per-unit-area emissions of Cl_2 and HOCl are notably elevated in Shanghai.

Response: Thank you for your question. The notably elevated per-unit-area emission intensity of Cl_2 and HOCl in Shanghai is due to its small area. We have added this discussion in Section 3.3 as follows: "Shanghai has the highest emission intensity of Cl_2 (60.07 kg km^{-2}) and HOCl (419.48 kg km^{-2}), which is due to its small area".