

Gridded inventories of historical usage for selected organochlorine pesticides in Gansu Province, China

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Abstract HCHs and DDTs were banned in 1983 in China; however, they are still remaining in various environmental media. Since endosulfan was introduced in China in 1994, it is widely used in agriculture. In this study, temporal and spatial uses of endosulfan, HCHs, and DDTs in Gansu province of China have been presented. It is estimated that the total usage is 701 tons for endosulfan between 1994 and 2007, 1,712 tons for HCHs between 1952 and 1983, and 462 tons for DDTs between 1951 and 1983, respectively. Endosulfan usage increased dramatically in 1998 due to its application on other crops except on cotton. The HCH and DDT usage displayed a rapid increase after 1972, reaching the peak in 1976 and in 1975, respectively; since then, they declined until being banned in 1983. The gridded usage inventories of these three kinds of organochlorine pesticides in Gansu province, with a $1/4^\circ$ longitude by $1/6^\circ$ latitude

resolution, have been created by using different crops for endosulfan and the area of dry farmland for HCHs and DDTs as surrogate data. The most intensive use was in northwestern regions for endosulfan and southeastern regions for HCHs and DDTs in Gansu province.

Keywords OCPs · Gridded inventories · Gansu province

Introduction

Organochlorine pesticides (OCPs) are a common name of a group of pesticides consisting of benzene and chlorine. Some OCPs are persistent organic pollutants (POPs) that are semivolatile, bio-accumulative, persistent, and toxic (Vallack et al. 1998; Jones and de Voogt 1999). Most OCPs have been eliminated or restricted in their use in many countries because of their persistence in the environment and bioaccumulation in biota. OCPs are ubiquitous pollutants due to their long-range transport potential. They have been detected in air, seawater, ice, and mammals in the Arctic and Antarctic (Tanabe et al. 1983a, b; Iwata et al. 1993), although these pristine regions have never been primary sources of OCPs. Considering their harmful effects on man and wildlife, many international agreements are now coming into effect to reduce future environmental burdens. One recent international agreement is the Stockholm Convention on POPs, which will eliminate or at least control 12 POP substances, of which nine are OCPs (aldrin, endrin, dieldrin, heptachlor, chlordane, mirex, toxaphene, DDTs, and hexachlorobenzene (HCB)) (UNEP 2001). Endosulfan, an OCP, is genotoxic in mammalian cells and generally considered to be toxic and classified by the World Health Organization and the US Environmental Protection Agency as priority pollutants and a nominator for inclusion in a future iteration of the persistent organic pollutants treaty (Jia et al. 2009).

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China started to produce and use technical HCHs and DDTs in 1952 and 1951, respectively. They were widely used in agriculture and were the main pesticides. The total amount of technical HCHs produced in China was 4.5 million tons before its use was banned in 1983 (Li et al. 1998), which accounted for about 30 % of total world production (Wei et al. 2007). The total production of commercial DDTs was more than 0.43 million tons from 1951 to 1983 (Wong et al. 2002). It was noted that technical HCHs and DDTs accounted for 78 % of the total pesticide production and usage in China before they were banned in 1983 (Hua and Shan 1996). Moreover, 3,200 tons of lindane (almost pure γ -HCH) was used between 1991 and 2000. And, DDT production also continued due to export demand and dicofol production after the restriction of technical HCHs and DDTs in 1983 (Zhang et al. 2002; Qiu et al. 2004, 2005). Due to toxicity to the health of humans and wildlife, endosulfan was included in the list of POPs in 2011. At present, the production, usage, and trade of endosulfan are banned in approximately 75 countries; however, it is still used in China. It was estimated that approximately 25,700 tons of endosulfan was used in China between 1994 and 2004, with an annual average use of 2,300 tons (Jia et al. 2009).

Within international conventions, the establishment of emission inventories for POPs is mandatory and provides the basis for further emission reductions among parties (UNEP 1999, 2001; Vestreng and Klein 2002). Furthermore, emission data and release estimates are also used by modelers, which aim to understand and predict source–receptor relationships for POPs at regional (Van Jaarsveld et al. 1997; Cohen et al. 2002) and global scales (Wania et al. 1999; Wania and Daly 2002).

By establishing gridded inventories of historical usage for POPs in a region, precision usage range for POPs can be understood, which can provide data support for the establishment of multimedia migration and conversion model, source–receptor relation analysis, health risk assessment, and other researches related to POPs. It is very important for policy makers to control and reduce national or regional POPs.

At present, the studies of inventories for POPs can be divided into two categories according to the scope of study. One is based on national or regional studies of inventories, and the results reflect the overall POP usage of the study area. The other one is gridded inventories by combining with GIS technology, which can more precisely reflect usage for POPs.

For national or regional studies of inventories, by collecting the data related with HCHs, inventories of usage for α -HCH, β -HCH, and γ -HCH from 1970 to 1996 were published in Europe. Pacyna et al. (2003) established inventories of usage for DDTs, HCB, benzo- α -pyrene, and dioxin from 1970 to 1995 in European countries. For gridded inventories, the global spatial distribution of PCBs was established by allocating emissions by country to the

GEIA grid system ($1^\circ \times 1^\circ$) and using population densities within each country as a surrogate parameter (Brievik et al. 2002). The global gridded inventories for α -HCH and γ -HCH usage with a $1/4^\circ$ longitude by $1/6^\circ$ latitude resolution were published by using a global arable land density from EROS as surrogate data (Li 1999). For the studies of Chinese gridded inventories, some OCPs such as technical HCHs and lindane (Li et al. 2001), DDTs (Li et al. 1999), and endosulfan (Jia et al. 2009) were published by using a global cropland database with a $1/4^\circ$ longitude by $1/6^\circ$ latitude resolution. In addition, Zhao (2005) created inventories of historical usage for chlordane with a $1/4^\circ$ longitude by $1/6^\circ$ latitude resolution in China by using the building area of housing newly built and urban population based on prefectures as surrogate data. Dou (2006) established gridded inventories of lindane usage by allocating usage by country to a $1/4^\circ$ longitude by $1/6^\circ$ latitude gridding system and using the area of controlling wheat midge as surrogate data. Liu (2007) established inventories of historical usage for HCHs, DDTs, and lindane in the Heilongjiang River Basin of China by allocating usage by province on a $1/4^\circ$ longitude by $1/6^\circ$ latitude gridding using different gridded cropland datasets with the same resolution. Zhang et al. (2010) created gridded inventories of usage with a $1/4^\circ$ longitude by $1/6^\circ$ latitude resolution of PCBs by using the population of China as surrogate data.

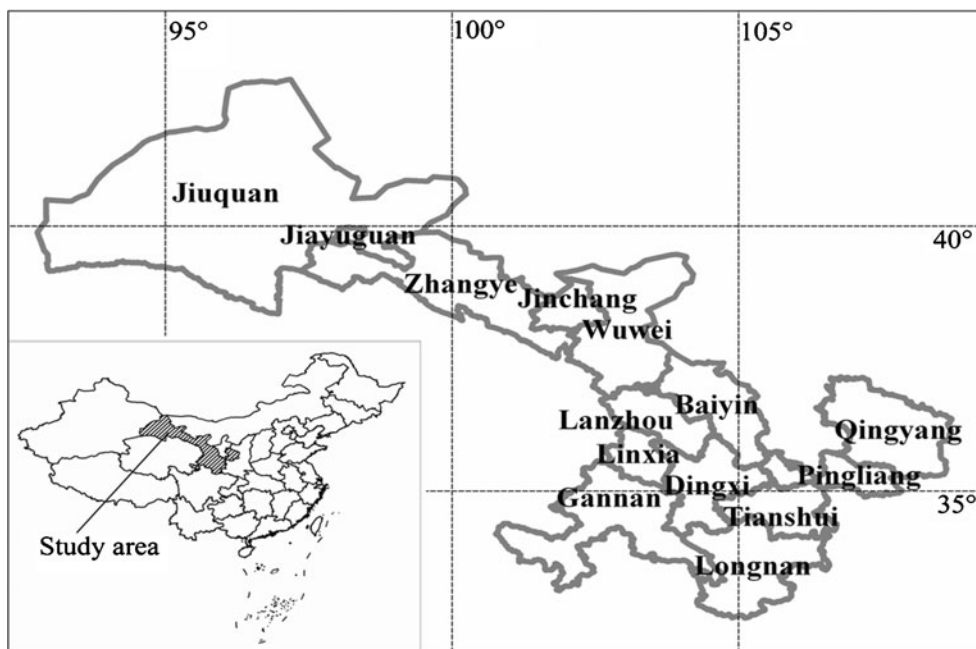
The arid and semi-arid area of northwest China accounts for 20 % of the total land area of China. With relatively higher altitude, this region also has very dry climate, and the annual precipitation decreased from the east to west. In northwest China, agriculture has been relatively well developed, such as irrigated agriculture in the Hexi Corridor area of Gansu province, one of the most important grain and cash crop production region. HCHs and DDTs were the most widely used pesticides in this area. While the inventories of OCPs have been studied extensively in northeastern, northern, and southeastern China, no extensive researches for inventories of OCPs in northwestern China were conducted. This study made an effort to establish gridded inventories of historical usage for selected OCPs, endosulfan, HCHs, and DDTs in Gansu province located in arid and semi-arid regions northwest of China (Fig. 1). The results of this work will provide data support for establishing OCP emission/residue inventories, health risk assessment, and other research related with OCPs in China.

Materials and methods

Study framework

In this study, we cited and improved the method for establishing gridded inventories of HCH usage presented

Fig. 1 The study area for inventories of selected OCPs



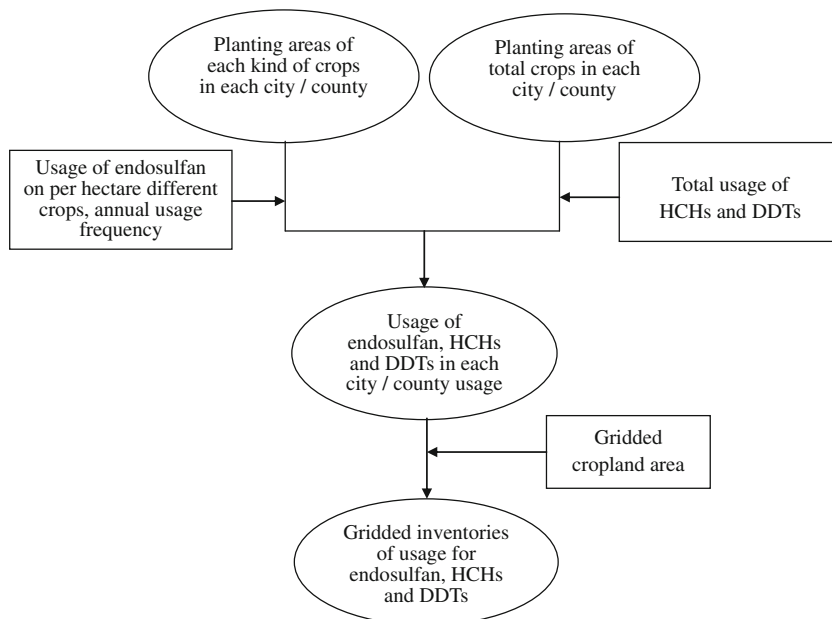
by Li et al. (1996) to establish the gridded inventories for historical usage for HCH, DDT, and endosulfan in Gansu province, China (Fig. 2). Firstly, according to the usage of endosulfan on different crops per hectare, annual usage frequency, and the planting areas of those crops, we can obtain the usage of endosulfan from each prefecture. Different with endosulfan, by using the total usage of DDTs and HCHs in Gansu province and the planting areas of the crops, we can get the usage of DDTs and HCHs from each prefecture. Then, we can establish gridded inventories of usage for endosulfan, DDTs, and HCHs in Gansu province, China, by allocating the usage of pesticides of each

prefecture into the grid.

Usage calculation

As an insecticide, endosulfan has been used only in agriculture in China. According to Chinese pesticides registered, endosulfan was used on six crops, e.g., cotton, tea, tobacco, wheat, apples, and pears (Pesticide Electronic Handbook 2006). There is almost no tea cultivation in Gansu province; thus, endosulfan usage on five crops, cotton, tobacco, wheat, apples, and pears, was estimated in this study.

Fig. 2 Framework to establish gridded inventories for endosulfan, HCHs, and DDTs



In order to estimate the use of endosulfan, the information of the average amount of endosulfan used on different crops per hectare, annual application frequencies of endosulfan on different crops, and fraction of cropland being treated by endosulfan were collected. For cotton, tobacco, wheat, and apples, the information of endosulfan usage was listed in Table 1. For pears, the annual application frequencies of endosulfan and amount used per hectare were collected from the Pesticide Electronic Handbook (2006). In addition, besides endosulfan, three other insecticides are also used on pears. By assuming that these four insecticides are equally used on pears, the fraction of cropland being treated by endosulfan is assumed to be one fourth for pears. Finally, we can collect the data of usage information of endosulfan for different crops, and these are listed in Table 1.

The usage of endosulfan can be estimated as follows:

$$U_{i,t} = U_p S_{i,t} n_i k_i \tag{1}$$

where U_{ij} is the total usage of endosulfan on crop i in year t , U_p is the amount used per hectare for different crops, S_{ij} is the area of cropland of crop i in year t , n_i is the annual application frequency of endosulfan on crop i , and k_i is the fraction of the cropland i that is treated by endosulfan.

The usage of DDT and HCH can be estimated as follows:

$$U_{i,j,t} = \frac{S_{i,j,t}}{\sum_{j=1}^n S_{i,j,t}} U_{i,t} \tag{2}$$

where $U_{i,j,t}$ is the total usage of pesticide i in prefecture j in year t , $S_{i,j,t}$ is the area of cropland treated by pesticide i in prefecture j in year t , and $U_{i,t}$ is the total usage of pesticide i in Gansu province in year t .

Usage gridding

The purpose of usage gridding is to allocate the use of pesticides to the areas where pesticides actually are used. One OCP could be used in a large amount in some areas, but little or

none in others. Accurate allocation of the use of pesticides is very important for policy makers to reduce OCPs.

In order to distribute pesticide usage database on prefecture to grid, the key is to collect the surrogate data. In this study, by using the similar method that has been applied to grid the use of pesticide (Li et al. 1999; 2000), gridded cropland datasets with 1/6° latitude by 1/4° longitude resolution for Gansu province were created to establish gridded inventories of usage for OCPs in Gansu province, China. The gridded land includes six types: gobi, desert, dry farmland, wetland, woodland, meadowland, and water area. In Gansu province, the pesticide was mainly used on cropland including a large number of dry farmland and rare wetland. Thus, in this study, gridded inventories of historical usage for selected OCPs in Gansu province are established by choosing dry farmland data from gridded land datasets with 1/6° latitude by 1/4° longitude resolution as surrogate data.

Results and discussions

Temporal use trend

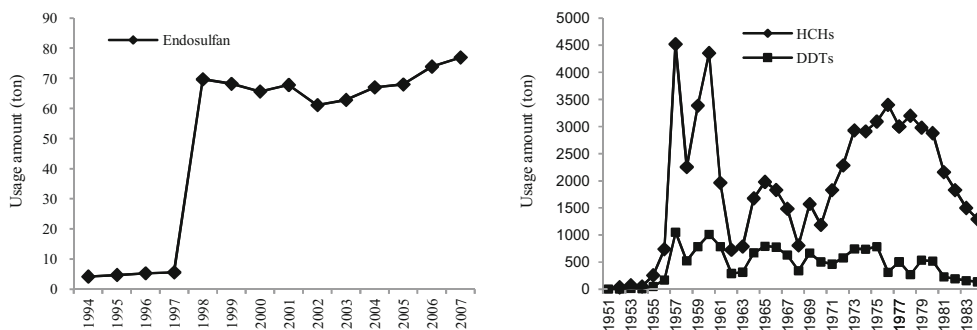
The total use of endosulfan between 1994 and 2007 is estimated at 701 tons, and annual average use is about 50 tons. Annual endosulfan usage from 1994 to 2007 in Gansu province is shown in Fig. 3, and it shows endosulfan usage dramatically increased in 1998, which is because all of the endosulfan was used on cotton from 1994 to 1998, and annual average usage is about 5 tons. From 1998, endosulfan was used on wheat, tobacco, apples, and other fruits, which resulted in a significant increase in endosulfan usage in 1998. From 1998 to 2007, the change of annual endosulfan usage displayed a relatively stable trend.

The total uses of HCHs between 1952 and 1983, and DDTs between 1951 and 1983, are estimated at 56,486 and 15,236 tons, and annual average use is about 1,712 and 462 tons, respectively. Figure 3 shows that annual HCH and DDT usage displayed three parabolic change trends

Table 1 Annual application frequencies of endosulfan, amount used per hectare of different crops, and fraction of cropland being treated by endosulfan in China (Jia et al. 2009; Pesticide Electronic Handbook 2006)

Crop	Application frequencies (n)	Amount used per hectare of different crops (g/ha)			Fraction of cropland being treated by endosulfan (k)
		Minimum	Maximum	Mean (U_p)	
Cotton	3	25	870	365.5	1/4
Wheat	1	60	120	90	1/4
Apples	3	75	438	170	1/4
Pears	4	125	438	147	1/8
Tobacco	2	87.5	525	350	1/4

Fig. 3 Annual endosulfan, HCH, and DDT, usage



(1951–1962, 1963–1968, 1969–1983). Compared with the first two stages, HCH usage increased and DDT usage decreased in the third stage. The HCHs and DDTs reached the peaks of 3,281 tons in 1976 and 758 tons in 1975, respectively, in the third stage; since then, they displayed a decline until they were banned in 1983.

The annual endosulfan usage on different crops is given in Fig. 4. The endosulfan usage on pears and tobacco has no obvious change from 1998 to 2007. The endosulfan usage on cotton increased from 1994 to 2007, especially after 2002. For apples, the endosulfan usage decreased from 1998 to 2002 and increased after 2002, which is due to the change of the apple planting area. The apple area decreased from 199.97 thousandha in 1998 to 163.53 thousandha in 2002, and since then, it increased to 247.64 thousandha in 2007 in Gansu province. For wheat, the cultivation area decreased from 1,330.16 thousand ha in 1998 to 924.14 thousandha in 2007, which resulted in the decrease of endosulfan usage on wheat from 29.62 tons in 1998 to 20.67 tons in 2007.

Figure 5 gives the total endosulfan usage on different crops. It shows that most of the endosulfan was applied to wheat and apples, which reached 283.13 and 237.24 tons between 1998 and 2007, respectively, and accounted for 67.8 % of the total endosulfan usage, followed by cotton (156.79 tons), pears (5.53 tons), and tobacco (4.49 tons).

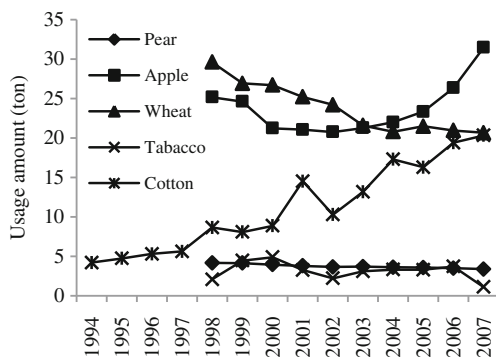


Fig. 4 Annual endosulfan usage on different crops

Usage based on prefectures

Figure 6 depicts selected OCP usage based on prefectures, and it shows that the prefecture with the highest usage of endosulfan was Dunhuang with large-scale cotton cultivation, followed by Tianshui, Jinta, Zhenyuan, and Qinan with a large number of wheat and apples. HCH and DDT usage were mainly distributed in areas with developed agriculture. HCH usage is mainly concentrated in Hunning, Dingxi, Zhenyuan, Tianshui, and Jingning, and DDT usage is mainly concentrated in Hunning, Dingxi, Jingning, Tianshui, and Zhenyuan. Table 2 listed the top ten prefectures with the highest usage of endosulfan, HCH, and DDTs, respectively. According to our estimation, total endosulfan usage of the top ten prefectures between 1994 and 2007 is 373 tons, 39 % of total usage of 79 prefectures in Gansu province. The total HCH usage between 1952 and 1983 and DDT usage between 1951 and 1983 of the top ten prefectures is 20,030 and 6,831 tons, which accounted for 31 and 35 % of the total usage of 79 prefectures in Gansu province.

Gridded usage

The selected OCP usage based on prefectures in Gansu province is the average use amount. However, one OCP could be used in a large amount in some agricultural regions and little or none in other regions, such as

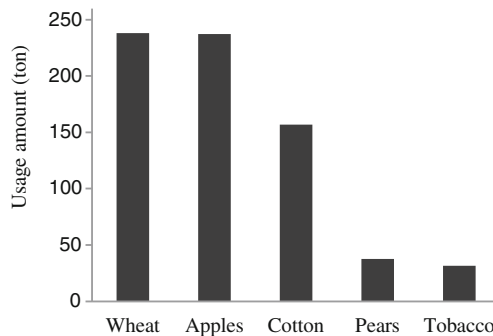


Fig. 5 Total endosulfan usage on different crops

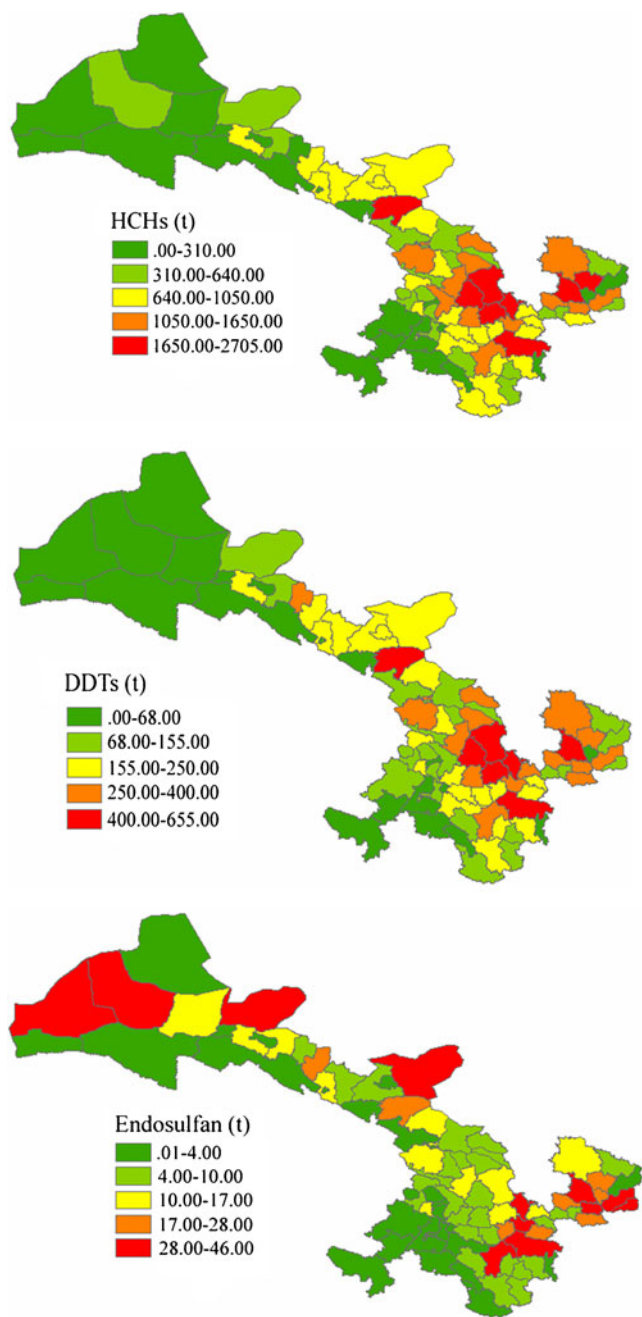


Fig. 6 Distribution of OCP usage based on prefectures

desert and gobi regions, which is very essential for policy makers.

Distribution of annual endosulfan usage was calculated on cotton from 1994 to 2007 and on wheat, apples, pears, and tobacco from 1998 to 2004 in China, and annual HCH usage from 1952 to 1983 and DDT usage from 1951 to 1983 were calculated according to area of dry farmland with a 1/4° longitude by 1/6° latitude resolution, and the results were given in Fig. 7. It shows that the most intensive use of endosulfan, HCHs, and DDTs was in northwestern and

Table 2 Top ten prefectures with the highest use of selected OCPs in Gansu province

Rank	Endosulfan (ton)		HCHs (ton)		DDTs (ton)	
	1994–2007		195–1983		1951–1983	
1	Dunhuang	45.1	Huining	2,701.4	Huining	652.1
2	Tianshui	41.0	Dingxi	2,421.3	Dingxi	588.0
3	Jinta	40.1	Zhenyuan	2,268.3	Jingning	554.7
4	Zhenyuan	37.7	Tianshui	2,065.6	Tianshui	501.2
5	Qinan	37.0	Jingning	2,036.9	Zhenyuan	496.0
6	Minqing	35.9	Tongwei	1,957.2	Qinan	470.0
7	Jingning	34.2	Wuwei	1,856.9	Tongwei	443.9
8	Jinchuan	34.1	Qingyang	1,652.1	Wuwei	394.9
9	Anxi	34.0	Ning	1,554.5	Qingyang	373.3
10	Li	39.8	Qinan	1,515.7	Ning	367.1

southeastern regions of Gansu province, which is due to developed agriculture in these regions. Figure 4 shows that endosulfan is mainly applied to wheat, apples, and cotton. In China, DDT was mainly used in agriculture for controlling armyworms, ball worm, pink ball worms, apple tortrix moths, and greenish brown hawk moths on wheat, maize, cotton, orchard, soybean, and sorghum (Cai et al. 1992). Crops on which HCH was widely used in China were rice, wheat, maize, cotton, soybean, sorghum, orchards, and some vegetables to attack a wide variety of pests for different crops (Wei et al. 2007). In Gansu province, wheat and apples were mainly distributed in the southeastern and Hexi corridor regions, and cotton was mainly concentrated in the northwestern region. In addition, a large area of desert and gobi existed in Gansu province, of which above 90 % are in the northwestern area. In addition, in the southwestern region, which plants crops, animal husbandry is the main economic pillar, and an extensive pasture is distributed in this region. Thus, OCPs were rarely used in the desert, gobi in the northwestern region, and pasture in the southwestern region.

Uncertainty

Uncertainty analysis for usage data is an important part of any inventory. It can clearly understand the limitations and deficiencies of the usage inventory, which can provide a basis for estimating the factors that lead to errors in the establishment of the inventory. Many factors could create uncertainties for these gridded usage inventories. For example, the regional and time differences were neglected in this study. Because they span a relatively large latitude range in Gansu province, natural conditions are very different for different regions, such as temperature and precipitation, which result in differences of pest situation for the same

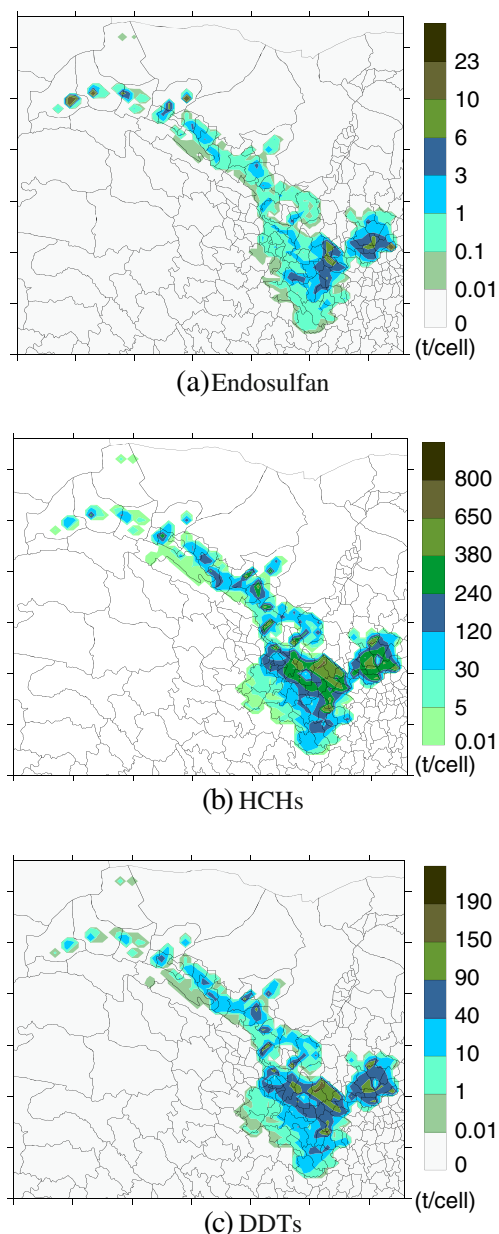


Fig. 7 Distribution of OCPs with a 1/4° longitude by 1/6° latitude resolution

crop in the different regions. Thus, for endosulfan, annual application frequencies and amount used for the same crop are different in the different regions. The application rate of endosulfan on the same crop could differ from year to year or season to season. In addition, the surrogate data that we used could be the sources for uncertainties in allocation of the usage. Ideally, statistical estimates of uncertainty would be provided for each province or grid cell. To make such estimates in this work is very difficult. In addition, except for dry farmland, the crops were also planted in a small number of other types of land in the grid with a 1/4°

longitude by 1/6° latitude resolution. The dry farmland with grid was selected as surrogate data to calculate the gridded inventories usage for endosulfan, HCHs, and DDTs, which also lead to the uncertainty of usage inventories.

Conclusions

Based on the total areas of major crops on which endosulfan was applied, the average application rates on individual crops, the fractions of the croplands that were treated by endosulfan, and the area of dry farmland treated with HCHs and DDTs, the temporal variation, spatial distribution of endosulfan usage from 1994 to 2007, HCH usage from 1952 to 1983, and DDT usage from 1951 to 1983 were generated at prefecture levels, as well as on a 1/4° longitude by 1/6° latitude gridding system.

The present study shows that, in Gansu province of China, the endosulfan between 1994 and 2007, HCHs between 1952 and 1983, and DDTs between 1951 and 1983 were approximately 701, 56,486, and 15,236 tons, and annual average use was 50, 1,712, and 462 tons, respectively. Endosulfan usage increased dramatically in 1998 due to application on other crops in 1998 except on cotton. The HCH and DDT usage displayed a rapid increase after 1972, reaching the peak in 1976 and in 1975, respectively; since then they declined until they were banned in 1983. For spatial distribution, the most intensive use of endosulfan, HCHs, and DDTs was in the northwestern and southeastern regions of Gansu province.

The inventories of gridded usage of endosulfan, HCHs, and DDTs in Gansu province of China with a 1/4° longitude by 1/6° latitude resolution will be used in creating the emission and residue inventories in the near future. This effort is especially important to understand POP usage information and reduce them for policy makers.

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