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RESEARCH ARTICLE

Gridded usage inventories of chlordane in China

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Abstract Chlordane (1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetra-hydro-4,7-methanoindane) is one of organochlorine pesticides (OCPs) which has been listed as one of the persistent organic pollutants (POPs) to be reduced and finally eliminated in the Stockholm Convention on Persistent Organic Pollutants, because of its great persistence, toxicity, bio-accumulation and long-range transport potential. It is critical to create a national chlordane usage inventories for China to compile chlordane emission inventories, which is helpful for carrying out risk assessments and other researches related to chlordane in China. The annual data of chlordane usage was calculated and modified in accordance with the reported annual production of chlordane which was caculated on the basis of the termite distribution, the data of chlordane usage rate and the annual new construction area (NCA). With the help of Geographic Information System, the usage data of this NCA were allocated to a grid system then, with a 1/4° longitude by 1/6° latitude resolution and a size for each grid cell of approximately 25 km by 25 km. Between 1988 and 2008, the total usage of chlordane in China was 2745 t, accounting for approximately 80% of the production in the same period. Zhejiang Province was the largest consumer of chlordane in China, whose usage adds up to 980 t, greatly exceeding other provinces/regions, followed by Jiangsu Province (534 t) and Sichuan Province (428 t). The region with the least usage of chlordane was Beijing. Provinces of Guizhou, Henan and Hebei did not use any chlordane, even though termites had occurred in these provinces. Gridded usage inventories showed that the intensive use of chlordane was concentrated in the south-east part of China, Yangtze River Delta and Pearl River Delta in particular. The satisfaction of the inventories was supported by the consistence between the estimated data of annual usage and the reported annual production of chlordane, and by the consistence between distribution pattern of chlordane's usage and ambient air concentration.

Keywords chlordane, inventories, termites, China, new construction area (NCA)

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1 Introduction

The chlordane (1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetra-hydro-4,7-methanoindane) is one of organochlorine pesticides (OCPs) and a complex technical mixture of chlorinated organic compounds [1]. Since the invention of chlordane in 1945 by Julius Hyman of the Velsicol Corporation, it had been widely used on agricultural crops, lawns, and gardens including vegetables, small grains, maize, other oilseeds, potatoes, sugarcane, sugar beets, fruits, nuts, cotton and jute, and also been extensively used as a fumigating agent [2]. In the environment, chlorinated organic compounds show great persistence, toxicity, bio-accumulation and long-range transport potential. These pollutants pose a direct toxic threat to human health, acting as a poison of the central nervous system. Chronic exposures produce a variety of sublethal effects as well, most notably related to altered blood chemistry and enzymatic activity [1]. Chlordane is a known carcinogen in test animals and a suspected human carcinogen that also attacks the lungs, nervous system, and skin [3].

Chlordane has been widely used in many countries. The USA first produced chlordane commercially in 1947, and the production of chlordane reached 9.5×10^6 kg in 1974. In 1978 the United States Environmental Protection Agency (US EPA) initiated a cancellation of registration for chlordane's use on food crops. After 1983, technical chlordane was used primarily as a termiticide, and the U.S.EPA banned all the uses in 1988 [4]. Due largely to chlordane's long-term persistence, toxicity, bio-accumulation and long-range transport potential, it was listed as one of the persistent organic pollutants (POPs) to be reduced and finally eliminated in the international agreements, including the 1998 Aarhus Protocol on POPs under the 1979 Geneva Convention on Long-range Trans-boundary Air Pollution (UNECE, 1998) and Stockholm Convention on Persistent Organic Pollutants (UNEP, 2003).

Some field efficacy trials of chlordane had been conducted in the 1960s, but the chlordane production of China before 1980 were almost all exported to Africa and only a very small portion had been used domestically. The chlordane had been forbidden to use on most of the agricultural crops, such as fruit tree, vegetable tea and so on since 1982, and was completely forbidden on all the crops in 1996. So only a very small amount of chlordane had been used in agriculture in China. However, chlordane had been used to protect new constructions from termite damage since 1978 [5]. Once new buildings were constructed, chlordane was sprayed over the soil in and around the bases of new constructions [6]. In 1999, chlordane was listed as an outdated pesticide in the *Phaseout List of Outdated Production Capacity, Technics and Products (First Group)* issued by the State Economic and Trade Commission of China [7]. Since then, cypermethrin, chlorpyrifos and other insecticides have been used as substitutes for chlordane in preventing termites [8] and the demand for chlordane decreased year by year. To implement the Stockholm Convention, China banned Chlordane production for all purposes in 2009. According to official statistics, more than 95% of China's chlordane production in history had been used in fighting termites to protect new constructions, a small amount had been used for the dike, wire and cable protection, and only a very small portion had used for the sugarcane production and sericulture [8,9].

Gridded Chinese inventories have been published for some OCPs, such as technical HCH and lindane [10,11] and DDT [12]. For chlordane, however, there are not any usage inventories available on any scales (national or provincial). The goals of this paper are to quantify the usage of chlordane in China and compile gridded historical usage inventories of chlordane for this country. Using the annual new construction area (NCA) as surrogate, gridded chlordane usage inventories in China between 1988 and 2008 with a 1/4° longitude by 1/6° latitude resolution is created. It is critical and helpful for carrying out risk assessments and other researches related to chlordane in China.

2 Materials and methods

2.1 Chlordane production in China

China started to produce chlordane during the 1950s on a pilot scale and reached industrial scale during the 1970s [13]. In 2002, there were approximately nine chlordane producers in China, which were all located in the Yangze River Delta area. In 1999, chlordane was listed as an outdated pesticide in the *Phaseout List of Outdated Production Capacity, Technics and Products (First Group)* issued by the State Economic and Trade Commission of China [7]. Since then, cypermethrin, chlorpyrifos and other insecticides have been used as substitutes for chlordane in preventing termites [8], therefore, the demand for chlordane decreased year by year. To implement the Stockholm Convention, China banned chlordane production for all purposes in 2009.

The production, domestic sale and export of chlordane from 1970 to 2008 in China can be divided into five periods (Fig. 1). The first period is from 1970 to 1979 and the chlordane production of this period reached a peak of approximately 230 t in 1974. The second one spans the period from 1980 to 1987, during which the production of chlordane was halted due to its occupational harm to the workers' health and outdated technique [8]. However, the production of chlordane was resumed in 1988, and then entered into its third period (1988–1995), which is the resuming period with a total use of 400 t. The fourth one (1996–2003) is the active period, when the production of this pesticide increased fast due to China's fast economic growth. The last one (2004–2008) is the phase-out period, when the production and usage of this pesticide declined year by year. More than 95% of chlordane produced in China during Period 3 and 5 had been used in fighting termites to protect new constructions, and no chlordane had been imported or exported in these periods [8,9].

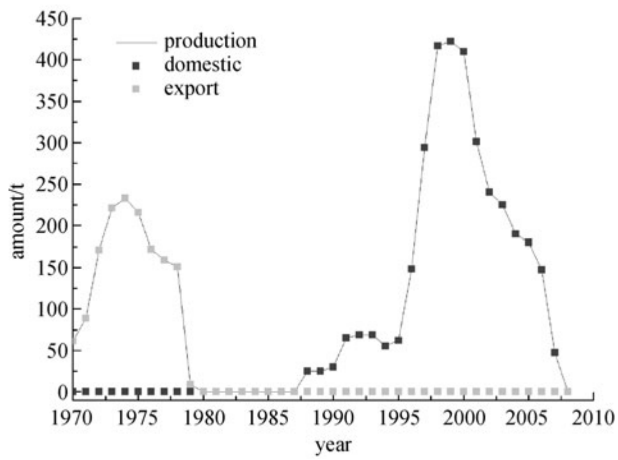

Fig. 1 Production, domestic sale and export of chlordane in China from 1970 to 2008

2.2 Chlordane usage in China

In China, actions were required to be taken for protecting new constructions from termite damage [5]. Once new buildings were constructed, chlordane was sprayed over the soil in and around the bases of new constructions [6]. To estimate the usage of chlordane in China, information of the termite distribution and chlordane usage rate were surveyed. Figure 2 shows the distribution of termites in China (shadow areas) [14]. In 2002, the National Center for Termite Control (CTC) carried out a nationwide survey on the usage of chlordane. The chlordane usage rate on new construction area (NCAs) of the surveyed cities from 1997 to 2001, which was obtained from this survey, formed the fundamental data in this inventory.

Based on the usage rate of chlordane on NCA, annual NCAs were used to estimate the annual chlordane usage in China. The data of prefectures or prefecture-level cities in China from 1988 to 2008 were collected from the National Bureau of Statistics of China (1989–2009) [15–35]. Because the chlordane rate data surveyed were given for cities, not for prefectures, so NCA surrogate data are needed to be compiled into the form of cities first.

Assuming that NCAs have direct relation with city/town population, we use city/town population of 2000 to distribute NCAs to city/town in this prefecture. The population data were collected from the "Data of 2000 Population Census at County Level" [27,36]. China's economic development is different among cities/towns of different levels, so the NCAs area cannot be simply divided in terms of the population proportion. To be more realistic, a weight factor was assigned to each. A weight factor of two was assigned to all cities at prefecture level (The cities of Beijing, Shanghai, Tianjin, and Chongqing were treated as prefecture-level cities); a weight factor of

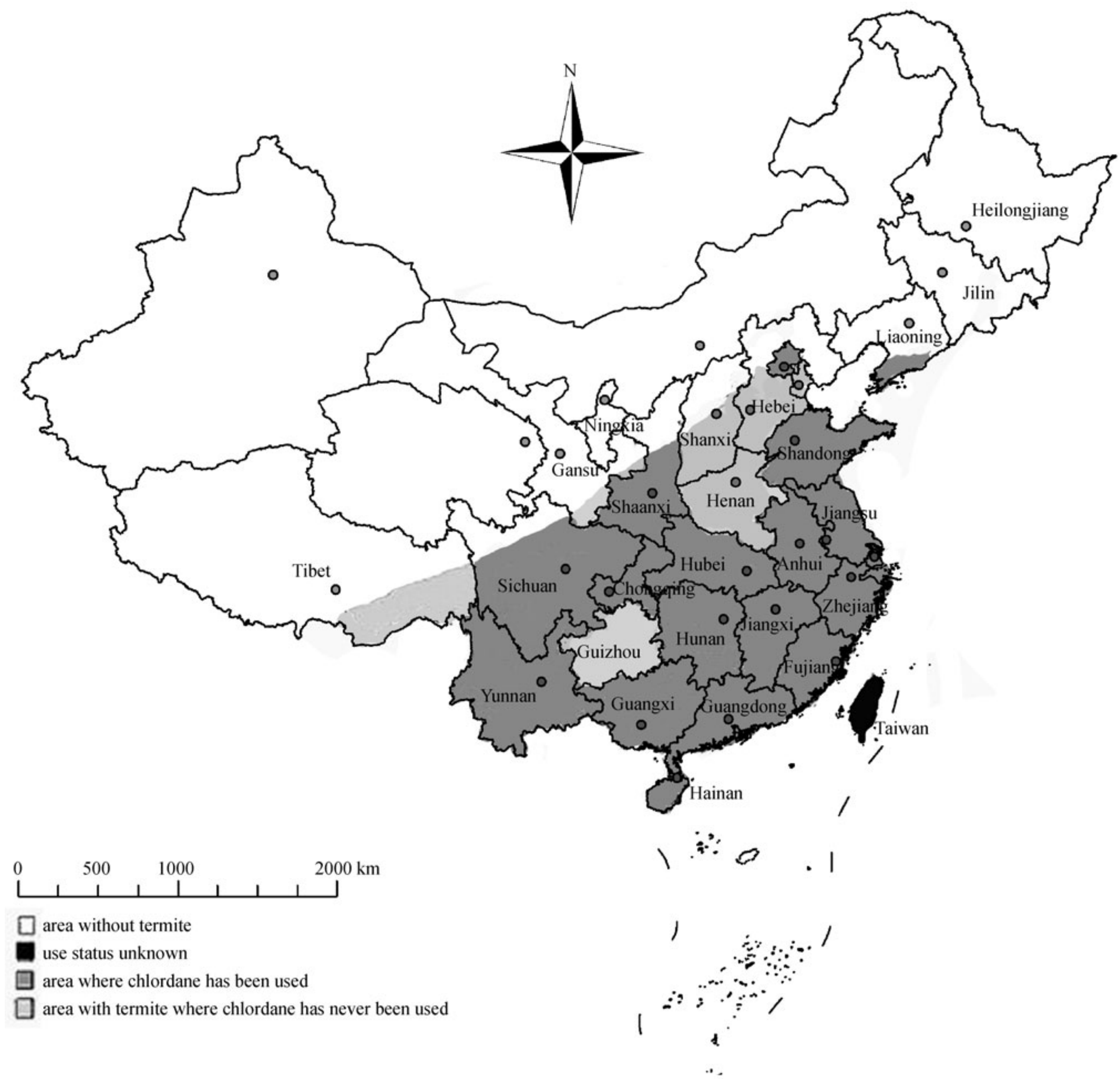

Fig. 2 Areas where termites prevail and areas where chlordane has been applied in China

1.5 was assigned to all county-level cities, and a weight factor of 1 was assigned to give all county-level towns. Thus, NCAs based on prefecture or prefecture-level cities were transferred to cities or towns according to

$$S_{ijd} = S_{ij}\frac{P_{ijd}W_{ijd}}{\sum_{d} P_{ijd}W_{ijd}}, \quad (1)$$

where, S_{ijd} is the area of new constructions in province i, prefecture j, city or town d. S_{ij} is the area of new constructions in province i, prefecture j, collected from *China Statistical Yearbook* (1988–2008) [15–35]. P_{ijd} is the town population in Province i, prefecture j, city or town d. W_{ijd} is the population weight factor assigned to this city or town.

To verify the rationality of this methodology, the distribution of population and NACs of 2000 treated with this methodology are compared (Figs. 3 and 4). China's urban population is mainly distributed in southern China, North China and South-west, which is consistent with the distribution of new housing area (NACs).

Using NCA as a surrogate, we estimate the usage of chlordane in China by assuming that chlordane are applied on newly constructed buildings. First, the average use of chlordane on NCA in prefectures and provinces is estimated by using the survey data and NCA data. Secondly, chlordane use in cities and towns is obtained by multiplying the chlordane average use rate(kg/km^2) by the NCA data in cities and towns. The details are described as below.

Step 1: The average usage of chlordane on unit NCA in the prefectures where survey data are available is calculated by

$$a_{ij} = \frac{\sum_{d} I_{ijd}}{\sum_{d} S_{ijd}}, \quad (2)$$

where a_{ij} is chlordane use rate on unit NCA in prefecture j, province i where the survey was conducted. I_{ijd} is the total surveyed chlordane usage in city/town d in prefecture j in five years (1997–2001), and S_{ijd} is the total NCA in city/town d in prefecture j for the same five years. The sum was carried out for all cities/towns d in prefecture j where the surveyed data were available.

Step 2: Similarly the average usage of chlordane on unit NCA in the province is calculated for all cities/towns in province i according to

$$a_{i} = \frac{\sum_{d} I_{id}}{\sum_{d} S_{id}}, \quad (3)$$

where a_i is the chlordane use rate on unit NCA in province i, I_{id} is the total surveyed chlordane usage in city/town d in province i in five years (1997–2001), and S_{id} is the total NCA for 1997–2001 in city/town d in province i. The sum was carried out for all cities/towns d in province i where the surveyed data are available.

Step 3: The chlordane usage in cities/towns is estimated. Chlordane usage was calculated by using Eq. (4) for the cities or towns in the prefectures where the survey was conducted, and by using Eq. (5) for the cities or towns in the prefectures where the survey was not conducted.

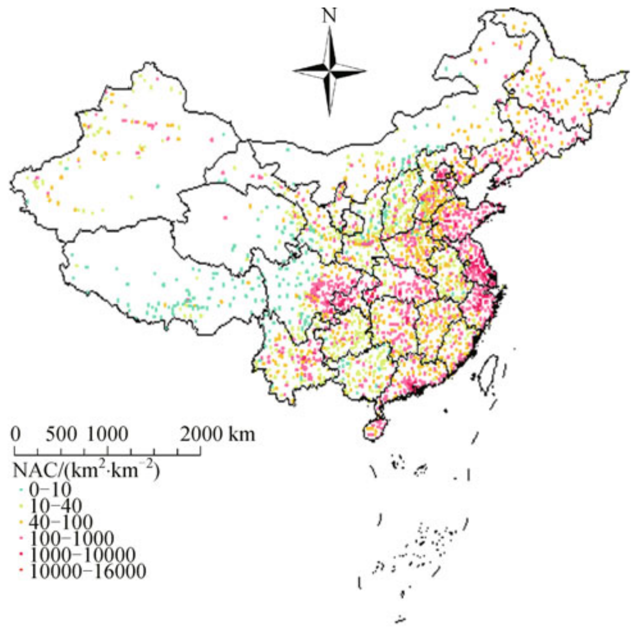

Fig. 3 Distribution of China's new construction area in 2000

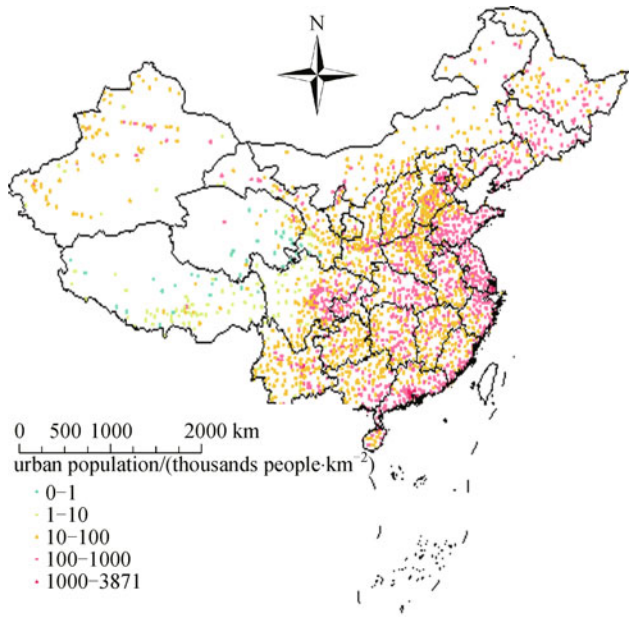

Fig. 4 Distribution of China's urban population in 2000

$$U'_{ijd} = S_{ijd} a_{ij}, \tag{4}$$

$$U'_{ike} = S_{ike} a_i, \tag{5}$$

where U'_{ijd} is chlordane usage in city/town d, prefecture j where the survey was conducted, and S_{ijd} is the NCA in city/town. Here chlordane use rate on unit NCA in prefecture j, a_{ij}, given by Eq. (2), is used. U'_{ike} is chlordane usage in city/town e, prefecture k where the survey was not conducted, and S_{ike} is the NCA in the city/town. In this calculation chlordane use rate on unit NCA in province i, a_i, given by Eq. (3), is used.

Step 4: Assuming that all the chlordane produced each year was used in the same year, the annual usage data given by Eqs. (4) and (5) for a particular year can be adjusted according to Eq. (6).

$$U_d = U'_d \frac{O}{\sum_d U'_d}. \tag{6}$$

where U_d is chlordane usage in city/town d after adjustment, U'_d is chlordane usage in city/town d, calculated from Eqs. (4) and (5). O is annual chlordane domestic sale. The sum was carried out for all cities/towns in China.

2.3 Chlordane usage gridding

It is crucial to allocate the usage of chlordane to areas where it had been actually used, which is the purpose of usage gridding. One of the most important characteristics of an inventory is its geographical area and spatial scale. Spatial data are often modeled digitally in one of two ways: as objects such as points, lines and polygons (vector format) or as a surface composed of regular grid cells or pixels with a value associated with each one (raster format). Collected data and information of chlordane usage in the previous sections are spatial data in vector format related to political units (cities/towns, counties, prefectures, or provinces), the usefulness of which is quite limited. In environmental study, pesticide usage data and other information in vector format have to be transferred to those in raster format, or on a special grid system.

Inventories of usage, emissions, and residues of different pesticides have been created on global scale with a 1° longitude by 1° latitude resolution [27] and on regional scale with a 1/4° longitude by 1/6° latitude resolution for China [10,12,37,38], the former Soviet Union [39,40], Canada [41], and the United States [11,42]. In this study, the surrogate data NCA based on prefecture (region) have been transferred to data based on city/town (point), thus it is not difficult to allocated NCA data to the grid system with a 1/4° longitude by 1/6° latitude resolution, then the use of chlordane can be allocated to the same grid system.

3 Results and discussion

3.1 Temporal chlordane use trend

Annual chlordane usage before adjustment was obtained from Eqs. (4) and (5) and is shown in Fig. 5 in comparison with the annual production which is also the annual usage after adjustment. Figure 5 indicated that our estimation of chlordane usage before adjustment was quite in agreement with the production before 1996, and lower than the production after 1996. The total usage before adjustment from 1988 to 2008 was 2745 t, accounting for approximately 80% of the production in the same period.

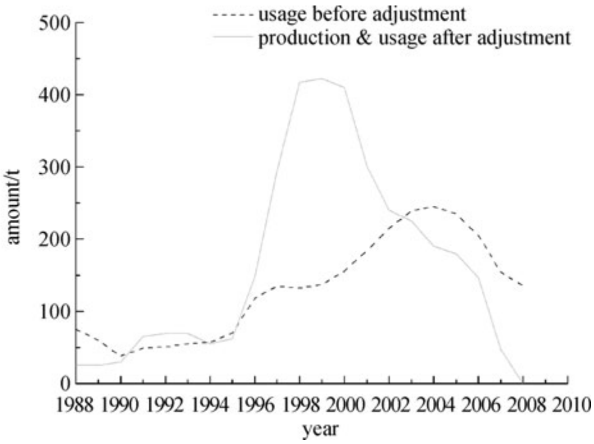

Fig. 5 Calculated annual usage and annual production of chlordane in China from 1988 to 2008

3.2 Chlordane usage based on provinces and prefectures

Figure 6 describes the accumulative usage of chlordane from 1988 to 2008 based on provinces or autonomous regions. Zhejiang Province was the largest consumer of chlordane in China, whose usage adds up to 980 t, far more than other provinces/regions, followed by Jiangsu Province (534 t) and Sichuan province (428 t). Beijing used the least chlordane. Provinces of Guizhou, Henan and Hebei had no chlordane use even though termites occurred in these provinces. On the city level, the city of Leshan in Sichuan province had the highest annual usage (about 13.4 t in 1999) and highest accumulated usage from 1988 to 2008 (around 120 t, Fig. 7).The very humid weather in the region could be the reason since this kind of weather make good living environment for termites.

Chlordane average use rates are given in Fig. 8 for the top 10 prefectures and in Fig. 9 for all provinces and autonomous regions. The prefecture with highest chlordane use rate is the city of Leshan, Sichuan Province (7.1 $kg \cdot km^{-2}$), followed by Lishui City, Yunnan Province (2.5 $kg \cdot km^{-2}$) and Zhangzhou City, Fujian Province (2.2 $kg \cdot km^{-2}$). The province with highest chlordane use rate is the Yunnan Province (0.98 $kg \cdot km^{-2}$), followed by

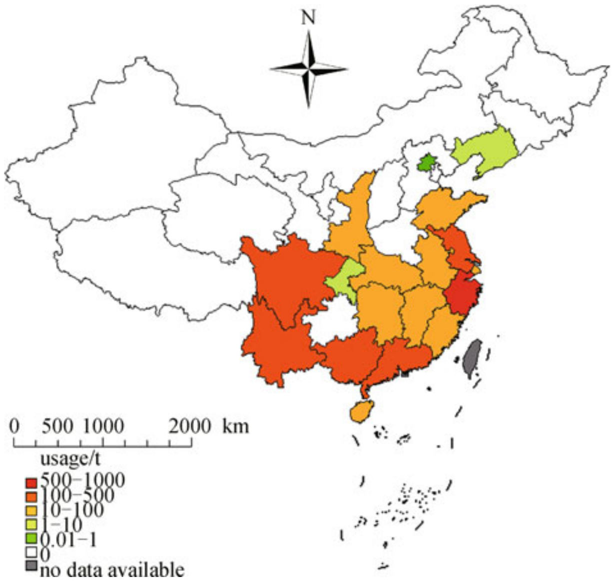

Fig. 6 Accumulative usage of chlordane from 1988 to 2008 based on provinces/autonomous regions

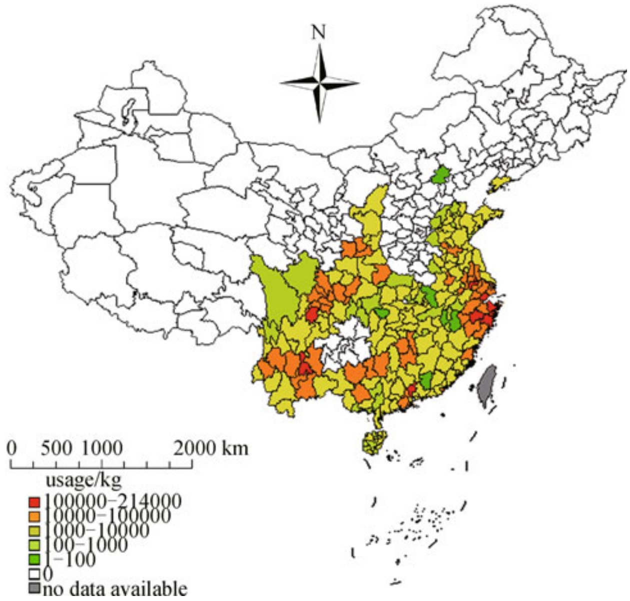

Fig. 7 Accumulative usage of chlordane from 1988 to 2008 based on prefectures

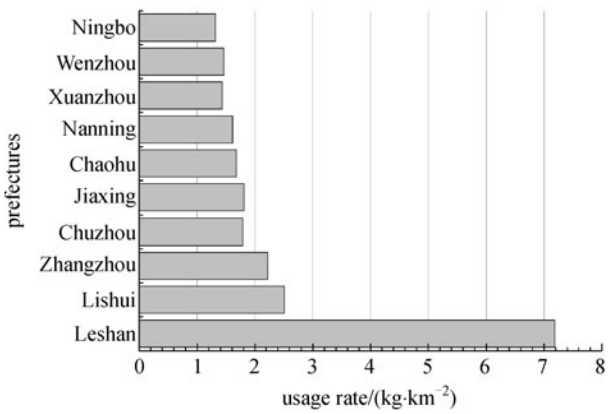

Fig. 8 Top 10 prefectures with highest chlordane use rate

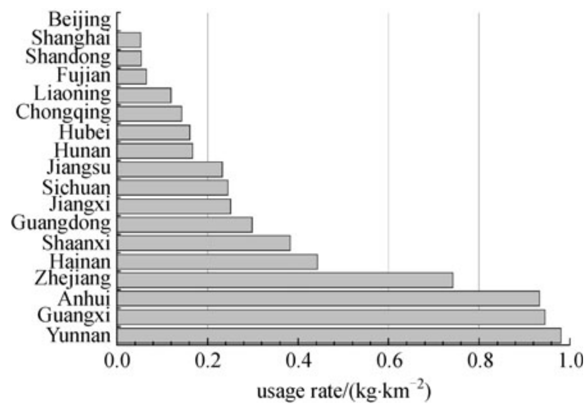

Fig. 9 Chlordane use rate for provinces and autonomous regions

Guangxi Zhuang Autonomous Region (0.94 $kg \cdot km^{-2}$) and Anhui Province (0.93 $kg \cdot km^{-2}$).

3.3 Gridded usage for chlordane

Gridded chlordane usages with a 1/4° longitude by 1/6° latitude resolution are given in Fig. 10 for 1988–2008. Chlordane usage was concentrated in the Yangze River Delta and the Pearl River Delta, which is not surprise since these regions were fast developing. The use of chlordane was also very heavy in the Sichuan Basin.

To verify the usage inventories, the ambient air concentrations of chlordane were monitored (Fig. 11). The results showed that the distribution pattern of air concentration was generally consistent with the usage pattern. The three sampling sites with highest chlordane air concentrations were in Guangzhou, Nanjing, and Fuzhou, which were all in the area with heavy chlordane use.

3.4 Uncertainty

There are two main sources of uncertainties in calculation of annual chlordane usage. First, the chlordane usage from 1988 to 2008 was estimated based on the mean usage rate of each province which was calculated from the survey information of 1997–2001, ignoring the time variation. Secondly, limited survey data were used to calculate the mean usage rate (for example, there was only one city in the provinces of Hainan and Shandong respectively where the survey was conducted), ignoring the space variation. Both of the reasons will cause uncertainty when the chlordane usage is estimated.

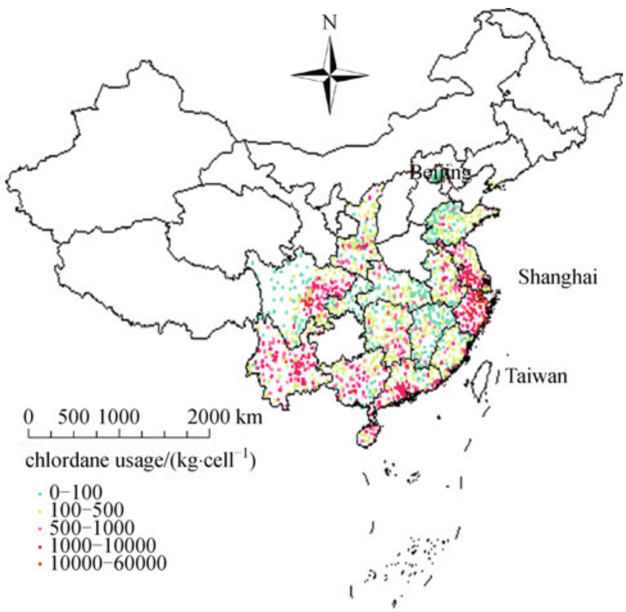

Fig. 10 Distribution of chlordane usage in China from 1988 to 2008 with 1/4° longitude by 1/6° latitude resolution

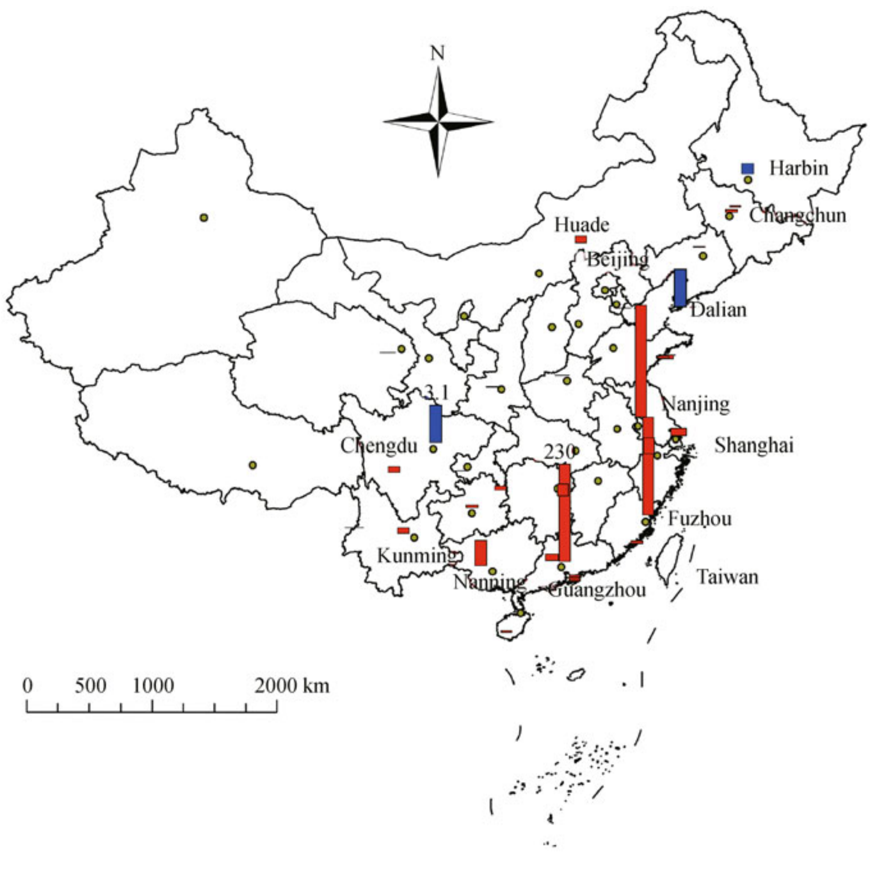

Fig. 11 Air concentration (pg/m^3) of total chlordane in China (The data for the red bars are calculated from the data by Jaward et al. (2005) [43]. The data for the blue bars are from monitoring data from Global Air Passive Sampling (GAPS) study [44]. Note the different scales for the red and blue bars.)

Although these uncertainties exist when chlordane usage is calculated, these inventories are believable due to the good quality of the information for chlordane production/usage in China. The satisfaction of the inventories is supported by the consistence between the estimation of the annual usage and the reported annual production of chlordane, and by the consistence between the spatial distribution of chlordane' usage and ambient air concentration.

4 Conclusions

The annual applications of chlordane from 1988 to 2008 in China were estimated based on the termite distribution, the survey data of use rate and the annual new construction area (NCA). It was estimated that approximately 2745 t of chlordane was used in China between 1988 and 2008, accounting for approximately 80% of the production in the same period. A comparison between our estimation and limited published usage/production data showed a good consistence. The spatial distribution of the application was generated at provincial and prefecture levels. With the help of GIS, chlordane usage of prefectures was transferred to a 1/4° longitude by 1/6° latitude gridding system.

The work described in this paper is obviously important. This is the first gridded chlordane usage inventories publicly available for China and it will pave the way for further chlordane study both in China and abroad.

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