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.

Major concerns:

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.

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.

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).

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.

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." This 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.

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.

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 Cl2 (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.

8. "Water treatment" and "Tap water use", are there any double counting? Water treatment should include "tap water use".

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.

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.

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.

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).

Specific comments:

1. 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."

2. 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).

3. 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; …).

4. Lines 75-80, lack punctuation: "and waste incineration The study pointed out."

5. Line 77, missing "." after "waste incineration".

6. 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.

7. Line 85: "chlorinecontaining" should be "chlorine-containing".

8. Line 115: The provided emission ratios of 0.84 and 0.11 for disinfectant use sources HOCl and Cl2, respectively, require an explanation regarding the origin of this data.

9. 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.

10. Lines 175-180, the numerical values for η_d and η_s need references.

11. 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?

12. 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?

13. 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?

14. 10. 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?

15. 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.

16. Line 375, please provide the emission ratios for HOCl and Cl_2 during pesticide application.

17. 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.

18. 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.

19. 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?

20. 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.

21. In section 3.3, it is suggested to provide reasons for higher chlorine emissions in different provinces to enhance the results analysis.

22. Line 740 mentions missing information on the meaning of the green line in Figure 4.

23. Line 760 suggests changing "Power" in the "Subsector" in Table 3 to "coal combustion."

24. Upon observing Figure S1, it raises the question of why the Per-unit-area emissions of Cl2 and HOCl are notably elevated in Shanghai.