Science of the Total Environment 771 (2021) 144644

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Anthropogenic emissions of atomic chlorine precursors in the Yangtze River Delta region, China

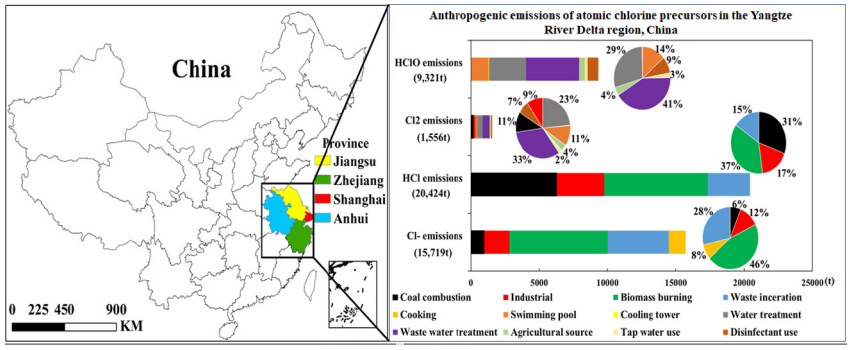
Xin Yi [a,b,c,1], Sijia Yin [a,b,1], Ling Huang [a,b], Hongli Li [a,b], Yangjun Wang [a,b], Qian Wang [a,b], Andy Chan [d], Dramane Traoré [a,b], Maggie Chel Gee Ooi [e], Yonghang Chen [c], David T. Allen [f], Li Li [a,b,*]

[a] School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China
[b] Key Laboratory of Organic Compound Pollution Control Engineering (MOE), Shanghai University, Shanghai 200444, China
[c] College of Environmental Science and Engineering, Donghua University, Shanghai 201620, China
[d] Department of Civil Engineering, University of Nottingham Malaysia, Semenyih 43500, Selangor, Malaysia
[e] Institute of Climate Change, National University of Malaysia, Bangi 43600, Selangor, Malaysia
[f] Centre for Energy and Environmental Resources, University of Texas at Austin, Austin, TX 78758, United States

HIGHLIGHTS

- Chlorine chemistry is important in the formation of secondary air pollution.
- A comprehensive emission inventory of chlorine precursors is developed for YRD based on a bottom-up methodology.
- Four chlorine precursor species and 12 source sectors are considered.
- Spatial and temporal allocations of the emissions are further discussed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 15 November 2020
Received in revised form 11 December 2020
Accepted 15 December 2020
Available online 28 January 2021

Editor: Pavlos Kassomenos

Keywords:
Chlorine
Emissions inventory
Yangtze River Delta

ABSTRACT

Chlorine radical plays an important role in the formation of ozone and secondary aerosols in the troposphere. It is hence important to develop comprehensive emissions inventory of chlorine precursors in order to enhance our understanding of the role of chlorine chemistry in ozone and secondary pollution issues. Based on a bottom-up methodology, this study presents a comprehensive emission inventory for major atomic chlorine precursors in the Yangtze River Delta (YRD) region of China for the year 2017. Four primary chlorine precursors are considered in this study: hydrogen chloride (HCl), fine particulate chloride (Cl^-) (Cl^- in $PM_{2.5}$), chlorine gas (Cl_2), and hypochlorous acid (HClO) with emissions estimated for twelve source categories. The total emissions of these four species in the YRD region are estimated to be 20,424 t, 15,719 t, 1556 and 9331 t, respectively. The emissions of HCl are substantial, with major emissions from biomass burning and coal combustion, together accounting for 68% of the total HCl emissions. Fine particulate Cl^- is mainly emitted from industrial processing, biomass burning and waste incineration. The emissions of Cl_2 and HClO are mainly associated with usage of chlorine-containing disinfectants, for example, water treatment, wastewater treatment, and swimming pools. Emissions of each chlorine precursor are spatially allocated based on the characteristics of individual source category. This study provides important basic dataset for further studies with respect to the effects of chlorine chemistry on the formation of air pollution complex in the YRD region.

* Corresponding author at: School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China.
E-mail address: Lily@shu.edu.cn (L. Li).
[1] These authors contribute equally to this work.

https://doi.org/10.1016/j.scitotenv.2020.144644
0048-9697/

1. Introduction

Atomic chlorine radical (Cl·) plays an important role in tropospheric atmospheric chemical reactions. For instance, Cl· can oxidise volatile organic compounds (VOCs) at rates of about 1 to 2 orders of magnitude faster than that of hydroxyl radicals (OH·) and this, may enhance the formation of ozone (Chang et al., 2002; Chang and Allen, 2006; Knipping and Dabdub, 2003; Tanaka et al., 2003; Wang et al., 2005, 2014b; Aschmann and Atkinson, 1995; Martin and Martin, 2010; Wang et al., 2020; Qiu et al., 2019a, 2019b), secondary organic aerosol (Wang and Ruiz, 2017; Qiu et al., 2019a, 2019b; Wang et al., 2020) and atmospheric oxidation capacity in the troposphere (Li et al., 2020a, 2020b). Field measurements in China have reported high concentrations of chlorine gas (Cl_2) (up to 1000 pptv) and nitryl chloride ($ClNO_2$) (up to 1200 pptv) in urban areas (Zhou et al., 2018; Xia et al., 2020), demonstrating that both Cl_2 and $ClNO_2$ level in China's urban areas is much higher than those observed in northern coastal cities of the United States (Spicer et al., 1998; Glasow, 2010; Liu et al., 2017; Stephen et al., 2019). Further studies reveal the existence of chlorine species that originated from anthropogenic sources in China (Tham et al., 2016; Wang and Ruiz, 2017; Yang et al., 2018). A comprehensive analysis on observed particulate chlorine (Cl^-) suggests high Cl^- loading in eastern China and the high mass ratios of Cl^-/sodium ion (Na^+) (1.75– 3.40) also confirms the existence of non-marine source of chlorine, amongst which coal combustion and biomass burning are considered as one of the main sources of Cl^- (Yang et al., 2018).

While atomic chlorine precursors are largely generated through chemistries in the atmosphere, the primary emissions are not negligible. Major primary precursor emissions including HCl, Cl_2, HClO and particulate Cl^- are emitted from both anthropogenic and natural sources (Finley and Saltzman, 2006; Khalil et al., 1999). An early chlorine related emission inventory has been developed for Cl_2/HClO over the Houston area of the United States in 2000 (Chang et al., 2001). Liu et al. (2018) developed the first anthropogenic chlorine emissions inventory for China (ACEIC) for year 2012, which includes emissions of HCl and Cl_2 from coal combustion and HCl from waste incineration plants. Hong et al. (2020) further updated the ACEIC with datasets for the year 2014 following the estimation method applied in their previous study (Liu et al., 2018). These studies are of great importance as they fill the gap between traditional emissions and chlorine related species. However, both ACEIC 2012 and 2014 only include HCl and Cl_2 emission from coal combustion and HCl emission from waste incineration stations. The emissions of HCl and particulate Cl^- from coal combustion, industrial processes, biomass burning and municipal solid waste incineration for China for year 2014 are later estimated (Fu et al., 2018), but gaseous Cl_2 and HClO are not included, while other anthropogenic sources (e.g. cooking, disinfectant usage, etc.) are not considered either. The global model study of Wang et al. (2019) emphasises the importance of natural sources of Cl but explicitly omitted HCl emissions from coal combustion in China because estimates were considered uncertain and small from a global perspective. On the contrary, a higher resolution model study by Li et al. (2020a, 2020b) supports the importance of anthropogenic Cl emissions in China and the need for improved bottom-up emission inventories. There are a number of inconsistencies in the literatures, and it further identifies the need for more complete inventories that support detailed modelling studies. An emission inventory of Cl_2/HClO, HCl, and Cl^- is developed recently for Shanghai (Li et al., 2020a, 2020b; Yi et al., 2020), the largest city in the YRD region with serious ozone (O_3) pollution as well as secondary aerosol problems (Gao et al., 2016; Shu et al., 2016). This indicates the anthropogenic emissions are significant, but as Shanghai only covers around 3% of the YRD region in area, it is thus important to develop an emission inventory of chlorine precursors in order to enhance our understanding of the O_3 and secondary pollution issues in the entire YRD region.

With these research motivations, we develop a comprehensive up-to-date anthropogenic emission inventory of major chlorine precursors, including gaseous HCl, Cl_2, HClO, and fine particulate Cl^- for the YRD region for the year 2017. Anthropogenic emissions with a total of twelve source categories are included. Emissions of each chlorine precursor are spatially allocated based on the distribution of individual source category. The temporal profiles are also given based on the activity characteristics for each source category. Comparison with other studies and uncertainties are also discussed. This study serves as an essential input dataset for further study with respect to the role of chlorine chemistry in the formation of O_3 and secondary aerosols in the YRD city-cluster.

2. Methodology

The study area is the Yangtze River Delta (YRD) region, which includes Shanghai, Jiangsu, Zhejiang and Anhui provinces, as shown in Fig. 1.

Fig. 2 lists the emission sources of different chlorine precursors considered in this study. Sources of HCl and Cl^- include the five main categories, namely: coal combustion, industrial processing, biomass burning, solid waste incineration and catering. Sources of Cl_2 and HClO include nine separate categories: coal combustion, chemical industry, cooling tower disinfection, water and wastewater treatment, usage of disinfectants, tap water use, and agriculture. Based on different technological processes and control measures, each source can be further divided into sub-categories. Details of sub-level sectors and associated chlorine precursors are listed in Table S1. A bottom-up method based on emission factors and activity data is applied to estimate the emissions of individual chlorine precursor from each source. The calculation method, sources of activity data and emission factors are summarized in Table 1. Details with respect to the emission calculations are provided in the Supplementary Information.

For spatial allocation of the emissions, the information on the latitude and longitude of power plants, industries, and waste incineration plants are collected and collated. Emissions from open biomass burning are allocated based on the locations provided in the National Center for Atmospheric Research (NCAR) - Fire Inventory from NCAR (FINN) fire spots. Emissions from other source sectors are mainly allocated based on population distribution of each city in the YRD region. For temporal allocation, we collect hourly activity intensity coefficients of various sources sectors (Mao et al., 2017; Zhang et al., 2015). In this study, the emission factor method is applied to calculate the emissions of the four chlorine precursors. The main calculation formulas are as follows:

a. HCl: The emissions of HCl are calculated based on the following equation,

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

where $A_{i,j}$ refers to the activity data, including the amount of coal combusted, industrial products, municipal solid waste incineration, straw combusted, etc.; $EF_{(HCl)i,j}$ represents the emission factors of HCl from different sources. i and j represents the county and subsectors respectively.

b. fine particulate Cl^-: The calculation method of $PM_{2.5}$ emissions has been established in details in previous studies (Zhao et al., 2013; Wang et al., 2014a, 2014b; Ma et al., 2017). The emissions of Cl^- are calculated based on the content of Cl^- in $PM_{2.5}$,

$$E_{Cl^-} = \sum_{i,j} A_{i,j} \times EF_{(PM_{2.5})j} \times M_j \quad (2)$$

where $A_{i,j}$ refers to activity data for $PM_{2.5}$ emissions, $EF_{(PM2.5)j}$ represents the emission factors of $PM_{2.5}$ from different sources; M is the fraction of Cl^- in primary $PM_{2.5}$ emissions. i and j represents the county and subsectors as above. The proportion of Cl^- in $PM_{2.5}$ adopted in this study is listed in Table S2.

The emission factors for Cl^- from biomass burning in China have been reported. Cl^- emissions from biomass burning are directly derived

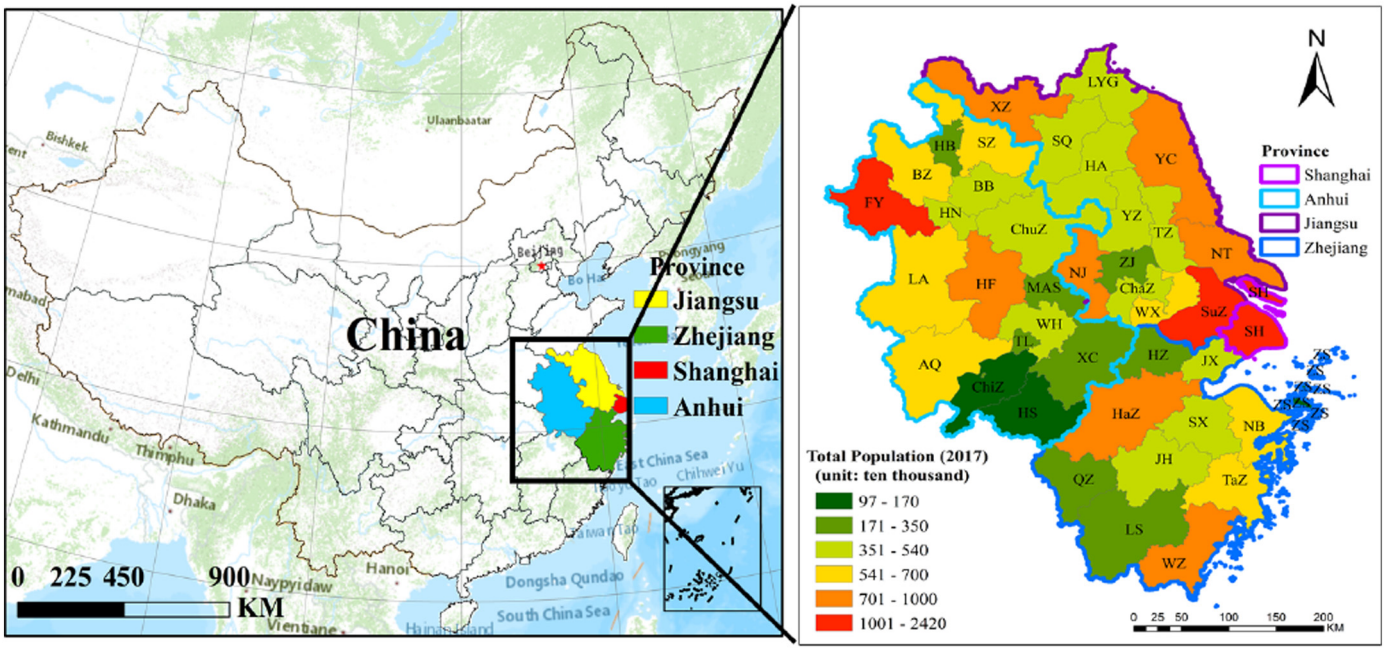


Fig. 1. Location of the Yangtze River Delta region and city-level population distribution.

from the activity level multiplied by the Cl^- emission factor. The emission factors of Cl^- from biomass burning adopted in this study are shown in Table S3.

c. Cl_2/HClO: The calculation methods of Cl_2 and HClO of different sources are slightly different. The main calculation method used is

$$E_{Cl_2/HClO} = \sum_i A_i \times (C_{ai} - C_{ri}) \times v \quad (3)$$

where $E_{Cl2/HClO}$ is the emission of Cl_2/HClO; A_i is the activity data; C_{ai} is the concentration of chlorine added in water, C_{ri} is the concentration of residual chlorine, v is the volatilisation rate. i represents the source type.

The detailed calculation methods for various sources including coal combustion, chemical industry, water and waste water treatment, use of biocides in cooling towers, disinfection of swimming pools, tap water and usage of chlorine-containing disinfectants have been reported in our previous study (Li et al., 2020a, 2020b). In addition, we also include emissions from agriculture in this article. There are many photochemical reactive chlorine-containing VOCs (e.g., fumigants and pesticides) in agricultural applications, which are especially important during the crop growing seasons. These chemicals can be precursors for chlorine radicals and may play a role in ozone or secondary organic aerosol (SOA) formation in agricultural regions. Zhang (2016) calculates that organic phosphorus, organic nitrogen, and organic sulphur pesticides accounted for approximately 70% of pesticide application in Guangdong, Jiangxi, and Hebei provinces. It is estimated that the use of organochlorine pesticides accounts for 30% of the total pesticides. The proportion of pesticides used is greater than 68%, and herbicides account for around 23% (Zhang, 2016). Information about several chlorine-containing pesticides and herbicides are shown in Table S4. The formula for calculating Cl_2/HClO emissions is shown in Eq. (4).

$$E_i = \sum_i \frac{T_i}{\rho} \times A_i \times C_i \times v, \quad (4)$$

where T_i is the amount of chlorine-containing pesticide used; ρ is the density of the insecticide; A_i is the effective component of chlorine; C_i is the proportion of molecular chlorine, v is volatile chlorine, and the value in this study is 30%, i represents the type of pesticide. The chlorine-containing herbicides are mainly liquid and have no density value, which do not need to be used ρ value in calculation.

3. Results and discussions

3.1. Estimated emissions of chlorine precursors

Table 2 shows the estimated emissions of chlorine precursors by source sector. The total emissions of HCl, Cl^-, Cl_2 and HClO in the YRD region in 2017 are estimated to be 20,424t (t), 15,719 t, 1556 t and 9331 t, respectively. The sectoral contributions from different emission category are presented in Fig. 3.

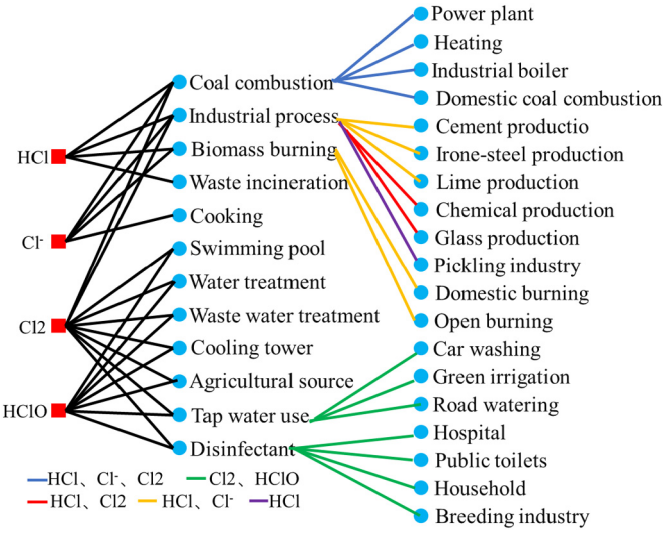


Fig. 2. Sources of chlorine precursors.

Table 1
Methods and data source applied to estimate chlorine emissions: Details of the activity data are listed in the Supporting Information.

Species	Source sector	Activity data	Emission factors
HCl	Coal combustion	The amount of coal burning, product output and waste incineration	Supplementary Eq. (19) (coal-fired)
	Industrial process		Table S8 (steel, cement, lime, chemical industry, biomass burning, garbage incineration)
	Waste incineration		
	Biomass burning-household-crop	Straw consumption	
	Biomass burning-household-firewood	Firewood consumption	
	Biomass burning-open	–	–
Cl^-	Coal combustion	The amount of coal burning, product output and waste incineration	Table S2 (the proportion Cl^- in $PM_{2.5}$)
	Industrial process		
	Waste incineration		
	Biomass burning	Biomass burning	Table S3 (biomass burning)
	Cooking	Number of households and catering companies	Wu et al., 2018
Cl_2	Coal combustion	Coal burning and exhaust gas production	Deng et al., 2014
	Industrial process		GB 31573-2015, GB 37823—2019
Cl_2/HClO	Swimming pool	Number of swimming pools	Chang et al., 2001
	Water treatment	Quantity of water and wastewater treatment	
	Waste water treatment		
	Cooling tower	Water supply of cooling tower	
	Agriculture source	Chlorine disinfectant usage	
	Tap water use-car washing	Car wash water consumption	
	Tap water use	Green irrigation and road watering	
	Disinfectant	Disinfectant usage in hospital	
		Public toilets and household	
		Breeding industry	

Major sources of HCl emissions include coal combustion (6283 t), industrial processing (3490 t), biomass burning (7543 t), and waste incineration (3108 t). Biomass burning represents the largest source of HCl emissions, accounting for 37% of total emissions, followed by coal combustion, industrial processing. The emissions of Cl^- are mainly from coal combustion (964 t), industrial process (1831 t), biomass burning (7222 t), waste incineration (4479 t) and cooking (1223 t). The largest anthropogenic source of Cl^- emissions is biomass burning, accounting for 46%.

Emissions of Cl_2 and HClO are mainly related to the usage of chlorine-containing disinfectants in water treatment (364 and 2746 t), wastewater treatment (510 and 3847 t) and swimming pools (170 and 1284 t). Cl_2 and HClO emissions from other sources including industrial processing, cooling towers tap water use, agricultural sources and disinfectant use are relatively small, each accounting for less than 10% of the total emissions.

3.2. Spatial distribution of HCl, Cl^-, Cl_2 and HClO

The spatial distributions of annual total HCl, Cl^-, Cl_2 and HClO emissions are shown in Fig. 4. The chlorine precursors are mainly distributed around the central-east and northwest of the YRD region. The distributions of HCl and Cl^- are similar, with large emissions in Anhui and Jiangsu province, due to the relatively large amount of household biomass burning and the relatively developed industry. The emissions of Cl_2 and HClO are higher in the central and eastern regions of the YRD region, especially in urban areas such as Shanghai, Suzhou, Hangzhou, Nanjing, mainly due to large amount of water treatment and wastewater treatment.

Table 2
Emissions of chlorine precursors from various source sectors in the YRD in 2017.

Source category	Sub-sector	HCl emissions (t)	Cl^- emissions (t)	Cl_2 emissions (t)	HClO emissions (t)	Percentage of total emissions (%)
Coal combustion	Power plant	2504	269	115	–	6.1
	Heating	625	193	32	–	1.8
	Industrial boiler	386	120	19	–	1.1
	Disperse coal combustion	2767	382	6	–	6.7
Industrial process	Cement production	1424	240	–	–	3.5
	Iron-steel production	204	1411	–	–	3.4
	Lime production	250	180	–	–	0.9
	Chemical production	811	–	137	–	2
	Glass production	211	–	10	–	0.5
	Pickling industry	591	–	–	–	1.3
Biomass burning	Domestic burning	6128	6117	–	–	26
	Open burning	1415	1105	–	–	5.4
Waste incineration	–	3108	4479	–	–	16.1
Cooking	–	–	1223	–	–	2.6
Swimming pool	–	–	–	170	1284	6.6
Water treatment	–	–	–	364	2746	9.3
Waste water treatment	–	–	–	510	3847	0.1
Cooling tower	–	–	–	3	26	1
Agricultural source	–	–	–	54	405	0.6
Tap water use	–	–	–	31	235	1.9
Disinfectant	–	–	–	105	788	6.1
Total		20,424	15,719	1556	9331	100

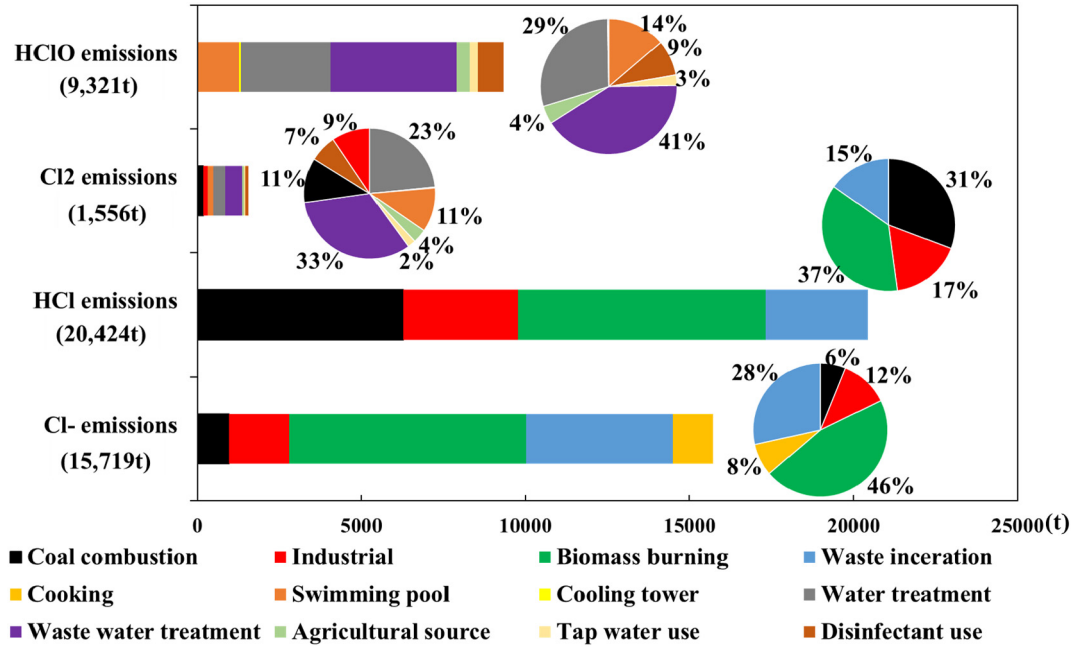


Fig. 3. Sectoral contributions of HCl, Cl^-, Cl_2 and HClO emissions in the YRD in 2017.

The spatial distributions of chlorine precursor emissions from different source sectors are shown in Fig. 5–6 and the Supporting Information. We obtain the specific latitudes and longitudes of coal-fired power plants, industrial coal-fired plants, steel plants, cement plants, garbage incineration plants and other enterprises from the 2018 environmental statistics. The domestic coal and household biomass burning according to rural population data, and cooking sources are allocated based on the respective commercial catering companies. Water treatment and wastewater treatment are allocated according to population, and is processed using SMOKE 3.7(Sparse Matrix Operator Kernel Emissions) for processing with a grid of 4 km × 4 km to obtain the result as shown in the Fig. 5. Coal combustion, industrial processing, waste incineration, water treatment and waste water treatment produce larger emissions in the central and eastern YRD region. Chlorine precursors produced by household biomass burning are mainly distributed in Anhui province, while large amounts of chlorine precursor emissions are produced by the open biomass burning in Anhui province. Cl_2 and HClO emissions in Shanghai, Suzhou, Nanjing, and Hangzhou are relatively high, with annual emissions exceeding 36 t and 350 t, respectively. This is mainly due to the large amount of water use, water and waste water treatment which is related to the population density. Huaibei, Chizhou, Huangshan, Xuancheng and Tongling (Anhui) have less Cl_2 and HClO emissions, with annual emissions lower than 6 t and 45 t respectively.

3.3. Temporal distribution of HCl, Cl^-, Cl_2 and HClO

The temporal profiles of different chlorine precursor emission sources are shown in Fig. 6. It is obvious that the emissions from 9:00 am to 12:00 am and from 17:00 pm to 19:00 pm are relatively high. Catering, household biomass burning, dispersed coal burning, industrial coal combustion and industrial processing have high contributions during this time period. From 22:00 pm to 5:00 am, the emission of chlorine precursors is relatively small, and the emissions from cooking sources, swimming pools, and water treatment sources are relatively small, which are closely related to people's daily activity. The opening hours of swimming pools are mainly distributed from July to September, which is obviously different from the emissions from other sources. The monthly changes in emissions from other sources have little variation.

3.4. Uncertainty analysis

We estimate the uncertainty of the chlorine emission inventory based on Monte Carlo methods (Zhao et al., 2011; Zheng et al., 2009). We obtain the pertinent activity level data and the uncertainty distribution of emission factors from literatures (Zhou et al., 2017; Zhao et al., 2011) (Table S9). The uncertainty of HCl, Cl^-, Cl_2 and HClO emissions in YRD in 2017 is shown in Fig. 7, with uncertainties range of −26.8 to 16.8%, −57.4% to 56.8%, −105.7% to 166.5%, −75.1% to 94.5% at 95% confidence intervals, respectively.

The uncertainty of HCl and Cl^- mainly comes from the chlorine content in coal, the removal efficiency of HCl by coal-fired facilities and different coal-fired control measures, and the data of household biomass combustion activity has greater uncertainty. The uncertainty of Cl_2 and HClO mainly comes from the use of chlorinated pesticides in agricultural sources and the volatilisation factor of chlorinated substances, and the lack of detection values for the amount of chlorine and residual chlorine in the water and wastewater treatment processes. The amount of water treatment and sewage treatment also has large uncertainty.

In order to reduce the uncertainty of the inventory, more emission measrurement data are needed, for example, the emission factors of HCl and Cl^- in industries such as steel, chemical and cement production, and the removal efficiency of HCl and Cl^- by different coal combustion control measures. Increasing the investigation of the use of chlorine-containing disinfection in the water treatment and wastewater treatment process, and obtain more information on the amount of chlorination and residual chlorine in the water treatment process. The proportion of Cl_2 and HClO volatilised in daily life applications are needed through experimental testing. In order to further verify the emission inventory, we recommend that chlorine precursors be observed in more locations and in different seasons, and the observed data is compared with the simulated data. Despite the uncertainty, the current inventory provides the latest and more comprehensive estimates of chlorine precursor emissions in the YRD region through

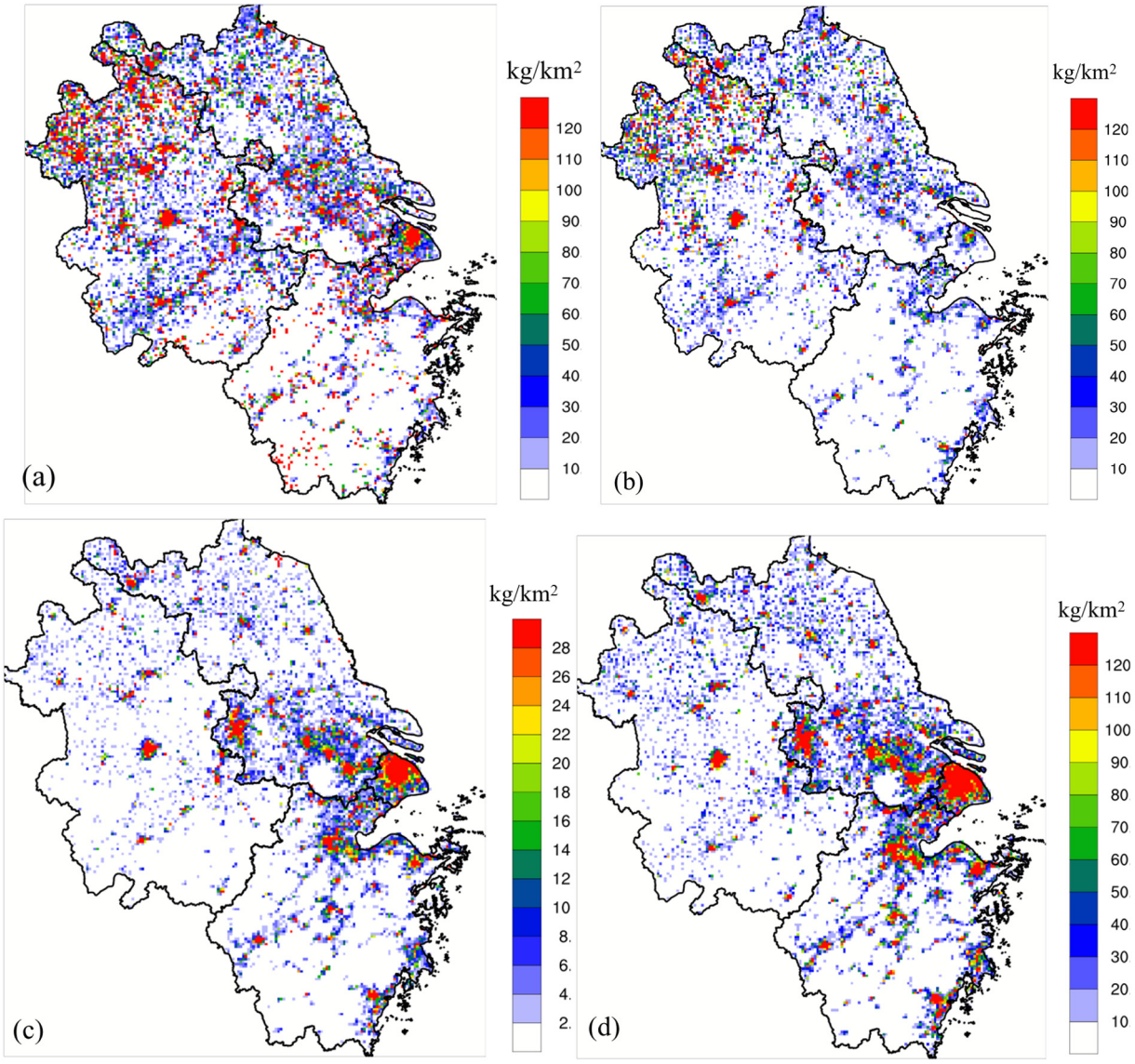


Fig. 4. Spatial distribution of (a)HCl, (b)Cl^-, (c)Cl_2 and (d)HClO emissions in the YRD in 2017.

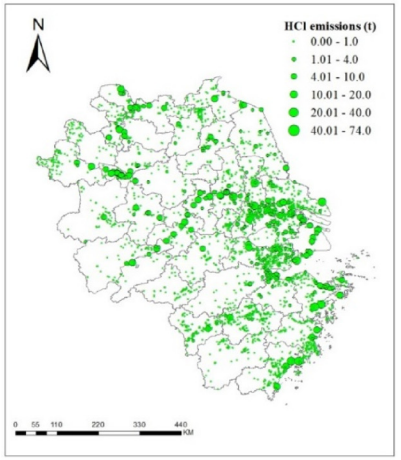
detailed local data, so as to better understand the impact of chlorine chemistry on the generation of air pollution in the YRD region.

3.5. Comparisons with other studies

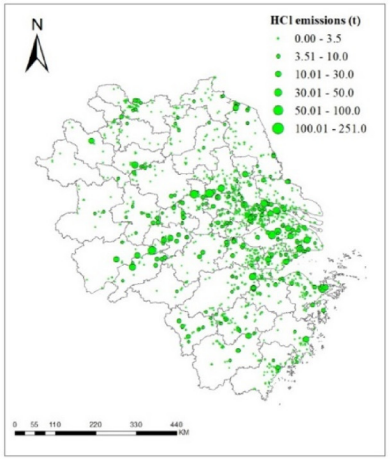
Table 3 shows comparisons between our results and previously reported data. The total emission intensity (emissions per unit area) over the YRD region is approximately one tenth of that estimated in Southeast Texas and 1.2 times higher than Beijing. The result of Cl_2 emissions estimated in our study is far lower than the emissions in Southeastern Texas. The main reason is that the use of chlorine-containing disinfectants in cooling towers in Southeastern Texas is more common, with a value of 6 t/day. There are few reports and studies on the use of chlorine-containing disinfectants in cooling towers in China. We use the industrial water consumption in the statistical yearbook and calculate the volatilisation of Cl_2 and HClO based on the circulating water volume of the cooling tower. The chlorine emissions from cooling towers in YRD (29 t) is much lower than that in Southeast Texas (2190 t).

We apply the same methods employed previously (Fu et al., 2018; Qiu et al., 2019a, 2019b) to estimate HCl and Cl^- emissions. However, both studies do not consider the emissions of chlorine precursors in the pickling, glass, and chemical industries. The industrial source of HCl in this study is more comprehensive. Compared with these studies (Fu et al., 2018) our results show a large contribution of HCl emissions from industrial sources while waste incineration only contributes a small proportion. The emissions calculated in this study are significantly different from those estimated due to the expansion of source categories and the different base year selected (Fu et al., 2018).

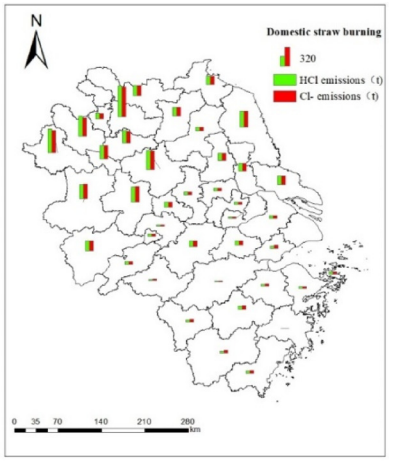
The HCl and Cl_2 emissions from coal combustion and waste incineration sources in China are estimated in a previous study (Liu et al., 2018), with emissions from coal combustion processes estimated at 232,900 t and 9400 t, respectively for the year of 2014. In this study, the emissions of HCl and Cl_2 from coal combustion in the YRD region are 6283 t and 172 t, respectively. The emissions of HCl from coal combustion are about 36 times higher than that of Cl_2, and our results are higher than the results reported earlier, which is 25 (Liu et al., 2018). The most likely reason is that the annual coal consumption for the year 2012 calculated is higher than the coal consumption adopted in our study (year 2017), and the efficiency of related flue gas treatment facilities has also improved thus the emissions of HCl and Cl_2 are lower in our study. This comparative results also indicate that the



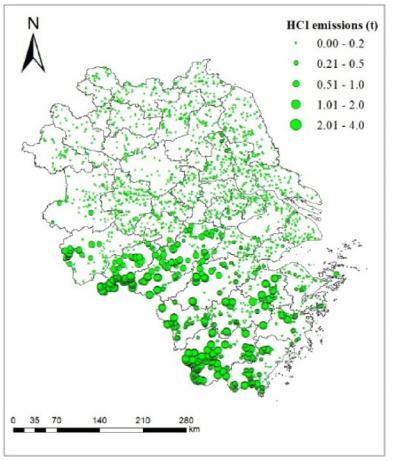
(a) HCl from coal combustion



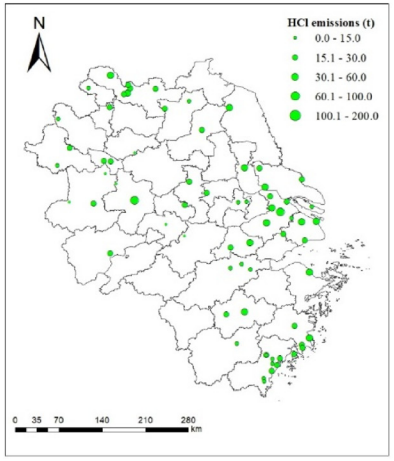
(b) HCl from industrial processing



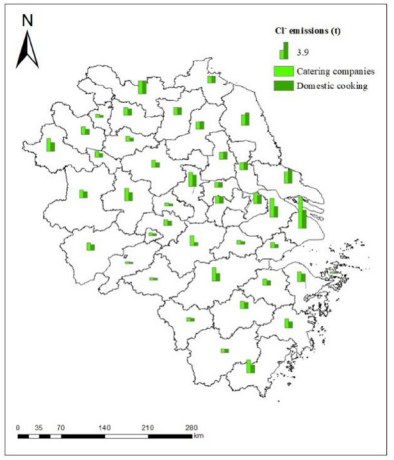
(c)HCl from household biomass burning



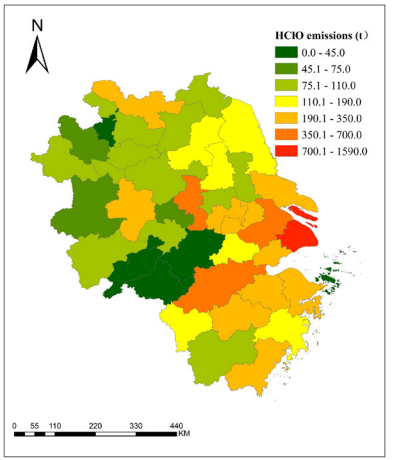
(d) HCl from open biomass burning



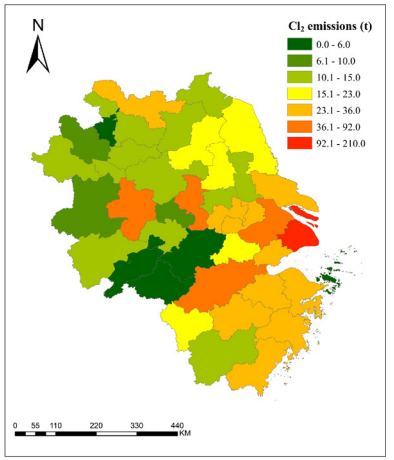
(e) HCl from waste incineration



(f) Cl^- emissions from cooking



(g)Area source of HClO emissions



(h) Area source of Cl_2 emissions

Fig. 5. Spatial distribution of HCl, Cl_2, Cl^- and HClO from different emission sources.

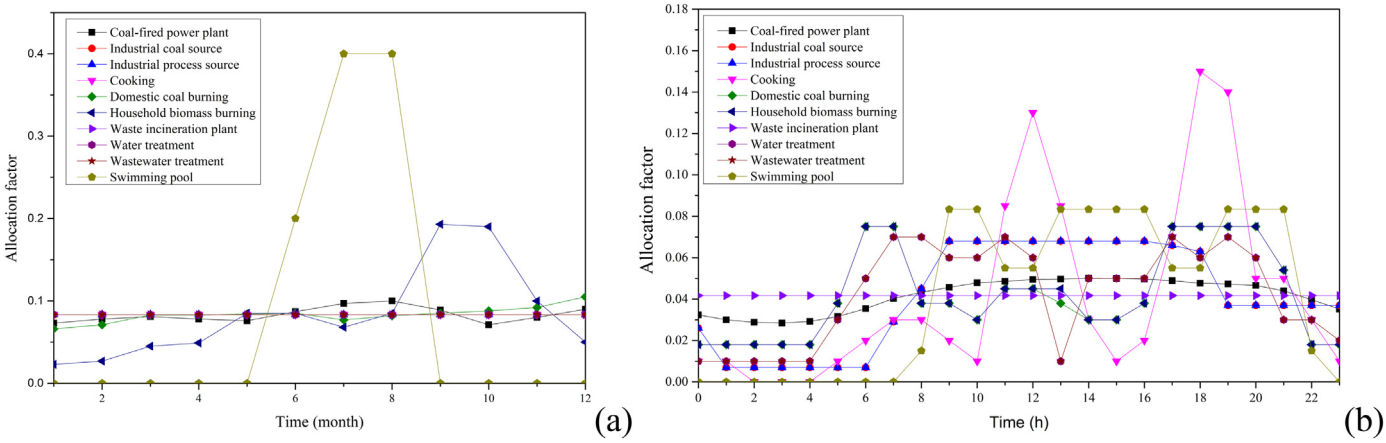


Fig. 6. Temporal distribution of different sources (a) monthly profile and (b) daily profile.

update of emissions inventory is very important due to the strict air pollution control measures conducted in China.

Cl^- emissions from commercial restaurants and households in the YRD region are also included in this study. Cl^- emissions from cooking in Beijing are estimated at 426.8 t, accounting for 75% of total Cl^- emissions (Qiu et al., 2019a, 2019b). In this study, Cl^- emissions in the YRD are estimated to be 1223 t, accounting for only 8% of anthropogenic Cl^- emissions. The large discrepancy is possibly due to the proportion of Cl^- in $PM_{2.5}$ in the cooking source in Beijing area (10%) is quite different from the factor applied in our study (1%) and the emissions from other sources may have been underestimated.

4. Conclusions

In this study, we establish a comprehensive emission inventory of chlorine precursors in the YRD region for the first time. Annual emissions of HCl, Cl^-, Cl_2 and HClO for the YRD region in the year 2017 are estimated at 20,424 t, 15,719 t, 1556 t and 9331 t, respectively. For HCl and Cl^-, biomass burning is the dominant contributor, accounting for 37% and 46% of total emission, respectively. Cl_2 emissions are primarily contributed by water and wastewater treatment, and swimming pool disinfection. Uncertainty analysis suggests that Cl_2 emissions have the largest uncertainty. Measurement of emission factors and

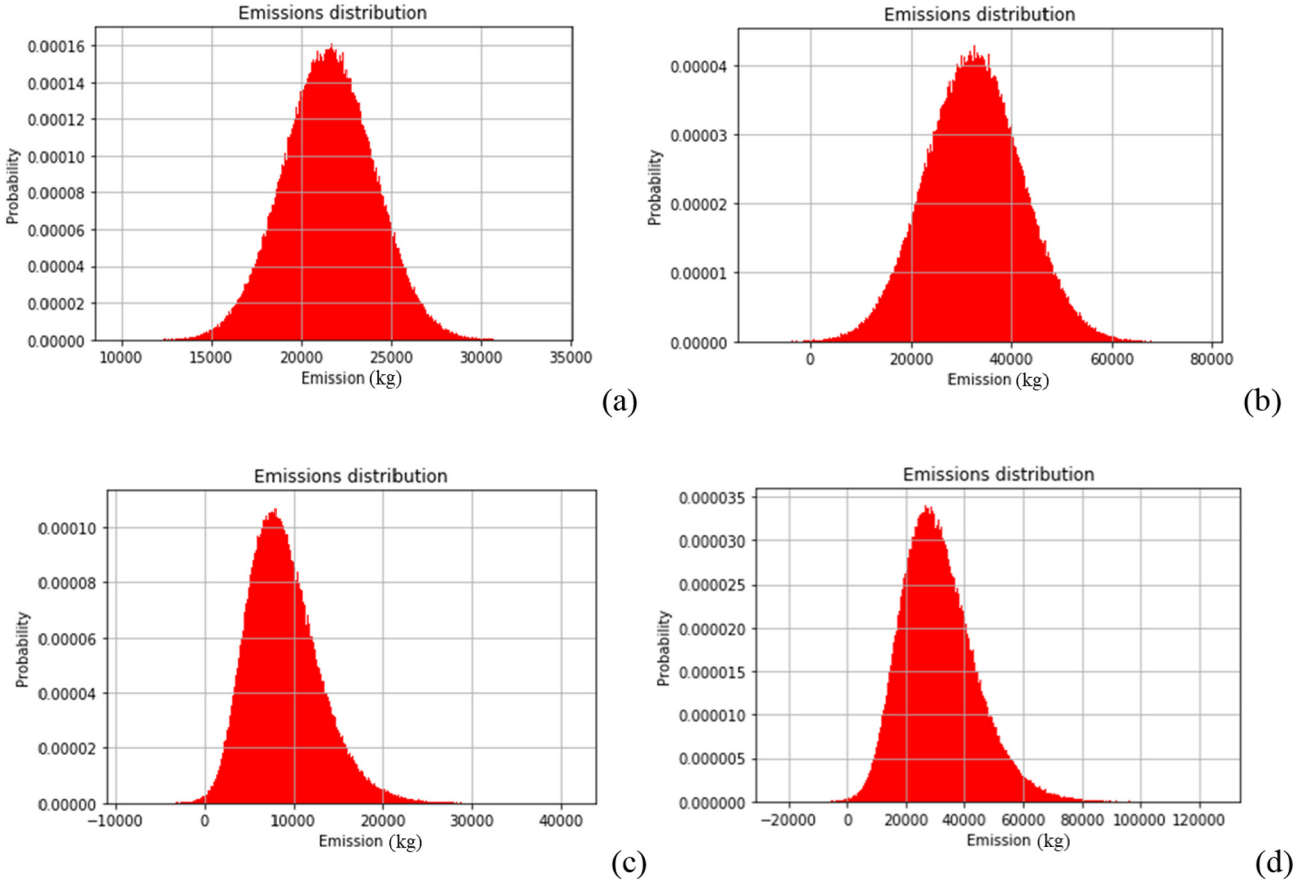


Fig. 7. Uncertainty distribution of HCl (a), Cl^- (b), Cl_2 (c) and HClO (d) emissions estimation.

Table 3
Comparisons of chlorine precursor emissions in different regions.

Region	Year	HCl emissions (t)	Cl^- emissions (t)	Cl_2 emissions (t)	HClO emissions (t)	Total chlorine emission rate by area (g/m^2)	Total chlorine emission rate by population ($g/per\ person*10^{-3}$)	Reference
Southeast Texas	2000	–	–	3453[a]	0	1.939	1.427	Chang et al., 2001
China	2012	235,800[b]	–	9400[c]	0	0.026	0.175	Liu et al., 2018
China	2014	223,400[d]	–	8900	–	0.024	0.166	Hong et al., 2020
China	2014	458,000[e]	486,000[f]	–	0	0.098	0.674	Fu et al., 2018
Beijing	2017	1893[g]	566[h]	29[i]	0	0.152	0.116	Qiu et al., 2019a, 2019b
Shanghai	2017	–	–	8650[j]	0	0.136	0.036	Li et al., 2020a, 2020b
Shanghai	2017	1207[k]	820[l]	–	0	0.32	0.083	Yi et al., 2020
YRD	2017	20,424[m]	15,719[n]	1556[o]	9331[p]	0.182	0.287	This study

[a] Water treatment: 110 t, waste water treatment: 73 t, disinfection of cooling tower: 2190 t, disinfection of swimming pools: 460 t, industrial sources: 365 t, sea salt: 110 t, tap water use: 110 t.
[b] Coal combustion: 232,900 t, waste incineration: 2900 t.
[c] Coal combustion: 9400 t.
[d] Coal combustion: 219,200 t, waste incineration: 4200 t.
[e] Coal combustion: 92,150 t (power plants, industrial boilers, domestic combustion), sources of industrial processes: 36,640 t (production of cement, steel, lime, bricks, HCl), biomass burning: 146,560 t (household, open), waste incineration: 187,780 t (incineration plant, open).
[f] Coal combustion: 24,300 t (power plants, industrial boilers, domestic combustion), sources of industrial processes: 29,160 t (production of cement, steel, lime, bricks, HCl), biomass burning: 364,500 t (household, open), waste incineration: 68,040 t (incineration plant, open).
[g] Coal combustion: 225 t (power plant, home combustion), industrial sources: 587 t, biomass burning: 0.18 t, waste incineration: 1080 t.
[h] Coal combustion: 41 t (power plant, home combustion), industrial sources: 89 t, biomass burning: 0.14 t, waste incineration: 8.5 t, cooking: 427 t.
[i] Coal combustion: 9 t (power plant, home combustion), industrial sources: 20 t.
[j] Coal combustion: 190 t (power plants, industrial, residential, other), industrial process sources: 20 t (chemical), water treatment: 850 t, wastewater treatment: 2700 t, cooling tower disinfection: 4000 t, swimming pool disinfection: 280 t, tap water use: 310 t, disinfectant use: 210 t, sea salt: 90 t.
[k] Coal combustion: 327 t (power plants, industrial boilers, domestic combustion other), sources of industrial processes: 134 t (production of cement, steel, HCl), biomass burning: 24 t (household, open), waste incineration plant: 722 t.
[l] Coal combustion: 82 t (power plants, heating, industrial boilers, domestic combustion, other), industrial process sources: 153 t (cement, steel), biomass combustion: 47 t (household, open), waste incineration: 498 t (incineration plant, open), cooking: 39 t (household, school, government agency, commercial catering), sea salt: 0.6 t.
[m] Coal combustion: 6283 t (power plants, heating, industrial boilers, household combustion), industrial process sources: 3490 t (cement, steel, lime, glass, chemical, pickling), biomass combustion: 7543 t (household, open), waste incineration: 3108 t (incineration plant, open).
[n] Coal combustion: 964 t (power plants, heating, industrial boilers, household combustion), industrial process sources: 1831 t (cement, steel, lime), biomass combustion: 7222 t (household, open), waste incineration: 4479 t (incineration plant, open), cooking: 1223 t, sea salt: 18,104 t.
[o] Coal combustion: 172 t (power plants, heating, industrial boilers, household combustion), industrial process sources: 147 t (chemical, glass), water treatment: 364 t, wastewater treatment: 510 t, cooling tower disinfection: 3 t, swimming pool disinfection: 170 t, tap water use: 31 t, disinfectant use: 104 t, agricultural sources: 54 t.
[p] Water treatment: 2746 t, wastewater treatment: 3847 t, cooling tower disinfection: 26 t, swimming pool disinfection: 1284 t, tap water use: 235 t, disinfectant use: 788 t, agricultural sources: 405 t.

atmospheric concentrations of atmospheric chlorine species are recommended in order to constrain and improve the accuracy of the inventories. Results from this study serve as a basis for further research regarding the impact of chlorine chemistry on the formation of ozone and secondary aerosols.

CRediT authorship contribution statement

X. Yi, S. J. Yin: Data collection and data analysis, Visualization, Writing.

Y. J. Wang, H. L. Li, Q. Wang: Modelling, Visualization, Investigation.

Maggie C. G. Ooi, Y. H. Chen: Data analysis,

L. Huang, A. Chan, David T. Allen, D. Traoré: Writing- Reviewing and Editing.

L. Li: Conceptualization, Methodology, Writing-Reviewing and Editing.

Declaration of competing interest

The authors declare that they have no conflict of interest.

Acknowledgement

This study is financially supported by the National Natural Science Foundation of China (NO. 42075144), Shanghai International Science and Technology Cooperation Fund (NO. 19230742500), the National Key Research and Development Program of China (NO.2018YFC0213600), the Shanghai Science and Technology Innovation Plan (NO.19DZ1205007), and the Shanghai Sail Program (No. 19YF1415600). We appreciate Dr. Dongmin Luo at California Air Resources Board for the valuable discussions and comments, which helped greatly to improve this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.144644.

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