https://doi.org/10.5194/egusphere-2023-1448
Preprint. Discussion started: 21 July 2023

ACEIC: a comprehensive anthropogenic chlorine emission inventory for China

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Abstract. Reactive 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. However, their anthropogenic sources remain uncertain and require further investigation. In previous studies, we developed the Anthropogenic Chlorine Emission Inventory for China (ACEIC) for the years 2012 and 2014. This inventory focused solely on the emissions of hydrogen chloride (HCl) and chlorine gas (Cl_2) from coal combustion and prescribed 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). The emission factors used in this updated inventory were primarily based on localized survey data. 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 summer (June-July) and 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

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anthropogenic sources of reactive chlorine species and can aid in the formulation of emission control strategies to mitigate secondary pollution in China.

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1 Introduction

Recent field and laboratory studies have revealed the crucial role of chlorine atoms (Cl) in tropospheric chemistry (Faxon and Allen, 2013; Qiu et al., 2019; 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., 2019; 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.

Anthropogenic activities are an important source of reactive chlorine species, including HCl, fine particulate Cl^- (pCl), Cl_2, and HOCl, which can produce Cl radicals. HCl can react with OH radicals to release Cl atoms (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 atoms 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 atoms, while HOCl can also be photolyzed to release Cl atoms (Finlayson-Pitts, 1993; Faxon and Allen, 2013). These reactive chlorine species are emitted from various anthropogenic sources (Chang and Allen, 2006; Yin et al., 2022). However, research on anthropogenic chlorine emission inventories in China is currently limited, and the temporal and spatial distribution of these emissions remains unclear. The wide range of anthropogenic chlorine sources in the atmosphere also poses challenges in establishing accurate emission inventories. 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. Therefore, it is of significant importance to develop a comprehensive inventory of anthropogenic chlorine emissions in China. Such an inventory 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.

Research on anthropogenic chlorine emission inventories in foreign countries began to develop in the early 21st century. In 1999, Mcculloch et al. (1999) established a global anthropogenic chlorine emission inventory called Reactive Chlorine Emissions Inventory (RCEI) based on statistical data from 1990. This inventory primarily focused on chlorine emissions from coal combustion and waste incineration. However, due to the rapid industrial and economic growth in China, the RCEI

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inventory established approximately 30 years ago does not accurately reflect the current situation of atmospheric chlorine emissions in China. In 2018, our team (Liu et al., 2018) has firstly established the Anthropogenic Chlorine Emission Inventory for China (ACEIC) based on the relevant basic data in 2012, which mainly considered emission of HCl and Cl_2 from coal combustion and prescribed waste incineration. The study found that the North China Plain, the Yangtze River Delta and the Sichuan Basin are the high-value areas of anthropogenic chlorine emissions in China. The ACEIC inventory has been successfully applied to the WRF-CMAQ model for simulating atmospheric pollutants. Subsequently, we updated the ACEIC inventory to the year 2014 (Hong et al., 2020). Compared to 2012, the total amount of HCl emitted from coal combustion in China decreased by approximately 6% in 2014, while the total amount of HCl emitted from waste incineration stations increased by around 45%.

In addition to our studies, there are some researches on anthropogenic chlorine emission inventory in China. Fu et al. (2018) also established a set of China's anthropogenic chlorine emission inventory, which considered HCl and fine particulate Cl^- emissions from coal combustion, industrial production processes, biomass combustion, and waste incineration The study pointed out that Northeast Plain and North China Plain are high emission areas of HCl. Zhang et al. (2022) developed a comprehensive global emission inventory of gaseous HCl and particulate Cl^- (pCl) from 1960 to 2014, including China. They estimated the emissions of HCl and pCl in China to be 707 Gg and 207 Gg, respectively. In recent years, 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). The results of Yin et al. (2022) found that the Cl_2, HOCl, HCl, and pCl emissions in mainland China in the year 2019 are estimated to be 7.8, 27.6, 270.3, and 183.5 Gg, respectively, with the major source contributors being use of chlorine-containing disinfectant (46%), use of chlorinecontaining disinfectant (100%), biomass burning (36.1%), biomass burning (78.0%), respectively. Despite these studies, 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). A comprehensive anthropogenic chlorine emission inventory which integrates the advantages of these studies is thus warranted.

Previously, we have preliminarily established the anthropogenic chlorine emission inventory for China (ACEIC) based on the basic data of 2012 and 2014 (Liu et al., 2018; Hong et al., 2020), but it only considered the HCl and Cl_2 emissions from coal combustion and HCl emissions from waste incineration stations. In this study, we further update and improve this anthropogenic chlorine emissions by including more reactive chlorine species and more completed sources, with 2018 as the base year. Section 2 demonstrates the data and method utilized to develop the updated chlorine emission inventory ACEIC.

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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 reactive chlorine species (HCl, fine particulate Cl^-, Cl_2, HOCl) from various anthropogenic activities across the 31 provinces in mainland China. These emissions are categorized into seven major source categories: (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.

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

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

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

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 reactive chlorine gases. In this study, the release ratio is assumed to be 0.84 for HOCl and 0.11 for Cl_2.

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. Generally, the activity data were obtained from the yearbook, government statistics, and Gaode's POI data. The emission data were collected and selected mainly 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. Table S1-S10 in the supplementary data summarize the activity data and emission factor.

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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, 2019a). 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):

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

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where i, j, k, l is the province, source sector, reactive 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
140 energy for a 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 denotes the ratios of the molar mass of chlorine atom to the molecular weight of reactive chlorine species (35.5/36:5, 1, 1 for HCl, fine particulate Cl^-, and Cl_2).

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2.2.2 Industrial production process

Reactive 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, 2019b), and those of industrial HCl was obtained from the National Bureau of Statistics (https://m.sohu.com/a/335035620_775892/?pvid=000115_3w_a, last access: 1 January 2023).

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For HCl and Cl_2, the emission factors of industrial productions of cement, iron, steel, hydrochloric acid, and flat glass, were obtained from the literature (see Table S2 and S3). 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 S4.

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.

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, 2019c). The emissions of HCl were calculated using the method purposed 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). 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%).

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 Table S2 and S4. η is the removal efficiency of $PM_{2.5}$ (99%) in the garbage incineration station (Nan, 2016).

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For the open waste incineration, there are 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 (Fu et al., 2018). F is the proportion of waste open burning of the total. Due to the imbalance of economy, urbanization, and garbage disposal technology popularization, the F value varied in different provinces. 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^- were shown in Table S2 and S4.

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 S5 and S6.

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 Table S2 and S7, respectively.

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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 S8.

2.2.6 Usage of chlorine-containing disinfectant

The emissions from disinfection 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 reactive chlorine species, respectively. W is the amount of chlorine-containing water. 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 reactive chlorine gases. Except for the activity data (W), other parameters in this equation for different usages of disinfectant are presented in Table S3.

a Cooling tower

In order 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 reactive 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:

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

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 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 estimation of emissions from water treatment relies on 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, 2019c).

c Waste water treatment

Waste water treatment encompasses the treatment of both medical waste water and domestic sewage. Prior to 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 Bureau of Statistics, 2019e). 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, 2019c).

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d Swimming pool

In order 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 with pool volumes of:

$$V_{i,j} = n_i \times \sum_j (a \times b \times h \times r) \quad (15)$$

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

where i and j represent provinces and size types. Swimming pool size types include standard swimming pools, semi-standard, and non-standard swimming pools. n is the number of swimming pools. The number of public swimming pools comes from the State Sports General Administration (https://www.sport.gov.cn/, last access: 1 January 2023), and the number of private swimming pools is estimated based on the ratio of residents' income to the number of swimming pools (Li et al., 2020). a, b, and h are the length, width, and depth of the swimming pool, as shown in Table S9. r represents the proportion of swimming pool types, with standard swimming pools accounted for 28%, semi-standard and non-standard swimming pools accounted for 72% (Li et al., 2020). 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 chlorinated 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). 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 of the swimming pool, and the outdoor swimming pool in this study is open all year, the indoor 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. R is the release ratio of HOCl and Cl_2.

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:

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

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). Considering the total health expenditure and the corresponding ratio, it is estimated to be 11,898.0 L in 2018. 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 and Cl_2.

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 farming types, and chlorine-containing species. S is the farming area, the pig and poultry farming area is calculated by the number of farming and the farming density, and the aquaculture area can be obtained directly from statistical data. U is the amount of disinfectant used per unit, and N is the disinfection frequency. f is the chlorine volatilization ratio, and these parameters can be found in Table S10. r is the proportion of chlorine-containing disinfectants used in the farm, which is 0.065 (Jing et al., 2019). R is the release ratio of HOCl and Cl_2.

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 and Cl_2. There are too little relevant data on household toilet disinfection, and its emission is estimated to be twice that of public toilets (Li et al., 2020).

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f Tap water use

Since 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 reactive 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, and the unit water consumption of lawn watering is 2L/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, and R is the release ratio of HOCl and Cl_2.

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. f is the chlorine volatilization ratio, which is 0.1 (Li et al., 2020). R is the release ratio of HOCl and Cl_2.

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. M is the proportion of insecticides and herbicides, 68% and 23% respectively. 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

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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 and Cl_2.

2.3 Spatial allocation

The estimated provincial chlorine emissions were further gridded into a 0.1° × 0.1° grid in order 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 emission 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 from other point sources with unavailable installed capacity, the emissions in each province were distributed equally 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 S11.

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). According to the proportion of the number of indoor and outdoor 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 Tables S12.

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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 process 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 7 major categories into 5 economic sectors, including power, industry, residential, agricultural, and biomass burning. 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,

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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). 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).

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}), while Shanghai has the highest emission intensity of Cl_2 (60.07 kg km^{-2}) and HOCl (419.48 kg km^{-2}). 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), while Ningxia and Shanghai are the provinces with the highest emission intensity of Cl_2 (39.01 g per people) and HOCl (109.72 g per people), respectively.

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

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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 monthly variation of anthropogenic chlorine emissions. 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. In contrast, emissions from other sectors remain relatively stable throughout the year. In October, the peak month, HCl emissions are approximately three times higher than in January, the month with the lowest emissions. For fine particulate Cl^-, the difference between the peak month (October) and the valley month (January) increases to over fivefold, primarily due to the dominant contribution from biomass burning. In contrast, the emissions of Cl_2 and HOCl show a different temporal variation pattern. 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 emission from garbage incinerators. We note that the emissions from garbage incinerators increase ~ 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. 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. The HCl produced by coal combustion in this study is ~2 times higher

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than their estimation, which is mainly due to the different emission factors and control technology. The HCl emission from waste incineration in this study is much lower, which is attributed the use of more detailed and lower open-air combustion rates in different provinces. The HCl emission from biomass burning is higher in this study, mainly due to the different estimation methods of household combustion rate and open combustion. We estimated lower HCl emissions than those of Zhang et al. (2022), which is mainly due to the lower estimation of coal combustion and waste incineration. They applied an upper limit of these emissions.

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 waste incineration are generally consistent across these studies. The emissions from waste incineration in Fu et al. (2018) and Zhang et al. (2022) is much higher than those in this study and Yin et al. (2022), which could be attributed to the fact that China has greatly reduced the amount of waste incineration in the open air in recent years, based on the basic principles of waste reduction, harmlessness and recycling. 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.

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). The emission from coal combustion in this study is ~2 times higher because we adopted relatively higher release ratios of chlorine. The emission from the usage of disinfectant is ~ 3 times higher because of the calculation method, the opening time of the swimming pool and the amount of chlorine added are higher than those of Yin et al.(2022). In addition, we include the emission from environmental disinfection and tap water use in this study, which account for 13% and 5% of the total emission from usages of disinfectant that can not be negligible. We also include the emission from usages of pesticide in rural areas, though its contribution to the total Cl_2 emission is small.

The research on the development of HOCl emission inventory is limited. This study and the study of 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 environmental disinfection, tap water use, and pesticide.

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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 S13 and S14. The uncertainties for HCl, fine particulate Cl^-, Cl_2, and HOCl emissions were estimated at a 95% confidence interval, resulting in ranges of -48% to 45%, -59% to 89%, -44% to 58%, and -44% to 79%, respectively (Table 6). According to the table, the uncertainties for HCl and fine particulate Cl^- in this study are comparable to those reported in other studies, while the uncertainties for Cl_2 and HOCl emissions are smaller in this study compared to the others.

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. 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 active chlorine. (3) Development of specific 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 (Yin et al., 2022). 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 measurements. This inventory includes 4 reactive 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.

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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 from June to July and 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 reactive 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 reactive chlorine species and is conducive to the development of emission control strategies to mitigate secondary pollution in

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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.

Financial support

This research has been supported by the Key-Area Research and Development Program of Guangdong Province (2020B1111360003), Guangdong Major Project of Basic and Applied Basic Research (2020B0301030004), Science and Technology Program of Guangdong Province (Science and Technology Innovation Platform Category) (2019B121201002), and the National Natural Science Foundation of China (42105097), and Guangdong Basic and Applied Basic Research Foundation (2023A1515010162).

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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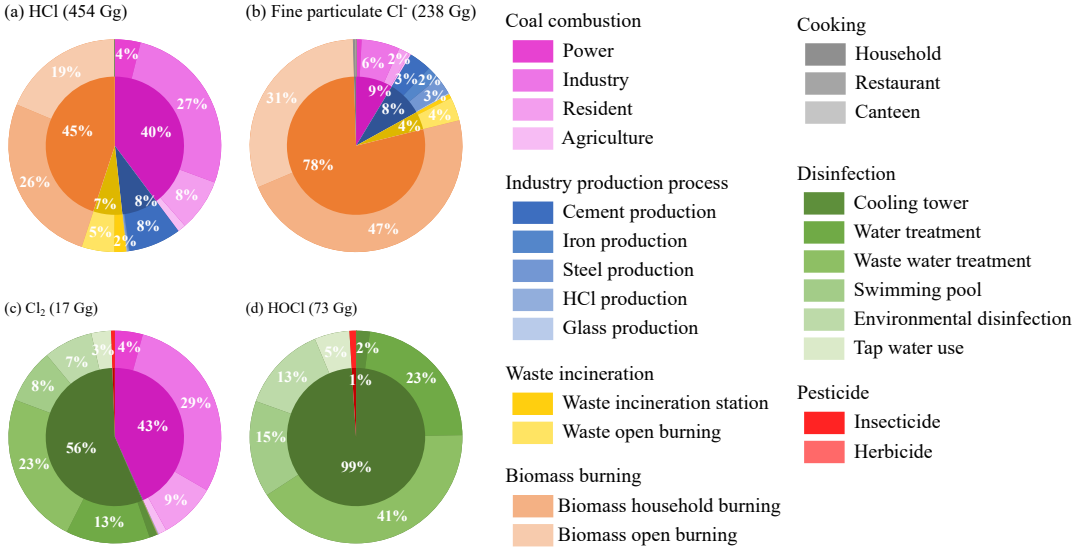

Figure 1 Contributions of different source categories to the anthropogenic emissions of HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) in 2018.

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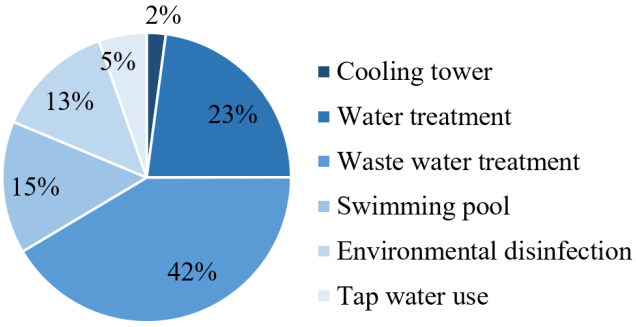

Figure 2 Proportion of different usages of chlorine-containing disinfectant in China.

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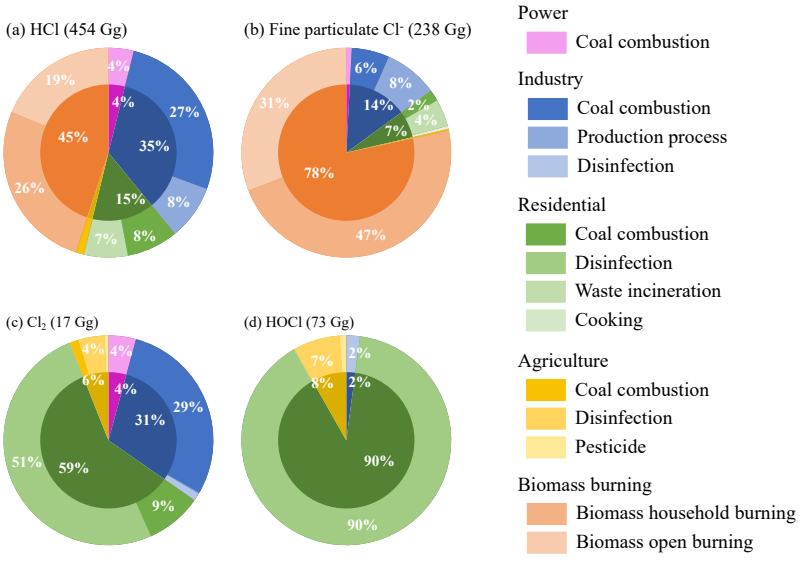

Figure 3 Contributions of different economic sectors to the anthropogenic emissions of HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) in 2018.

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

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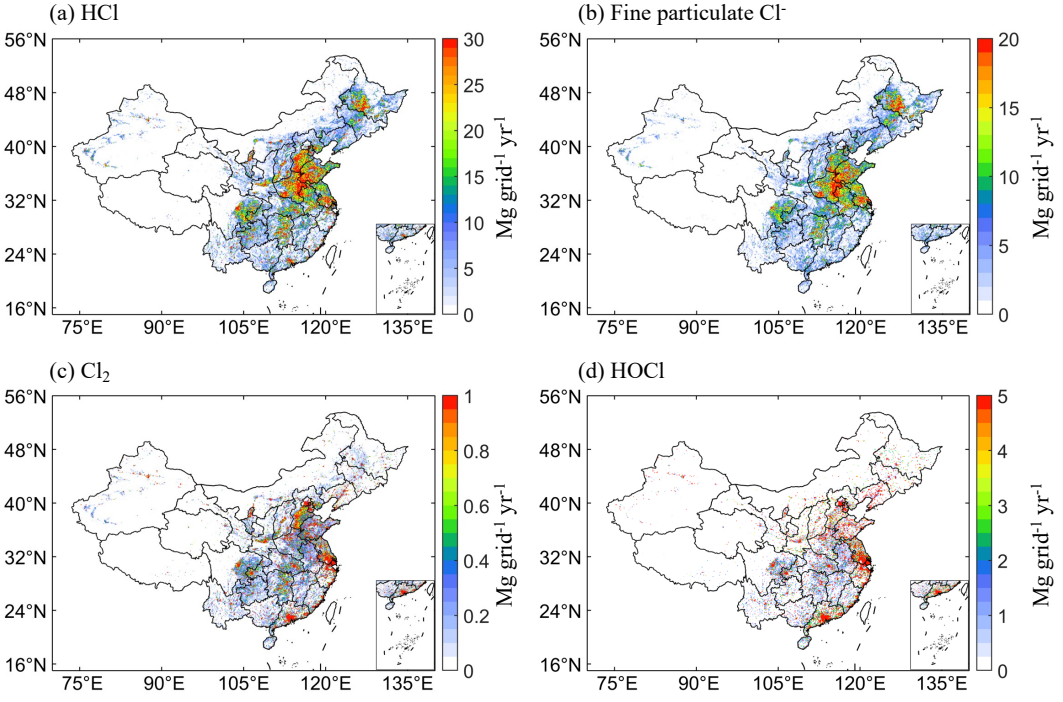

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Figure 5 Spatial distribution of anthropogenic HCl, fine particulate Cl^-, Cl_2, and HOCl emissions in 2018.

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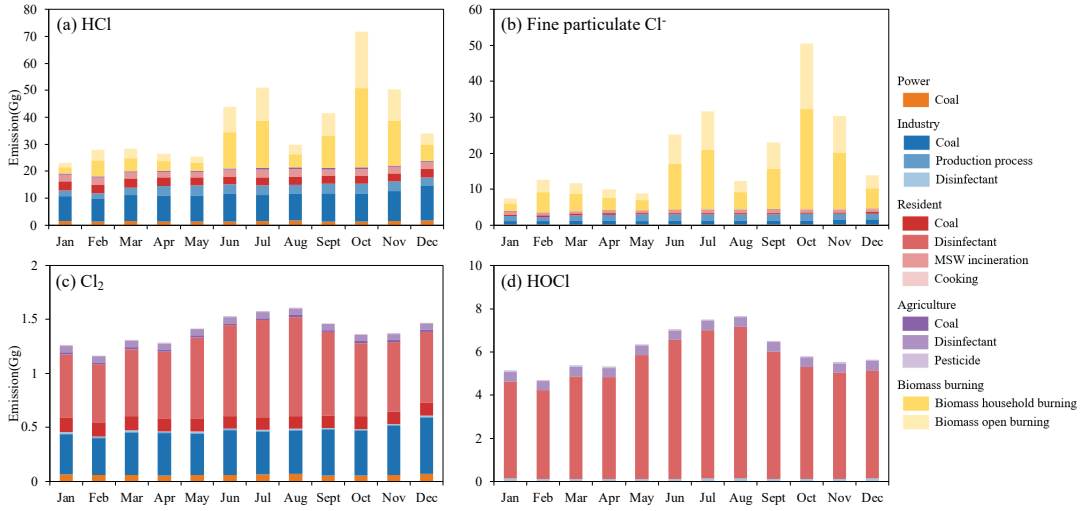

750 Figure 6 Monthly variation of anthropogenic HCl (a), fine particulate Cl^- (b), Cl_2 (c), and HOCl (d) emissions by economic sector in 2018.

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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 use
Base year	2018
Spatial resolution	$0.1^o \times 0.1^o$
Temporal resolution	Monthly

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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

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

Sector	Subsector	Emission (Gg)			
		HCl	pCl	Cl_2	HOCl
Power	Power	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

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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

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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.

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Table 6 Uncertainties of the ACEIC inventory and comparisons with other studies.

Study	Region	HCl	pCl	Cl_2	HOCl
This study	China	-48%~45%	-59%~89%	-44%~58%	-44%~79%
Yin et al. (2022)	China	-23%~56%	-42 %~74%	-44%~82%	-65%~87%
Zhang et al. (2022)	China	-22%~132%	-57%~48%	-	-
Fu et al. (2018)	China	-33%~83%	-40%~82%	-	-
Yi et al. (2021)	YRD	-27%~17%	-57%~57%	-106%~167%	-75%~95%
Li et al. (2020)	Shanghai	-	-	-85%~85%	-85%~85%