ACEIC: a comprehensive anthropogenic chlorine emission inventory for China

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Abstract. Chlorine species play a crucial role as precursors to Cl radicals, which can significantly impact the atmospheric oxidation capacity and influence the levels of trace gases related to climate and air quality. Several studies have established the chlorine emission inventory in China in recent years, but the emission remains uncertain and requires further investigation. Anthropogenic Chlorine Emission Inventory for China (ACEIC) was the first chlorine emissions for China based on local data developed in our previous study, which only includes the emissions from coal combustion and waste incineration. In the present study, we updated this inventory to include data from a more recent year (2018) and expanded the range of species considered (HCl, fine particulate Cl^-, Cl_2, and hypochlorous acid (HOCl)) as well as the number of anthropogenic sources (41 specific sources). Compared with previous studies, this updated inventory considered more anthropogenic sources, used more localized emission factors, and adopted more refined estimation methods. The total emissions of HCl, fine particulate Cl^-, Cl_2, and HOCl in mainland China for the year 2018 were estimated to be 454 (-48%~45%), 238 (-59%~89%), 17 (-44%~58%), and 73 (-44%~79%) Gg, respectively. To facilitate analysis, we aggregated the chlorine emissions from various sources into five economic sectors: power, industry, residential, agriculture, and biomass burning. HCl emissions were primarily derived from biomass burning (45%), industry (35%), and residential (15%) sectors. The biomass burning and industry sectors accounted for 78% and 14% of the fine particulate Cl^- emissions, respectively. Residential and industry sectors contributed 59% and 31% of the total Cl_2 emissions. HOCl emissions were predominantly from the residential sector, constituting 90% of the total emissions. Notably, the usage of chlorine-containing disinfectants was identified as the most significant source of Cl_2 and HOCl emissions in the residential sector. Geographically, regions with high HCl and fine particulate Cl^- emissions were found in northeast China, the North China Plain, and the Sichuan Basin, whereas the Pearl River Delta, Yangtze River Delta, and Beijing-Tianjin-Hebei regions exhibited elevated levels of Cl_2 and HOCl emissions. Regarding monthly variation, emissions of HCl and fine particulate Cl^- were relatively higher during early spring (February to April) and early autumn

(August to October) due to intensified agricultural activities, while Cl_2 and HOCl emissions were higher in the summer months due to increased demand for water disinfection. This updated inventory contributes to a better understanding of anthropogenic sources of chlorine species and can aid in the formulation of emission control strategies to mitigate secondary pollution in China.

1 Introduction

Recent field and laboratory studies have revealed the crucial role of chlorine radical (Cl) in tropospheric chemistry (Faxon and Allen, 2013; Qiu et al., 2019a; Young et al., 2014; Peng et al., 2022). As highly reactive radicals, Cl can significantly impact the abundance of trace gases related to climate and air quality. Specifically, in the lower troposphere, Cl can initiate the oxidation of volatile organic compounds (VOCs), elevate the levels of conventional radicals (OH, HO_2, and RO_2), and produce ozone (O_3) and secondary aerosols that contribute to air pollution and alter the earth's radiation budget and climate (Qiu et al., 2019a; Wang et al., 2019; Li et al., 2021; Wang et al., 2020b). Furthermore, Cl reacts rapidly with methane, which is the most abundant hydrocarbon and the second-most important greenhouse gas emitted into the atmosphere (Li et al., 2022; Strode et al., 2020). Consequently, the study of chlorine chemistry in the troposphere has garnered increasing attention within the atmospheric chemistry community.

Chlorine species from anthropogenic activities, including HCl, fine particulate Cl^- (pCl), Cl_2, and HOCl, are important precursors of Cl radicals. HCl can react with OH radicals to release Cl radicals (Riedel et al., 2012). Particulate Cl^- provides aerosol surfaces for heterogeneous reactions with N_2O_5, producing $ClNO_2$ that is rapidly photolyzed into Cl radicals after sunrise (Thornton et al., 2010; Bertram and Thornton, 2009; Roberts et al., 2009; Osthoff et al., 2008). During the daytime, Cl_2 can be swiftly photolyzed, releasing two Cl radicals, while HOCl can also be photolyzed to release Cl radicals (Finlayson-Pitts, 1993; Faxon and Allen, 2013). These chlorine species are emitted from various anthropogenic sources (Chang and Allen, 2006; Yin et al., 2022), posing challenges in estimating their emissions. The development of chlorine emission inventories would enhance our understanding of the emission characteristics and primary sources of these emissions. Furthermore, it would provide crucial input data for numerical simulations of air quality, ultimately improving the accuracy of atmospheric pollutant predictions.

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. However, due to the rapid industrial and economic growth, this inventory cannot accurately represent the current situation of atmospheric chlorine emissions in China. The first anthropogenic chlorine emission inventory using local data in China (Anthropogenic Chlorine Emission Inventory for China (ACEIC)) was developed by our team (Liu et al., 2018), which considered the HCl and Cl_2 emissions from coal combustion and waste incineration for the year 2012. We then updated this inventory to the year 2014 (Hong et al., 2020). A more detailed chlorine emission inventory for the same year was established by Fu et al. (2018), including the HCl and pCl emissions from coal combustion, industrial production

processes, biomass combustion, and waste incineration. Zhang et al. (2022) also developed a comprehensive global emission inventory of HCl and pCl from 1960 to 2014, including China. Qiu et al. (2019b) compiled an updated emission inventory for Beijing in 2014 with pCl emissions from cooking considered. Lately, a team from Shanghai University has successively established anthropogenic chlorine emission inventories for Shanghai (Yi et al., 2020; Li et al., 2020), Yangtze River Delta region (Yi et al., 2021), and China (Yin et al., 2022), with the consideration of more chlorine species (HCl, pCl, Cl_2, and HOCl) from various sources.

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.

In this study, we update and improve the ACEIC inventory we developed previously to a more comprehensive one, which includes 4 chlorine species from 41 specific anthropogenic sources, with 2018 as the base year. Compared with the previous studies, this updated inventory considered more anthropogenic sources, used more localized emission factors, and adopted more refined estimation methods, which aims to reduce the uncertainty of chlorine emission estimations. The structure of this paper is organized as follows. Section 2 demonstrates the data and method utilized to develop the updated chlorine emission inventory ACEIC. Section 3 presents the characteristic of anthropogenic chlorine emissions, and Section 4 discusses the uncertainty and limitation of this inventory. The conclusion is summarized in Section 5.

2 Data and Method

2.1 Emission estimation

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): (1) coal combustion, (2) industrial production processes, (3) waste incineration, (4) biomass burning, (5) cooking, (6) usage of chlorine-containing disinfectants, and (7) usage of pesticides. To estimate anthropogenic chlorine emissions in China for the year 2018, we have employed the "emission factor" method, which is represented by the following equation.

$$E_{HCl} = \sum_{i,j} A_{i,j} \times EF_{(HCl)i,j} \quad (1)$$

$$E_{PCl} = \sum_{i,j} A_{i,j} \times EF_{(PM_{2.5})i,j} \times M_{i,j} \quad (2)$$

$$E_{Cl_2/HOCl} = \sum_{i,j,k} A_{i,j} \times \left(CD_{i,j} - CR_{i,j}\right) \times f_j \times R_k \quad (3)$$

Generally, the estimation of HCl and pCl emissions was carried out using formulas 1 and 2, respectively, while the calculation of Cl_2 and HOCl emissions utilized formula 3. In these formulas, the variables i, j, and k represent provinces, emission sources, and chlorine precursors, respectively. The symbol E represents emissions, A represents activity data, and EF represents emission factors. The variable M in equation 2 represents the proportion of Cl^- in $PM_{2.5}$. For equation 3, the variable CD represents the amount of chlorine added, CR represents the residual chlorine, f represents the volatilization rate of chlorine, and R represents the release ratios of chlorine gases.

2.2 Activity data and emission factor

The activity data and emission factor used to calculate the chlorine emissions of each source category are demonstrated in this section. 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. The selection process of emission factors was guided by the principles of prioritizing domestic local areas, prioritizing the most

recent year, and giving precedence to field observations. By adhering to these principles, the study seeks to minimize the inherent uncertainty associated with emission factors to the greatest extent possible.

125

2.2.1 Coal combustion

Chlorine in the coal can be released into the atmosphere through coal combustion. We estimated the emissions from coal combustion based on the coal consumption data, which were derived from the provincial energy balance spreadsheets of the "China Energy Statistical Yearbook" (National Bureau of Statistics, 2019e). We classified coal consumption into 4 economic

130 sectors: (1) power plants. (2) industry, including industrial processes, construction processes, and heating plants. (3) residential, including residential and commercial activities, transportation, and others. (4) agriculture. The emissions from coal combustion were calculated using the formula proposed in our previous study (Liu et al., 2018):

$$E_{i,j,k} = A_{i,j} \times c_i \times \sum_k \left[X_{j,k} \times R_{j,k} \times \left(1 - \eta_{d_{j,k}}\right) \times \left(1 - \eta_{s_{j,k}}\right)\right] \times \rho \times \frac{1}{MM_k} \quad (4)$$

135

where i, j, k, l are the province, source sector, chlorine species, and the energy allocation type (type of boiler and control device combination). A is the coal consumption by province in each source category, and c is the chlorine content in coal by province. Instead of using chlorine content in raw coal produced in each province (Liu et al., 2018), we adopted the data from the study of Fu et al. (2018), which is the value of consumed coal considering the coal transportation. X is the fraction of energy for a

140 sector (energy allocation ratio), and R is the chlorine release rate. η_d is the removal efficiency of dust-removal facilities, and η_s is the removal efficiency of sulfate-removal facilities. The values of X, R, η_d, and η_s 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). MM refers to the chlorine content in chlorine species (35.5/36:5, 1, 1 for HCl, pCl, and Cl_2).

145

2.2.2 Industrial production process

Chlorine species can be released during some industrial production processes. For example, the production of cement, iron, and steel will emit HCl and fine particulate Cl^-, and HCl will be volatilized during the production of hydrochloric acid, while the production of flat glass will produce HCl, fine particulate Cl^-, and Cl_2. The emissions from these industrial production

150 processes were estimated based on their production. The data of cement, iron, steel, and flat glass by province were collected from China Industry Statistical Yearbook (National Bureau of Statistics, 2019c), and those of industrial HCl were obtained from the National Bureau of Statistics (https://m.sohu.com/a/335035620_775892/?pvid=000115_3w_a, last access: 1 January 2023).

For HCl and Cl_2, the emissions were calculated using formula 1 and the emission factors of industrial productions of cement, iron, steel, hydrochloric acid, and flat glass were obtained from the literature (see Table S4 and S5). For fine particulate Cl^-, the emission factors (EF_{pCl}) were calculated from the emission factor of $PM_{2.5}$ ($EF_{PM2.5}$) and the Cl^- proportion in $PM_{2.5}$ (M):

$$EF_{pCl,i} = EF_{PM_{2.5},i} \times M_i \quad (5)$$

where i denotes different industrial production processes. These parameters for the industrial production of cement, iron, steel, and flat glass are shown in Table S6.

2.2.3 Waste incineration

The garbage disposal method in China primarily involves garbage incineration, which releases HCl and pCl during the incineration process. While China prohibits the open burning of garbage, there are still instances of open garbage incineration in certain areas. Therefore, garbage incineration can be categorized into two methods: garbage incineration stations and open garbage incineration. Currently, only the emissions from domestic waste are considered in this study.

For the emission from garbage disposal incinerators, the waste incinerations in each province/city were collected from the China Urban-Rural Construction Statistical Yearbook (National Bureau of Statistics, 2019a). The emissions of HCl were calculated using the method proposed in our previous study (Liu et al., 2018):

$$E_i = A_i \times EF \times (1 - \eta_d) \times (1 - \eta_s) \quad (6)$$

where i is the province, and A is the amount of waste incineration. EF is the emission factor of HCl during the incineration process. Here we used the value 2.2 g kg^{-1} reported by Emmel et al. (1989). η_d is the chlorine removal efficiency of dust-removal facilities (25.1%), and η_s is the chlorine removal efficiency of sulfate-removal facilities (95.5%) (Liu et al., 2018).

The estimation of fine particulate Cl^- emissions from garbage disposal incinerators was conducted using the following formula:

$$E_i = A_i \times EF_{PM2.5} \times M \times (1 - \eta) \quad (7)$$

In the formula, the variable i represents the province, $EF_{PM2.5}$ denotes the emission factor of $PM_{2.5}$, and M signifies the proportion of Cl^- in $PM_{2.5}$. The specific values for these parameters can be found in Tables S4 and S6. η is the removal efficiency of $PM_{2.5}$ (99%) in the garbage incineration station (Nan, 2016).

For the open waste incineration, there is no relevant statistical data at present, so the amount of open incineration was estimated as follows.

$$A_i = \left(P_{i,u} \times MSW_u \times F_{i,u} + P_{i,r} \times MSW_r \times F_{i,r}\right) \times B \times 365 \quad (8)$$

where i, u, and r represent the province, urban, and rural data, respectively. P is the population. MSW is the per capita waste production rate, which is 1.2kg/person/d in urban areas and 0.79kg/person/d in rural areas (Wang et al., 2017; He et al., 2010). 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. B is the garbage combustible rate, which is assumed to be 0.6 based on the study conducted by Fu et al. (2018). The emission factors of HCl and fine particulate Cl^- are shown in Table S4 and S6.

2.2.4 Biomass burning

Biomass burning includes household burning and open burning. Both of them include straw burning and firewood burning. The emissions from straw burning were calculated as follows:

$$A_{i,j,k} = P_{i,j} \times R_j \times D_j \times C_j \times F_{i,j,k} \quad (9)$$

where i, j, and k represent respectively the province, crop type, and combustion type. P represents the crop yield, R signifies the ratio of straw to crop products, and D represents the dry matter ratio. C denotes the combustion efficiency, and F represents the rates of household and open combustion. Specific values for these parameters can be found in Tables S7 and S8.

For the firewood burning, the emissions were estimated using the Eq. 10:

$$A_i = \frac{P_i}{S_i} \times B \times T \quad (10)$$

where i represents the province. P represents the rural population, while S represents the average size of rural households. The division P/S represents the number of rural households. B represents the consumption rate of firewood, and T represents the number of days on which firewood is burned. According to the research of Yi et al. (2021), it is assumed that each rural household burns 2 kg of firewood per day, and the total number of burning days in a year is 260. The emission factors of HCl and fine particulate Cl^- from straw and firewood combustion are shown in Tables S4 and S9, respectively.

2.2.5 Cooking

Chlorine emissions from cooking are mainly due to the usage of edible salt, which is released in the form of aerosols during the cooking process (Zhang et al., 2017). The emissions from cooking activities were categorized into three types: household catering, social catering, and canteen catering. In social catering, small and medium-sized catering enterprises contribute to more than 80% of the total emissions (Wu et al., 2018). Canteen catering includes school canteens and unit canteens. The formula for estimating pCl emissions from cooking was as follows:

$$E_{i,j} = N_{i,j} \times n_j \times Q_j \times H_j \times D_j \times (1 - \eta) \times EF_{PM2.5,j} \times M \quad (11)$$

where i and j represent provinces and catering types, respectively. N represents the number of households (i.e. population/family size), the number of social restaurants, the number of students and faculty members in middle schools and colleges, and the number of public institutions and government agencies. n is The number of furnaces, and Q is the smoke emission. H represents the cooking time per day, D is the cooking days, and η is the removal efficiency of the flue gas scrubber. $EF_{PM2.5}$ is the emission factor of $PM_{2.5}$, the above parameters are based on the study of Wu et al. (2018). M is the proportion of Cl^- in $PM_{2.5}$. According to the local measurement of Li et al. (2018), the average proportion of Cl^- in $PM_{2.5}$ is 1.545%. The parameters related to the emission factors of cooking are shown in Table S10.

2.2.6 Usage of chlorine-containing disinfectant

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). The rapid air exchange between indoor and outdoor during the ventilation process can help the emissions of chlorine gases into the atmosphere.

The emissions from the disinfection process were estimated as follows:

$$E_{i,j} = W_i \times (CD - CR) \times f \times R_j \quad (12)$$

where i and j are the province and chlorine species, respectively. W is the amount of water for disinfection treatment. CD and CR are the chlorine dose added to the water and the residual chlorine after disinfection, respectively. f is the volatilization ratio of chlorine, and R is the release ratio of chlorine gases. In this study, the release ratios of HOCl and Cl_2 during the disinfection process are 0.84 and 0.11, respectively (Wong et al., 2017). Except for the activity data (W), other parameters in this equation for different usages of disinfectant are presented in Table S5.

a Cooling tower

To prevent the breeding of bacteria and algae in the circulating water of the cooling tower and reduce the cooling efficiency, it is necessary to add disinfectant regularly and maintain a certain concentration. The volatilization of cooling water will cause the release of chlorine gases into the atmosphere. The chlorine emission was estimated based on the amount of supplementary water that was regularly added to the cooling tower. It was estimated according to the following equation:

$$W_i = A_i \times L \times S \quad (13)$$

where i is the province, and A is the industrial water consumption. These provincial activity data were obtained from the China Environmental Statistics Yearbook 2019 (National Bureau of Statistics, 2019d). L is the proportion of cooling water, which accounts for about 60% of industrial water (Hou and Zhang, 2015). S is the proportion of supplementary water. Here we used the value of 2.4% (Wang et al., 2020a; Zhao, 2015).

b Water treatment

During the water treatment process, which typically involves coagulation, sedimentation, filtration, and disinfection, the use of chlorine-containing disinfectants is a common practice for effective disinfection (Ge et al., 2006). As a result of this disinfection process, emissions of HOCl and Cl_2 are released into the atmosphere. The emission of water treatment is estimated using formula 12. The water amount that needs disinfection (W_i) 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.

c Waste water treatment

Waste water treatment encompasses the treatment of both medical waste water and domestic sewage. Before discharge, medical waste water undergoes disinfection to prevent the dissemination of pathogens. Similarly, domestic sewage is disinfected before being released into natural water bodies to safeguard the ecological balance against the proliferation of algae and microorganisms. The treatment capacity for medical waste water can be estimated based on the number of hospital beds and the rate of waste water production.

$$W_i = N_i \times Q \times D \quad (14)$$

where i is the province, and N is the number of hospital beds, which was derived from the China Health Statistics Yearbook 2019 (National Health Commission of the People's Republic of China, 2019). Q is the waste water production rate, each bed produces 0.62 m^3 of medical waste water per day (Zhou, 1987). D is the number of days of disinfection, which is 365.

The emissions from domestic waste water were estimated based on the provincial sewage treatment capacity. These data were obtained from the China Urban and Rural Construction Statistical Yearbook 2019 (National Bureau of Statistics, 2019a).

d Swimming pool

To inhibit bacterial growth and ensure a sanitary environment in swimming pools, regular disinfection of the pool water is necessary (Wang et al., 2002). 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:

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

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.

The HOCl and Cl_2 emissions of the swimming pool are calculated:

$$E_{i,k,l} = V_i \times \sum_k (y \times z \times D) \times c \times f \times R_l \quad (17)$$

where i, k, and l are provinces, indoor and outdoor types (outdoor and indoor swimming pools), and chlorine species, respectively. y is the proportion of indoor and outdoor types, with outdoor swimming pools accounting for 59.43% and indoor swimming pools accounting for 38.93% according to the survey of State Sports General Administration (https://www.sport.gov.cn/, last access: 1 January 2023). 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. z for the dosage, according to the research of Wang et al. (2002), the outdoor swimming pool uses 2g/m^3 of strong chlorine per day, and the indoor swimming pool uses 1.25g/m^3 every day. 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. c is the mass fraction of available chlorine in strong chlorine, which is 90% (Wang et al., 2002). f is the chlorine volatilization ratio, which is 0.2 (Li et al., 2020). R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017).

e Environmental disinfection

Environmental disinfection includes hospital disinfection, breeding disinfection, and toilet disinfection. For hospital disinfection, chlorine emissions are related to the number of hospitals and the amount of disinfectant used, and the emissions are as follows:

$$E_{i,k} = n_i \times U \times c \times f \times R_k \quad (18)$$

where i and k represent provinces and chlorine-containing species, and n is the number of hospitals. U denotes the quantity of disinfectant utilized. In 2007, the average volume of chlorine-containing disinfectant used in Taizhou hospitals was reported to be 2329.2L (Sun et al., 2007). 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. c is the chlorine content of the disinfectant, which is 1000 mg/L. f is the chlorine volatilization ratio, which is 0.3 (Li et al., 2020). R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017).

Breeding disinfection is found in pig farming, poultry farming, and aquaculture. The chlorine emissions from these sources are calculated using the following formula:

$$E_{i,j,k} = S_{i,j} \times U_j \times N_j \times r \times f \times R_k \quad (19)$$

where i, j, and k represent provinces, different breeding types, and chlorine-containing species. S is the breeding area. The breeding areas of pigs and poultry are calculated by the number of pigs and poultry and the breeding density. The number of pigs and poultry and the aquaculture area can be obtained from statistical data (National Bureau of Statistics, 2019b). U is the amount of disinfectant used per unit area, and N is the disinfection frequency. f is the chlorine volatilization ratio, and these parameters can be found in Table S12. r is the proportion of chlorine-containing disinfectants used for breeding, which is 0.065 (Jing et al., 2019). R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017).

Toilet disinfection includes disinfection of public toilets and household toilets. Estimates of emissions during the disinfection of public toilets are as follows:

$$E_{i,k} = n_i \times U \times c \times N \times f \times R_k \quad (20)$$

where i and k represent provinces and chlorine-containing species, and n is the number of public toilets. U is the daily disinfectant dose of each toilet, which is 500 mL. c is the disinfectant concentration, which is 5%. N is the disinfection frequency, which is 365 times/year. f is the chlorine emission rate, which is 0.3. The parameters are all based on the study by Li et al. (2020). R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017). There is too little relevant data on household toilet disinfection, and its emission is estimated to be twice that of public toilets (Li et al., 2020).

f Tap water use

After water treatment, there is still residual chlorine in the tap water at the end of the pipe network, the residual chlorine will be released into the atmosphere during the use of tap water. In this study, we considered the emissions from car washing, lawn watering, road sprinkling, and pipe leaks. For the former three sources, the chlorine emissions were estimated using the following formula:

$$E_{i,j,k} = S_{i,j} \times U_j \times N_j \times CR \times R_k \quad (21)$$

where i, j, and k represent the province, source category, and chlorine species, respectively. S is the number of car washes, green area, and road area. U is the unit water consumption, each car wash uses about 6.6m^3 of car wash water per day (Jing et al., 2014), and the unit water consumption of lawn watering is 1.5L/m^2, and road sprinkling is 1L/m^2 (Li et al., 2020). N is the frequency of water use. Car wash shops use water 365 times a year, lawn watering is 45 times, and road sprinkling is 50 times (Li et al., 2020). CR is the amount of residual chlorine, which is 0.86 mg/L (Li et al., 2020), and R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017).

For the pipeline leaks, the chlorine emissions are estimated as follows:

$$E_{i,k} = W_i \times CR \times f \times R_k \quad (22)$$

where i and k represent provinces and chlorine-containing species. W is the amount of water loss. CR is the amount of residual chlorine, which is 0.86 mg/L (Li et al., 2020). f is the chlorine volatilization ratio, which is 0.1 (Li et al., 2020). R is the release ratio of HOCl (0.84) and Cl_2 (0.11) (Wong et al., 2017).

2.2.7 Usage of pesticide

Active chlorine is also released during pesticide application, thereby enhancing the activity of the atmosphere. The pesticides in this study include insecticides and herbicides. The chlorine emissions from them are calculated using the following formula:

$$E_{i,j} = P_i \times O \times M \div \rho \times A \times c \times f \times R_j \quad (23)$$

where i and j represent provinces and chlorine-containing species, respectively. P is the amount of pesticide usage, O is the proportion of organochlorine pesticides, and the use of organochlorine pesticides accounts for 30% of pesticides (Zhang, 2016). M is the proportion of insecticides and herbicides, 68% and 23% respectively (Zhang, 2016). ρ is the density, the density of the insecticide is 1.359g/ml, the herbicide is mainly liquid, and the density value is 1. A is the active ingredient content, the insecticide is 94.8g/L, and the herbicide c is the chlorine content, the chlorine content of pesticides is 13.08%, and the chlorine content of herbicides is 11.48%. f is the chlorine volatilization ratio, which is 0.3. The above parameters are all based on the research of Yi et al. (2021). 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).

2.3 Spatial allocation

The estimated provincial chlorine emissions were further gridded into a 0.1° × 0.1° grid to derive a gridded, model-ready emission inventory. The spatial allocation of emissions was handled separately according to point sources and area sources. For point sources with clear location identification, including coal-fired emissions from power plants, coal-fired emissions from heating, emissions from waste incineration stations, and biomass open combustion emissions, the point source emissions will be directly located in the grid according to the longitude and latitude coordinates of the point source. For the emissions from power plants and garbage disposal incinerators, the emissions in each province were distributed to each location according to its installed capacity. 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). 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. For the area sources, the provincial emissions were spatially disaggregated onto grid cells using empirically selected spatial proxies such as population density (total, urban, and rural). The detailed spatial allocation factors of each source category and their sources are listed in Table S13.

2.4 Temporal allocation

The annual chlorine emissions estimated above were further allocated to each month using the corresponding activity data or the selected monthly proxies. For the emissions of coal-fired power plants and cooling towers, the monthly emissions are allocated according to the thermal power generation. For the emission of heating coal, the time distribution coefficient is set from mid-November to mid-March according to the heating conditions in different regions of China. For the emissions from the industrial production process and the emissions from pesticide use, the time distribution is based on the output of industrial products and pesticides respectively. The monthly output of thermal power generation, industrial products, and pesticides can be found in the National Bureau of Statistics (https://www.stats.gov.cn/, last access: 1 January 2023). 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. According to the proportion of the number of outdoor and indoor swimming pools, it is assumed that 60% of the swimming pools are open from mid-May to mid-September, 40% of the swimming pools are open all year round, and the time distribution coefficient of the disinfection discharge of the swimming pool is set accordingly. For other coal burning and environmental disinfection emissions that are not sensitive to time change, the monthly emissions can be distributed by days. Other emissions will be allocated according to the existing research. The monthly allocation factors for each source category are presented in Table S14.

3 Results

3.1 Anthropogenic chlorine emission for China (ACEIC)

The ACEIC (Anthropogenic Chlorine Emission Inventory for China) inventory for the year 2018 was developed in this study. The general information of this inventory is shown in Table 1. It includes emissions from 31 provinces in mainland China. We estimated the total emissions of HCl, fine particulate Cl^-, Cl_2, and HOCl in mainland China to be 454, 238, 17, and 73 Gg, respectively. The estimated emissions by source category are presented in Table 2. Figure 1 shows the contribution of different source categories to the total emission. For HCl emission, biomass burning and coal combustion are the primary sources, accounting for 45% (204 Gg) and 40% (180 Gg) of the total, respectively. The emissions from industrial production processes and waste incineration make up 8% (38 Gg) and 7% (31 Gg), respectively. For fine particulate Cl^-, biomass burning is the major contributor to the total emission (78%, 186 Gg), followed by coal combustion (9%, 21 Gg), industrial production process (8%, 20 Gg), and waste incineration (4%, 10 Gg). For Cl_2, it is mainly from the usage of chlorine-containing disinfectants

(56%, 9 Gg) and coal combustion (43%, 7 Gg). For HOCl emissions, the usage of chlorine-containing disinfectant is the major contributor (99%, 72 Gg). We note that the usage of chlorine-containing disinfectants is the major source of Cl_2 and HOCl. Figure 2 presents the proportion of different usages of chlorine-containing disinfectants. The waste water treatment account for 42% of the total, followed by water treatment (23%), swimming pool (15%), environmental disinfection (13%), tap water use (5%), and cooling tower (2%).

3.2 Anthropogenic chlorine emission from different economic sectors

We aggregated the anthropogenic chlorine emissions from 41 specific sources into 5 economic sectors, including power, industry, residential, agricultural, and biomass burning (Table S2). Table 3 provides the estimated emissions for HCl, fine particulate Cl^-, Cl_2, and HOCl, while Figure 3 illustrates the contribution of each economic sector to the total emissions. HCl emissions were predominantly attributed to biomass burning (45%, 204 Gg), industry (35%, 160 Gg), and the residential sector (15%, 67 Gg). For fine particulate Cl^- emissions, biomass burning accounted for the majority with 78% (186 Gg), followed by the industry sector with 14% (34 Gg). Cl_2 emissions were primarily sourced from the residential sector (59%, 10 Gg) and the industry sector (31%, 5 Gg). As for HOCl emissions, the residential sector dominated, contributing 90% (65 Gg) of the total.

3.3 Anthropogenic chlorine emission in different provinces

Fig. 4 and Table 4 display the regional variations in anthropogenic chlorine emissions across different provinces. 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.

The spatial distribution of anthropogenic chlorine emissions reveals distinct patterns when considering per-unit-area (Fig. S1) and per-capita (Fig. S2) intensity by province. 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.

3.4 Spatial distribution of the emission

Figure 5 showcases the intricate spatial distribution of anthropogenic chlorine emissions with a high-resolution granularity of 0.1° × 0.1°. Generally, eastern China exhibits significantly higher emissions compared to Western China. The distribution pattern of HCl and fine particulate Cl^- closely resemble one another, attributable to their primary sources: biomass burning and coal combustion. Similarly, Cl_2 and HOCl display comparable distribution patterns due to their predominant contribution from the usage of chlorine-containing disinfectants. Regarding HCl emissions, elevated levels are primarily concentrated in key regions such as the North China Plain (NCP), Northeast China, Sichuan Basin (SCB), Yangtze River Delta (YRD), and Pearl River Delta (PRD). The spatial distribution pattern of fine particulate Cl^- closely mirrors that of HCl emissions, except for slightly lower emissions observed in the PRD region. For Cl_2 and HOCl, regions with high emissions encompass the PRD, YRD, Beijing-Tianjin-Hebei region (BTH), SCB, and the central area of Hubei province. These regions exhibit higher population densities and economic development levels, which stimulate the demand for cleaning products and consequently contribute to elevated chlorine emissions.

We also present the spatial distribution of anthropogenic chlorine in various energy sources (Fig. S3) and sectors (Fig. S4). In terms of energy consumption sources, coal emissions are mainly concentrated in North China, Central China, Southwest China, and Northwest China. The emissions from industrial production and waste treatment are mainly concentrated in the BTH, YRD, PRD, and SCB. The spatial distribution of emissions from biomass combustion is similar to that of pCl emissions, while the emissions from disinfectant use are similar to the spatial distribution of Cl_2 and HOCl emissions. Concerning the sector-based distribution, the distribution of power plants is similar to the distribution of coal emissions. The emissions from industrial

and civil sectors are mainly concentrated in BTH, YRD, PRD, and SCB. The agricultural sector is mainly concentrated in the southern region. Biomass combustion, as mentioned above, is similar to the distribution of pCl.

3.5 Temporal variation of the emission

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.

The emissions of Cl_2 and HOCl show a different temporal variation pattern from those of HCl and pCl. Their monthly variations are primarily driven by the residential sector, particularly the usage of chlorine-containing disinfectants. Emission peaks occur during summer, corresponding to the high demand for water disinfection. Lower emissions of Cl_2 and HOCl are observed in February.

4 Discussion

4.1 Comparison with previous studies

We compared our results with the chlorine emissions in China in recent years from other previous studies (Table 5). We regrouped their emission into our source category for a comprehensive comparison. The HCl emission in this study is higher than the emissions in our previous study due to the inclusion of emissions from industrial processes and biomass burning. The HCl emission from coal combustion in this study (2018) is lower than those in 2012 and 2014 due to the reduction of coal consumption in recent years after the implementation of the Clean Air Action in China in 2013. The HCl emission from waste incineration in this study is higher because our previous study only included emissions from garbage incinerators. We note that the emissions from garbage incinerators increased ~ 2 times from 2014 to 2018, which is due to the increased number of garbage incinerators built in China in recent years. The total HCl emission estimated by Fu et al. (2018), Zhang et al. (2022), and Yin et al. (2022) are 458 Gg (2014), 705 Gg (2014), 270 Gg (2019), respectively. Our estimations (454 Gg) are within the

range of their results. In this study, HCl emissions from coal combustion are about twice as high as those reported by Fu et al. (2018) and Yin et al. (2022), but significantly lower than the estimation by Zhang et al. (2022). This discrepancy primarily arises from the varied application of emission factors and control technologies. For coal combustion, our study relies on source data derived from on-site observations, differing from Fu et al. (2018) who used control technology application ratios based on national policies, Yin et al. (2022) who employed foreign application ratios, and Zhang et al. (2022) who used an S-curve formula leading to overestimations. Our control technology application ratios are based on domestic research literature, which is deemed more reasonable. Furthermore, our study reports substantially lower HCl emissions from waste incineration compared to Fu et al. (2018) and Zhang et al. (2022). This reduction is attributed to advancements in China's municipal solid waste management, with greater emphasis on centralized waste collection and reduced open burning compared to the situation in 2014. Additionally, considering the economic disparities, urbanization, and uneven waste treatment technology adoption among provinces, we have adopted a differentiated approach, utilizing waste treatment rates from the Statistical Yearbook to estimate open burning ratios for each province. This approach offers a more nuanced representation of provincial differences compared to the uniform national open burning ratios employed by Yin et al. (2022) and Zhang et al. (2022). In our study, biomass combustion is a notable contributor to HCl emissions, primarily due to disparities in estimations of household combustion rates and outdoor burning methods compared to previous research. Our household combustion rates are substantiated by documented sources, while our approach to open burning employs a bottom-up emission factor method, mitigating potential underestimations resulting from satellite-based detection, which may overlook small-scale or short-term fire events. Moreover, our estimate of HCl emissions in the Yangtze River Delta (YRD) region is approximately twice as high as that reported by Yi et al. (2021), who underestimated HCl emissions due to their exclusive focus on centralized coal combustion, neglecting dispersed coal combustion. In contrast, our study not only encompasses urban waste incineration facilities but also considers emissions from county towns and open burning.

The total emission of fine particulate Cl^- in this study is within the range of the emissions reported by Fu et al. (2018), Zhang et al. (2022), and Yin et al. (2022). All of these studies highlight biomass burning as the major contributor (78%, 75%, 57%, and 78% in respective studies). The emissions from coal combustion, industrial processes, and biomass burning are generally consistent across these studies. Regarding waste incineration, emissions reported by Fu et al. (2018) and Zhang et al. (2022) are notably higher than those in our study and Yin et al. (2022). This difference can be attributed to significant improvements in China's municipal solid waste management since 2014, with a greater emphasis on centralized waste collection and reduced open burning, leading to decreased emissions. 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. The estimated emissions from the YRD in this study are higher than those from Yin et al. (2022) because our activity data is considered more comprehensively.

The total emission of Cl_2 in this study is higher than our previous studies because we included additional emissions from usages of disinfectants and pesticides. Yin et al. (2022) and our studies both demonstrated the emission from usages of chlorine-containing disinfectants is as important as those from coal combustion. However, the total Cl_2 emission in this study is ~2 times higher than those estimated in the study of Yin et al. (2022). In this study, emissions from coal combustion are roughly twice as high as those in Yin et al. (2022) due to the use of locally measured emission factors and the application ratio of domestic control technologies. The emissions from disinfectant use in this study are about three times higher than those in Yin et al. (2022). This increase is attributed to the expanded sources of emissions considered in this study, as well as improvements in the calculation methods for chlorine addition in swimming pools. Firstly, this study includes emissions from county-level water treatment, county-level wastewater treatment, tap water usage, and environmental disinfection processes, accounting for 4%, 6%, 5%, and 13% of the total emissions from disinfectant use, respectively. These sources are not negligible. Secondly, localized improvements were made to some emission factors. Regarding the chlorine addition in swimming pools, while Yin et al. (2022) used a concentration of 1 mg/L from national standards, this study cited experimental research literature and used outdoor and indoor chlorine addition rates of 1.8 mg/L and 1.125 mg/L, respectively. Lastly, we improved the estimation method for swimming pool emissions by distinguishing between the open times of indoor and outdoor pools. Unlike the assumption in Yin et al. (2022) that all pools are open in the summer, this study assumes year-round operation for indoor pools and only summer operation for outdoor pools, which is a more realistic scenario. This led to increased disinfectant use in swimming pools, resulting in higher emissions compared to Yin et al. (2022). The estimated emissions from the YRD in this study are higher than those from Yi et al. (2022) because we consider dispersed coal combustion during coal combustion, and activity data such as water treatment, wastewater treatment, and environmental disinfection also consider rural areas. The estimated emissions in Shanghai are half that of Li et al. (2020) because their main source of emissions is overestimated from cooling towers, which was also mentioned in the study of Yin et al. (2022).

The research on the development of HOCl emission inventory is limited. This study and Yin et al. (2022) both showed that the emission from the usage of chlorine-containing disinfectants is the major contributor to HOCl emission. However, we estimated the HOCl emission to be ~ 3 times higher than those estimated by Yin et al. (2022). As mentioned above, this higher

estimation can be attributed to the addition of emissions from swimming pools, environmental disinfection, tap water use, and pesticides.

4.2 Uncertainty and limitation

We applied the Monte Carlo method to quantify the uncertainties of the ACEIC inventory. Normal distributions with coefficients of variation (CV) ranging from 5% to 50% were assumed for activity data according to previous studies (Yi et al., 2021; Li et al., 2020; Zheng et al., 2022; Fu et al., 2018). For the emission factors, probability distributions were fitted for parameters with adequate measurement data. For parameters with limited measurement data, probability distributions were assumed as uniform or log-normal distributions. The detailed uncertainty assumptions for the activity data and emission factors are summarized in Tables S15 and S16. 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.

Indeed, the ACEIC inventory has its limitations, and further studies are necessary to develop a more comprehensive chlorine emission inventory in China. Some of the areas that require attention and improvement include: (1) More localized measurements and studies on emission factors: There is a need for additional local measurements and studies to obtain more accurate and representative emission factors for chlorine emission. This is particularly important for sectors such as waste treatment, where more localized observation data is needed to improve the accuracy of emission estimates. (2) Inclusion of emissions from additional anthropogenic activities: 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 chlorine species. (3) Application of specific spatial and temporal allocation factors: Temporal allocation factors, which account for the varying contributions of different sectors over time, need further refinement. The proportion of departmental contributions in each province can differ, resulting in distinct temporal characteristics of emissions. By addressing these limitations and conducting further research in these areas, it will be possible to develop a more robust and comprehensive chlorine emission inventory for China.

5 Conclusion and implication

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. This inventory includes 4 chlorine species (HCl, fine particle Cl^-, Cl_2, and HOCl) and 41 specific anthropogenic sources. We estimate that the total emissions of HCl, fine particle Cl^-, Cl_2, and HOCl in mainland Chinese in 2018 were 454 (-48%~45%), 238 (-59%~89%), 17 (-44%~58%) and 73 (-44%~79%) Gg, respectively.

In terms of energy consumption sources, for HCl emissions, biomass combustion and coal combustion are the main sources, accounting for 45% and 40% of the total, respectively. For fine particle Cl^-, biomass combustion is the main contributor (78%) to the total emissions. For Cl_2, it mainly comes from the use of chlorine-containing disinfectants (56%) and coal burning (43%). For HOCl emissions, the use of chlorine-containing disinfectants is the main reason (99%). Among them, waste water treatment (42%) and water treatment (23%) are the human activities with the highest emissions during the use of chlorine-containing disinfectants.

From the perspective of the economic sector, HCl emissions mainly come from the biomass combustion (45%) and industrial (35%) sectors. Biomass combustion and the industrial sector are the main sources of fine particulate chlorine, accounting for 78% and 14% of total emissions, respectively. Cl_2 emissions mainly come from the residential and industrial sectors, accounting for 59% and 31% of the total emissions, respectively. HOCl emissions mainly come from the residential (90%) sector. The use of chlorine-containing disinfectants is the most important source of Cl_2 and HOCl in the residential sector.

From the perspective of provincial emissions, Heilongjiang has the highest emissions of HCl (38.27 Gg) and fine particle Cl^- (27.18 Gg). Guangdong has the highest emissions of Cl_2 (1.40Gg) and HOCl (8.61Gg). High HCl and fine particle Cl^- emission areas are located in the northeast, North China Plain, and Sichuan Basin, while Cl_2 and HOCl emission areas are located in the Pearl River Delta, Yangtze River Delta, and Beijing-Tianjin-Hebei region. In terms of monthly changes, due to active agricultural activities, the emissions of HCl and fine particle Cl^- are relatively high during early spring (February to April) and early autumn (August to October), while the emissions of Cl_2 and HOCl are higher in summer due to the high demand for corresponding disinfection water.

The results of this study demonstrate that chlorine-containing disinfectants are significant sources of Cl_2 and HOCl. The demand for these disinfectants has risen substantially in recent years due to the spread of COVID-19 worldwide. It is

anticipated that the increase in chlorine emissions following the outbreak of COVID-19 will play a more important role in tropospheric chemistry. Furthermore, the long-term trends of chlorine emissions are largely unknown due to most studies focusing on emissions from a specific year. Therefore, an inter-annual emission inventory of chlorine species is essential for controlling emissions. It is worth noting that Cl_2 and HOCl emissions typically peak during the summer when severe O_3 pollution frequently occurs in many parts of China. Therefore, future investigations should assess the impact of chlorine emissions on summertime O_3 formation. This inventory provides valuable insights into the anthropogenic sources of chlorine species and is conducive to the development of emission control strategies to mitigate secondary pollution in China. We suggest that air quality modeling studies include the chlorine emission inventory to accurately simulate tropospheric chlorine chemistry.

Author contributions

Y.M.L. and Q.F. initiated the research. Y.M.L. designed the research framework. S.T.L. and Y.M.L. collected the materials. S.T.L. calculated the emissions and drew the figures. Y.M.L. and S.T.L. analyzed the results and wrote the paper with inputs from all authors. All authors contributed to the discussion and improvement of the paper.

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Competing interests

The authors declare that they have no conflict of interest.

Code/Data availability

The code or data used in this study are available upon request from Yiming Liu (liuym88@mail.sysu.edu.cn) and Siting Li (list23@mail2.sysu.edu.cn).

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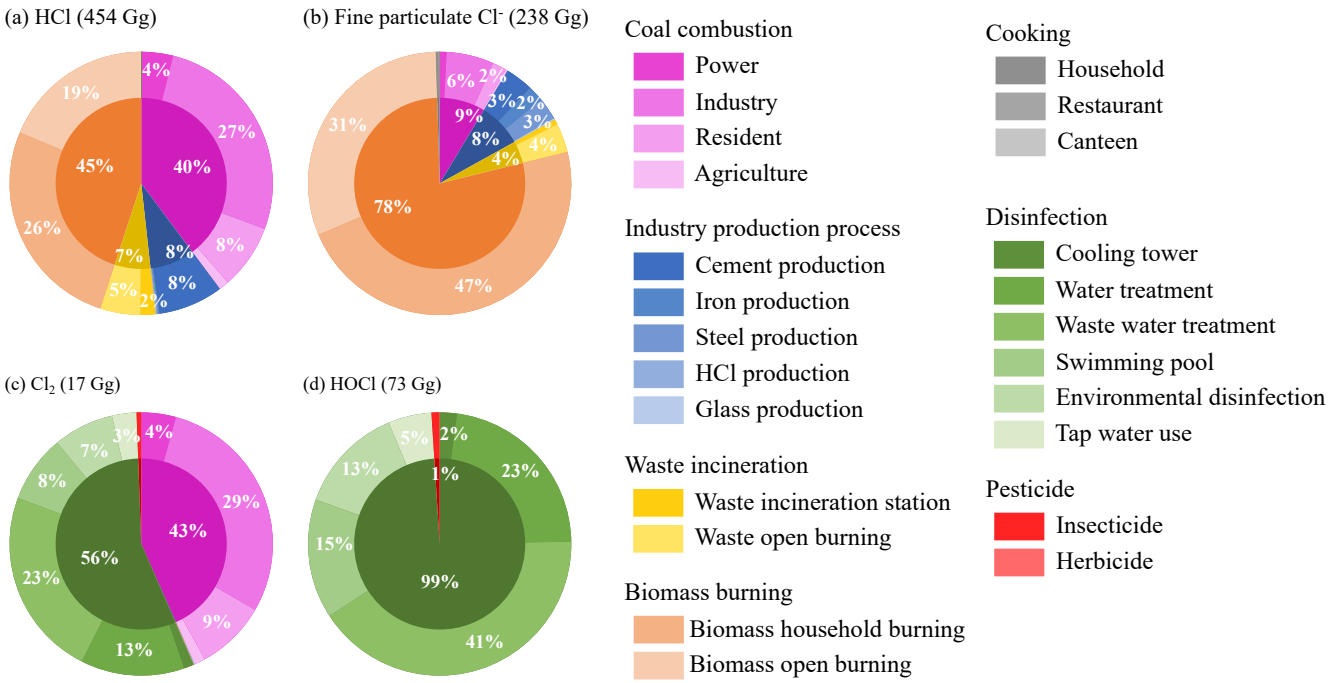

Figure 1 Contributions of different source categories to the anthropogenic emissions of HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) in 2018. The aggregation of 7 primary source categories from 41 specific sources can be found in Table S1.

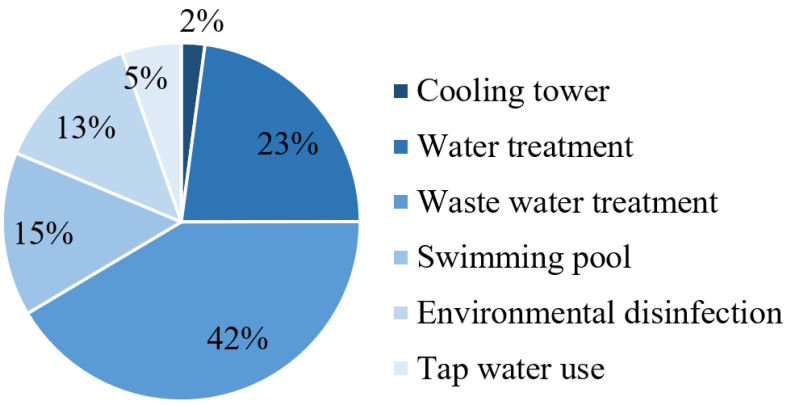

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Figure 2 Proportion of chlorine emissions from different usages of chlorine-containing disinfectant in China.

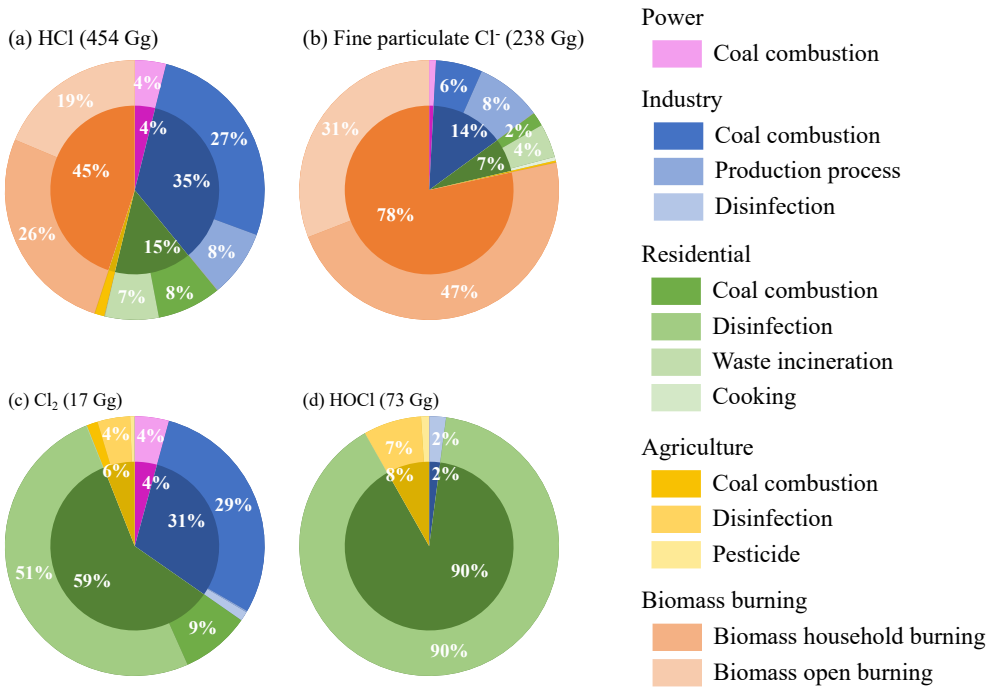

Figure 3 Contributions of different economic sectors to the anthropogenic emissions of HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) in 2018. The aggregation of 5 economic sectors from 41 specific sources can be found in Table S2.

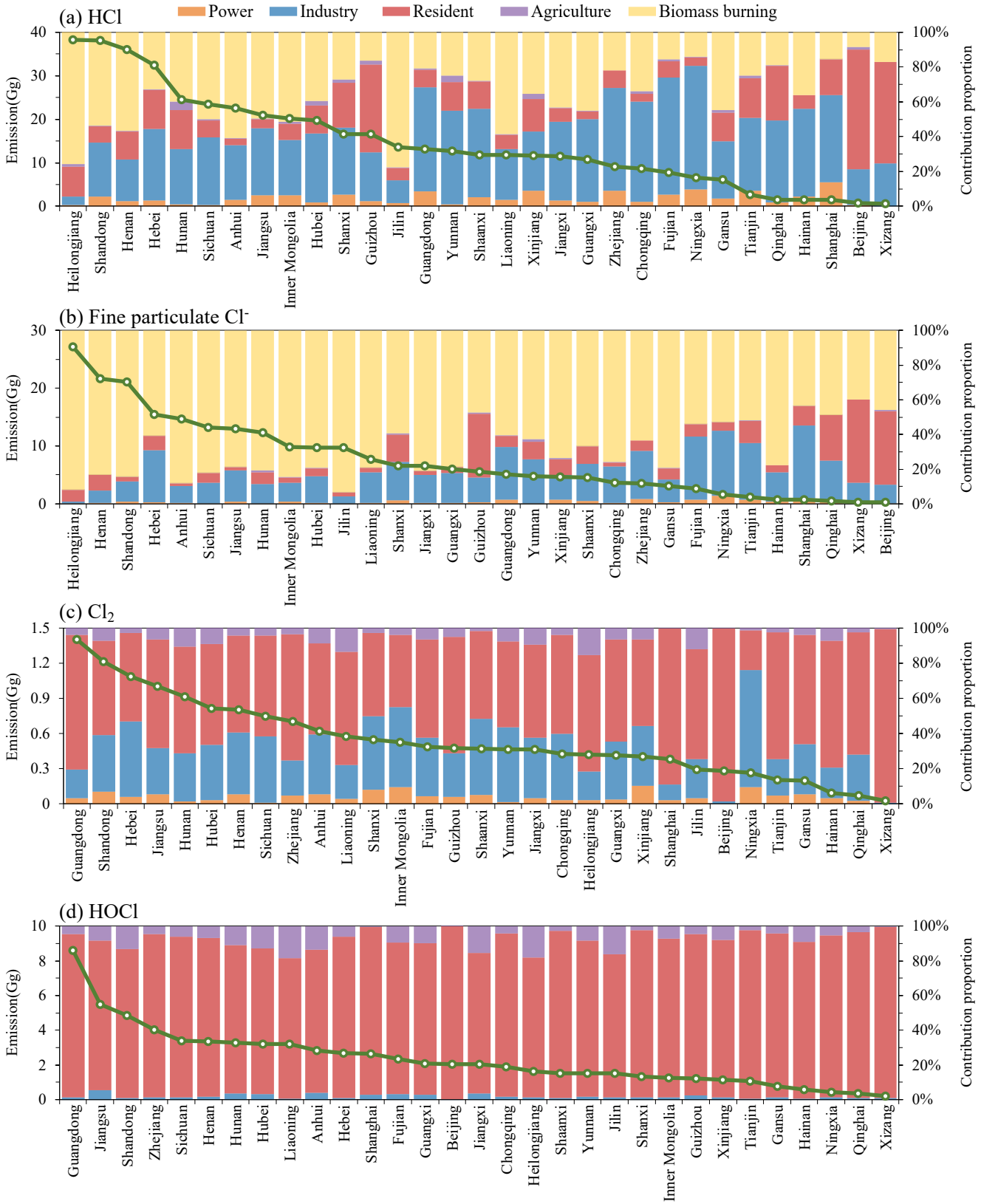

Figure 4 Emissions (green line) and contribution proportions of HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) by province in 2018.

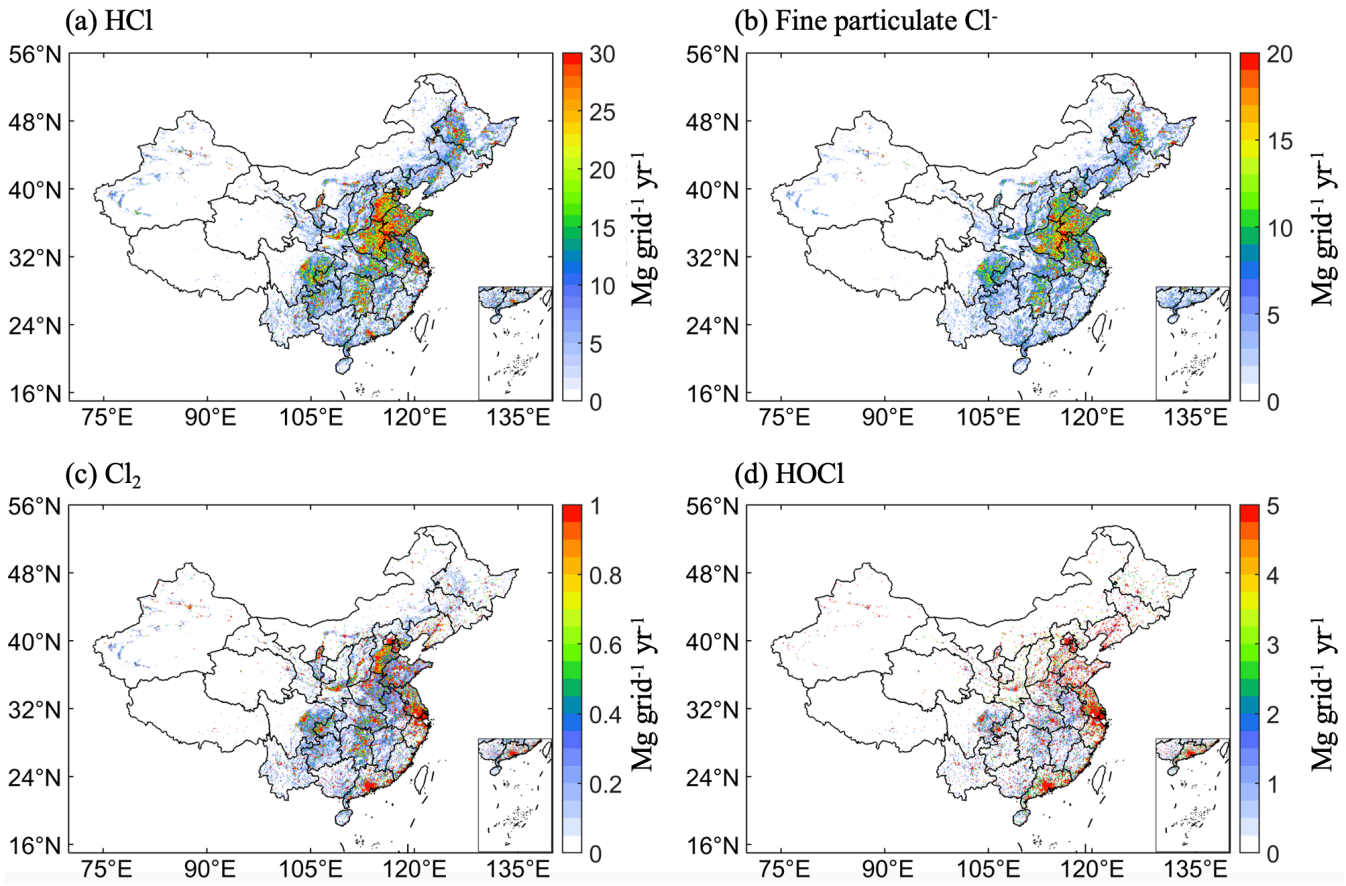

Figure 5 Spatial distribution of anthropogenic HCl, fine particulate Cl^-, Cl_2, and HOCl emissions in 2018 at 0.1° × 0.1° resolution.

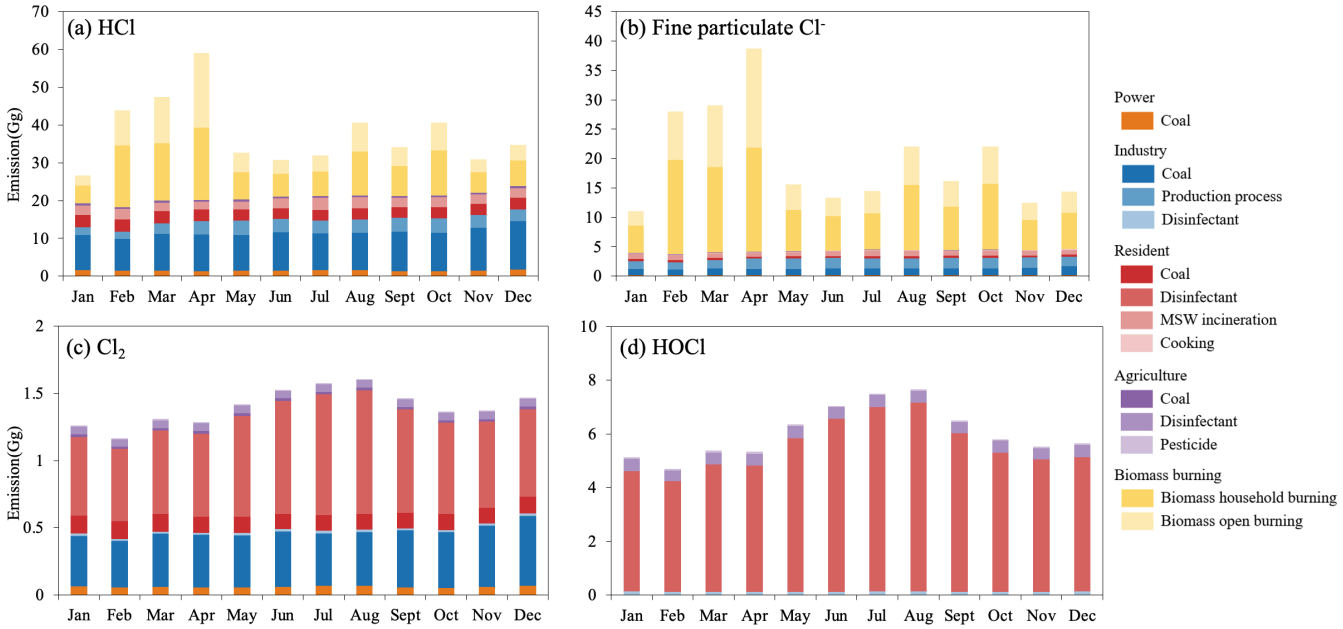

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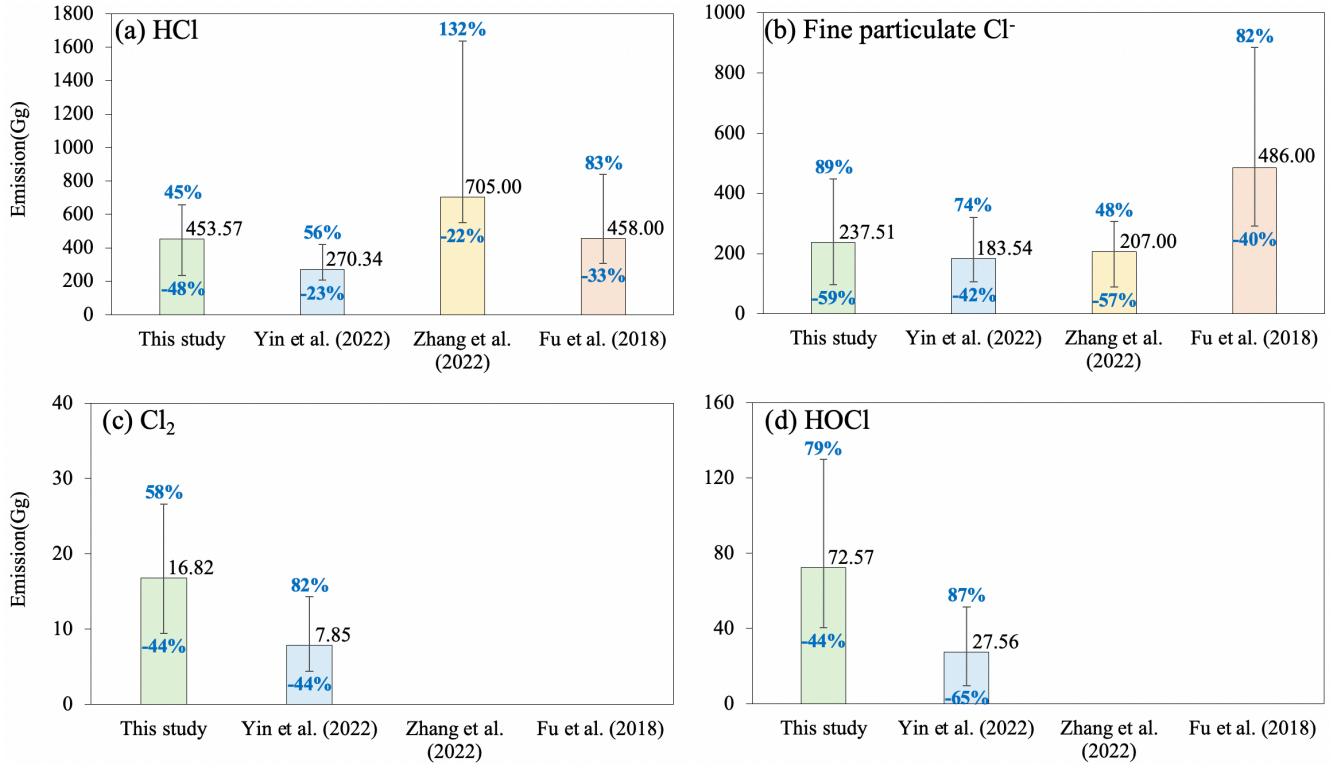
Figure 6 Monthly variation of anthropogenic HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) emissions by economic sector in 2018.



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

Table 1 General information about the ACEIC inventory

Item	Information
Name	Anthropogenic chlorine emissions for China (ACEIC)
Domain	31 provinces in mainland China
Species	HCl, fine particulate Cl^-, Cl_2, and HOCl
Source categories	7 major categories: (1) coal combustion, (2) industrial production process, (3) waste incineration, (4) biomass burning, (5) cooking, (6) usages of chlorine-containing disinfection, and (7) usages of pesticide
Base year	2018
Spatial resolution	$0.1^o \times 0.1^o$
Temporal resolution	Monthly

860

Table 2 Anthropogenic chlorine emission in China by source category in 2018.

Source category	Sub-category	Emission (Gg)			
		HCl	pCl	Cl_2	HOCl
Coal combustion	Power	17.69	2.01	0.71	
	Industrial	121.18	13.77	4.89	
	Residential	36.00	4.09	1.45	
	Agriculture	5.66	0.64	0.23	
	Sum of coal combustion	180.53	20.51	7.29	0.00
Industrial production process	Cement production	35.99	8.06		
	Iron production	0.46	4.64		
	Steel production	0.74	6.57		
	HCl production	0.62			
	Flat glass production	0.54	0.47	0.03	
	Sum of industrial production process	38.35	19.74	0.03	0.00
Waste incineration	Incineration station	8.32	1.52		
	Open burning	22.43	8.47		
	Sum of waste incineration	30.76	9.99	0.00	0.00
Biomass burning	Household burning	119.23	112.72		
	Open burning	84.70	73.42		
	Sum of biomass burning	203.93	186.13	0.00	0.00
Cooking	Household		0.72		
	Restaurant		0.36		
	Canteen		0.05		
	Sum of cooking	0.00	1.13	0.00	0.00
Disinfectant	Cooling tower			0.20	1.53
	Water treatment			2.15	16.41
	Waste water treatment			3.90	29.81
	Swimming pool			1.39	10.65
	Environment disinfectant			1.25	9.58
	Tap water use			0.51	3.87
	Sum of disinfectant	0.00	0.00	9.41	71.85
Pesticide	Insecticide			0.09	0.71
	Herbicide			0.01	0.02
	Sum of pesticide	0.00	0.00	0.10	0.73
Sum of all categories		453.57	237.51	16.82	72.57

865 Table 3 Anthropogenic chlorine emission in China by economic sector in 2018.

Sector	Subsector	Emission (Gg)			
		HCl	pCl	Cl_2	HOCl
Power	Coal combustion	17.69	2.01	0.71	
Industry	Industrial coal combustion	121.18	13.77	4.89	
	Industrial production process	38.35	19.74	0.03	
	Industrial usage of disinfectant	0.00	0.00	0.20	1.53
	Sum of industry	159.53	33.51	5.12	1.53
Residential	Residential coal combustion	36.00	4.09	1.45	
	Residential usage of disinfectant			8.52	65.09
	Waste incineration	30.76	9.99		
	Cooking		1.13		
	Sum of residential sector	66.75	15.21	9.98	65.09
Agriculture	Agricultural coal combustion	5.66	0.64	0.23	
	Agricultural usage of disinfectant			0.69	5.23
	Agricultural usage of pesticide			0.10	0.73
	Sum of agriculture	5.66	0.64	1.01	5.96
Biomass burning	Biomass household burning	119.23	112.72		
	Biomass open burning	84.70	73.42		
	Sum of biomass burning	203.93	186.13		
Sum of all sectors		453.57	237.51	16.82	72.57

Table 4 Anthropogenic chlorine emissions by province in 2018.

Province	Emission (Gg)			
	HCl	pCl	Cl_2	HOCl
Beijing	0.77	0.24	0.28	2.05
Tianjin	2.75	1.22	0.20	1.06
Hebei	32.46	15.46	1.09	2.67
Shanxi	16.65	6.62	0.55	1.33
Inner Mongolia	20.21	9.89	0.53	1.27
Liaoning	11.86	7.66	0.58	3.19
Jilin	13.66	9.67	0.29	1.50
Heilongjiang	38.27	27.18	0.42	1.64
Shanghai	1.44	0.69	0.38	2.66
Jiangsu	20.98	12.95	1.00	5.50
Zhejiang	9.18	3.58	0.70	4.04
Anhui	22.62	14.67	0.62	2.84
Fujian	7.81	2.69	0.49	2.34
Jiangxi	11.53	6.57	0.46	2.03
Shandong	38.10	21.13	1.22	4.86
Henan	36.05	21.60	0.80	3.35
Hubei	19.69	9.69	0.81	3.22
Hunan	24.45	12.36	0.91	3.27
Guangdong	13.19	5.13	1.40	8.61
Guangxi	10.83	5.98	0.41	2.09
Hainan	1.47	0.74	0.10	0.58
Chongqing	8.62	3.70	0.43	1.88
Sichuan	23.41	13.18	0.75	3.39
Guizhou	16.53	5.56	0.48	1.22
Yunnan	12.69	4.79	0.47	1.52
Xizang	0.61	0.28	0.03	0.20
Shaanxi	11.87	4.50	0.47	1.53
Gansu	6.15	3.04	0.20	0.77
Qinghai	1.47	0.50	0.07	0.34
Ningxia	6.62	1.59	0.27	0.44
Xinjiang	11.63	4.67	0.40	1.13
Sum	453.57	237.51	16.82	72.57

870

Table 5 Comparison of anthropogenic chlorine emissions with other studies. The values in brackets are the proportion of different source categories.

Species	Study	Year	Total	Coal combustion	Industrial process	Waste incineration	Biomass burning	Cooking	Usage of disinfectant	Usage of pesticide
HCl	This study	2018	453.57	180.53 (40%)	38.35 (8%)	30.76 (7%)	203.93 (45%)			
(Gg)	Liu et al. (2018)	2012	235.80	232.90 (99%)		2.90 (1%)				
	Hong et al. (2020)	2014	223.40	219.20 (98%)		4.20 (2%)				
	Fu et al. (2018) [a]	2014	458.00	87.00 (19%)	36.60 (8%)	187.80 (41%)	146.60 (32%)			
	Zhang et al. (2022) [a]	2014	705.00	310.2 (44%)	21.15 (3%)	338.40 (48%)	35.25 (5%)			
	Yin et al. (2022)	2019	270.34	80.61 (30%)	57.09 (21%)	35.01 (13%)	97.63 (36%)			
pCl	This study	2018	237.51	20.51 (9%)	19.74 (8%)	9.99 (4%)	186.13 (78%)	1.13 (1%)		
(Gg)	Fu et al. (2018) [a]	2014	486.00	24.30 (5%)	29.20 (6%)	68.00 (14%)	364.50 (75%)			
	Zhang et al. (2022) [a]	2014	207.00	20.70 (10%)	16.56 (8%)	51.75 (25%)	117.99 (57%)			
	Yin et al. (2022)	2019	183.54	11.86 (6%)	11.88 (6%)	9.78 (5%)	143.19 (78%)	6.85 (4%)		
Cl_2	This study	2018	16.82	7.29 (43%)	0.03 (0%)				9.41 (56%)	0.10 (1%)
(Gg)	Liu et al. (2018)	2012	9.40	9.40 (100%)						
	Hong et al. (2020)	2014	8.90	8.90 (100%)						
	Yin et al. (2022)	2019	7.85	3.46 (44%)	0.78 (10%)				3.61 (46%)	
HOCl	This study	2018	72.57						71.85 (99%)	0.73 (1%)
(Gg)	Yin et al. (2022)	2019	27.56						27.56 (100%)	

[a] The emissions from different source categories are estimated by multiplying the total emission with the corresponding proportions reported in the literature.