Global α-hexachlorocyclohexane emission with 1^ox1^o longitude/latitude resolution

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Abstract

Technical hexachlorocyclohexane (HCH) is an organochlorine insecticide used throughout the world. Total global technical HCH usage between 1948 to 1997 has been estimated as 10 million tonnes. Annual technical HCH usage data are apportioned to a grid system with 1^ox1^o longitude/latitude resolution by using a global cropland dataset as the surrogate data. Global emissions of α-HCH, the major isomer contained in technical HCH, are also gridded on a 1^ox1^o longitude/latitude grid system for 1980 and 1990 due to 10 consecutive years of application of this insecticide. Total global α–HCH emissions in 1980 were 217 kt, among which, 95.5% was contributed by the current use of technical HCH in 1980, and 4.5% by the residues due to usage since 1970. Total global α–HCH emissions in 1990 were 37 kt, among which, 92.9% was contributed by the current use of technical HCH in 1990, and 7.1% by the residues due to usage since 1980. Emissions in the South Polar region were zero in both 1980 and 1990. In 1980 approximately 80% of the global α–HCH emissions originated in the Northern Hemisphere midlatitudes, and followed by around 14% in the Northern Hemisphere tropics, while approximately 48% of the global α–HCH emissions originated in the Northern Hemisphere tropics, and followed by around 40% in the Northern Hemisphere midlatitudes in 1990.

1. Introduction

Hexachlorocyclohexane (HCH), also called benzene hexachloride (BHC), is an organochlorine insecticide available in two formulations: technical HCH and lindane. A total of eight HCH isomers have been identified in technical HCH; however, only the α-, β-, γ-, δ- and ε-isomers are stable and commonly identified in technical formulations. Lindane contains more than 90% of γ-HCH (*IRPTC*[1]). Although only the γ-HCH exhibits any significant insecticidal activity, all isomers are acutely and chronically toxic (Metcalf[2]; Agency for Toxic Substances and Disease Registry[3]; Smith[4]; Willett et al.[5]). Certain HCHs cause central nervous system, reproductive, and endocrine damage.

Technical HCH is the most widely used pesticide in the world. According to Li[6], global technical HCH usage from 1948 to 1997 was estimated to be around 10 million tonnes, far more than the tonnage of any other pesticide used in history. The usage of HCH and other organochlorine insecticides contributes to increased agricultural yield, protection of livestock, and the elimination of vector-transmitted disease. On the other hand, however, the global contamination caused by usage of technical HCH and other pesticides over the past five decades has been found in virtually every environmental compartment and every geographic region (Woodwell et al.,[7] Waid,[8] Tanabe,[9] Tanabe et al.,[10] Kawano et al.,[11] Barrie et al.,[12] Bidleman et al.,[13] Li[6]).

The atmospheric modeling community world wide has been trying to understand the sources, occurrence, and pathways of the contaminants and to predict how these inputs, and concentrations in the various environmental compartments may change in the future by developing various models to simulate the physical, chemical, and hydrometeorological process (Pudykiewicz & McMillan,[14] Strand,[15] Wania & Mackay,[16] Wania *et al.*,[17]). Recently, by using a zonally-averaged global multimedia distribution model, *Wania et al.*[17] discovered a dramatic decrease in the global emission of α-HCH that is reflected in rapidly decreasing concentrations in the atmosphere and sea water, except in the Arctic Ocean, which apparently is the last refuge for α-HCH in the global environment.

In order to obtain good results, all models require gridded emission data of the pollutants. The model results will vary significantly depending on the available emission data, and how this emission data is processed into input for models.

For organochlorine pesticides, efforts to estimate usage/emissions on a global scale were began in the early 1990s. In 1993, Voldner & Li[18] made an estimation for global use of toxaphene, and this effort was extended to several organochlorine pesticides, including technical HCH, lindane, and DDT (Voldner & Li[19]). The first global gridded usage inventories of α–HCH and γ–HCH were created in 1996 on a 1°x1° longitude/latitude grid system for circa 1980 and 1990 with large space gaps for numerous countries due to lack of information (Li et al.[20]). In order to fill the space gap, the relationship between technical HCH usage and environmental contamination on a local level has been studied. Li et al.[21] compared the use of technical HCH and contamination by this insecticide in

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China and Japan. The results of the comparison indicate that the contamination caused by use of this pesticide in China during the beginning of 1980s was comparable to that in Japan at the beginning of 1970s, due to the similarity of the total average use-density of technical HCH over arable land between China and Japan. Similar comparisons were also made for insecticide DDT (Li et al. [22]). Although the concentration data were produced from samples with rather limited spatial coverage, and are not necessarily broadly representative, the comparison made in these two papers suggests a clear linkage between environmental contamination by technical HCH and DDT and the average accumulated usage of these insecticides over arable land. Based on this assumption, a global annual usage inventory of technical HCH was produced (Li[6]). Global technical HCH usage data for the years of 1950, 1960, 1970, 1980, and 1990 are distributed to a grid system with 1°x1° longitude/latitude resolution by using a global cropland dataset as the surrogate (Li[23]).

This paper presents the continuous efforts described above. Section 2 presents α-HCH usage on a 1°x1° longitude/latitude grid system for 1980 and 1990. Gridded emission factors are introduced in Section 3. Combining the gridded α-HCH usage and emission factor, gridded α-HCH emissions on a 1°x1° longitude/latitude grid system for 1980 and 1990 are presented in Section 4. Section 5 gives the results and discussions to end the paper.

2. Gridded α-HCH Usage Data

Although small amount of α-HCH may result from isomerization of lindane (almost pure γ-HCH) upon exposure to sunlight, we only consider α-HCH from the use of technical HCH. Generally, technical HCH contains 55-80% of α-HCH (Metcalf[2]), and we use the value of 70% to convert technical HCH usage to α-HCH usage.

The gridded usage data for technical HCH from 1948 to 1990 (Li[23]) are used here to produce gridded α-HCH usage data for 1980 and 1990, shown in Figures 1 and 2, respectively. The total global usage of α-HCH was estimated to be around 312 kt in 1980, and 52.5 kt in 1990. The usage data for α-HCH, given in Figure 1, indicates that in 1980, while some countries, such as Canada, the United States, and Japan did not use this insecticide, the intensive use of α-HCH (more than 100 t of α-HCH per each grid cell) was in the eastern and central parts of China, the most parts of India, the North part of Nigeria, some area in Australia, and some western European countries. Fig. 2 shows that the main areas using this insecticide were mostly in regions of India and Vietnam in 1990, while China, the three Northern European countries, many western European countries, along with Canada, the United States, and Japan, did not use any amount of this insecticide.

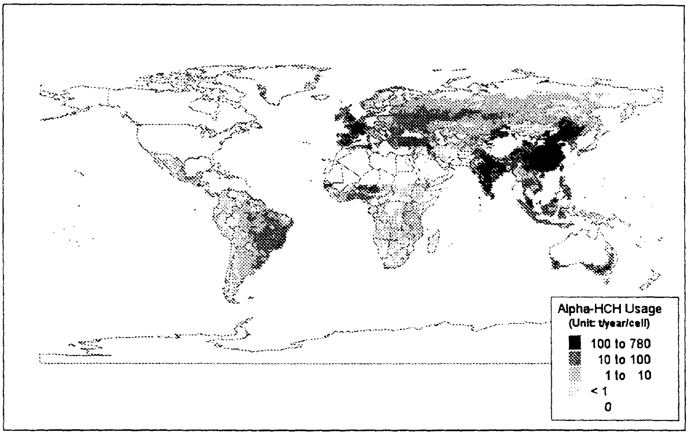

Figure 1. Gridded global α–HCH usage on a 1°x1° lat/long grid system for 1980.

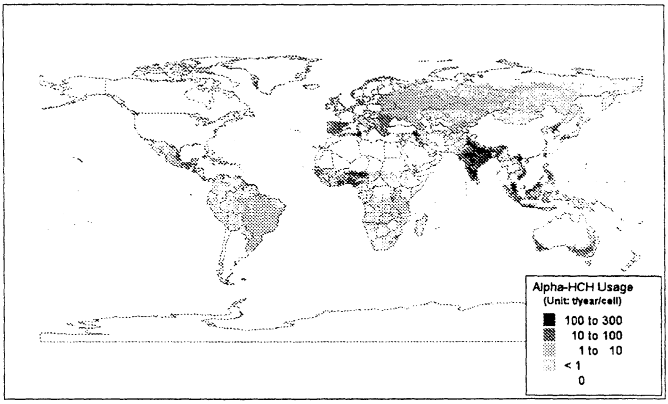

Figure 2. Gridded global α–HCH usage on a 1°x1° lat/long grid system for 1990.

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3. Emission Factors

In order to compile emission inventories for pesticides, emission factors should be calculated first. Emission factors represent the emission potential of pesticides in a geographical area and are not emission estimates on their own. The emission factor of organochlorine pesticides is a very complicated function of a large number of variables. It depends on physical and chemical properties of pesticides, application modes of the pesticides, the meteorological conditions when they are applied, the soil type, and the temperature. Prior to Scholtz et al.[24,25] there was no reliable way to estimate the emissions of pesticides to the atmosphere. Their study detailed estimated emission factors for twenty pesticides for North America[24] and on a global scale.[25]

Based on annual emission factor given by Scholtz *et al.*,[25] emission factors of α-HCH for the spray application mode and for soil incorporated application mode are compiled and given in Figures 3 and 4. There are some differences between the new sets of data presented here and the original one. First, the area where the emission factor covers have been redistributed from Matthew's cultivation density map[26] to a new cropland map (Li[23]). Secondly, emission factors given by Scholtz et al.[25] are one-year overall values, while those presented here are separated as two different kinds, one for spray mode, another for soil incorporation mode. Finally, emission factors in some tropical and sub-tropical region are modified substantially.

One year overall emission factors of α-HCH given by Scholtz et al.[25] show that the emission factors in the Southern India are in the range between 0.65 to 0.7, and those in the Southeastern Asian countries, such Philippines and Indonesia, in the range between 0.13 and 0.5. These ranges seem too low. For example, in Tamil Nadu, South India, it was estimated that about 99.6% of the applied HCH in the paddy areas of this watershed were removed to the air over a year (Tanabe[27]), which is much higher than the results predicted from Scholtz's model.

Figures 3 and 4 show that the emission factors for the spray application mode are much greater in magnitude than those for soil incorporation mode. This is not surprising since, for a spray application, the pesticide is totally contained within the upper 1 cm of the soil surface thus making most of the pesticide readily available for volatilization at the soil. If a vegetation canopy is present during a spray application, then a fraction of the applied pesticide will be intercepted by the canopy where it can volatilize directly to the atmosphere or drip to the soil during a precipitation event. By the time of the till in the second year, major portion of the α-HCH has already volatilized from the soil surface and canopy, and only a small portion remains in the soil ready for future emission.

In this paper, although emission factors were derived by using meteorological data in 1989, they are used for all years to calculate emissions of α-HCH.

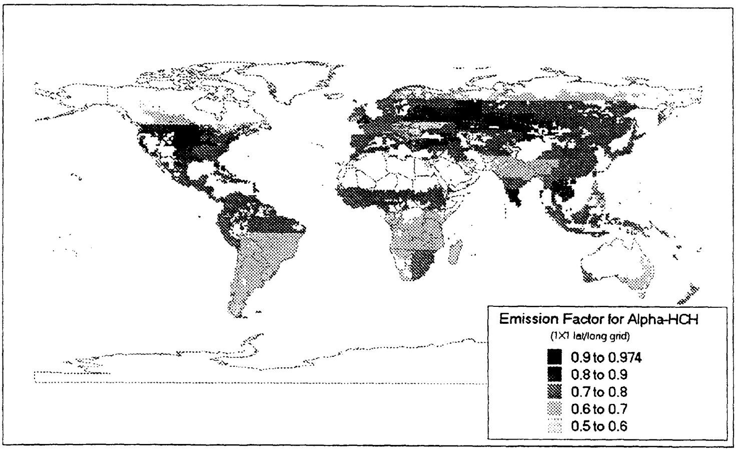

Figure 3. Gridded global emission factor of α–HCH for spray application mode on a 1°x1° lat/long grid system.

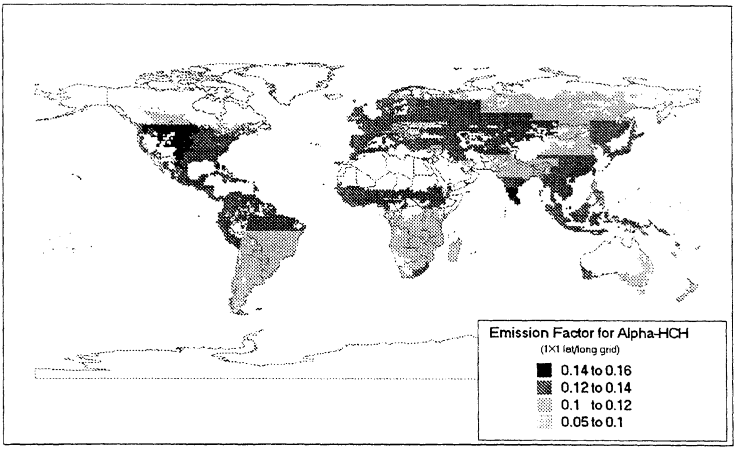

Figure 4. Gridded global emission factor of α–HCH for soil incorporation application mode on a 1°x1° lat/long grid system.

4. Emissions of α-HCH

Emissions of pesticides have been reported only locally. For example, Berdowsky et al.[28] produced emissions of γ-HCH in Europe for 1990. It is apparent that the computation of emissions on a grid covering the whole world will require considerable human and computer resources. The difficulty in undertaking such a large number of computations is that some of the input data required for the computations, such as timing and mode of application and degradation rates in the soil, are not well known. Within the scope of the present study, a number of assumptions are made for the sake of simplicity.

We assume that 90% of all technical HCH used in each country every year was applied as spray mode, and 10% applied as seed treatment mode. No technical HCH was used in the form of soil incorporation. When technical HCH is applied as a spray, the fraction of α-HCH emitted to the atmosphere is calculated by using emission factors for the spay mode. When technical HCH is applied in the form of seed treated, the insecticide is effectively applied beneath the soil surface with little, if any, exposed to the atmosphere. The residues in the soil are exposed to the air during the next tilling event. The fraction of α-HCH emitted to the atmosphere from the residues is calculated by using emission factors for the soil incorporation mode. Only one plant/harvest cycle is assumed in a given year, although it is not uncommon to have two or more plant/harvest cycles in a given year. This will not create too much error if we only consider annual emissions. Thus emissions of all α-HCH residues are assumed to occur at the time of tilling in each year. Although a half life of 400 days was used for α-HCH in Scholtz *et al.*'s study,[24,25] a value of one year (365 days) is used here.

The usage data and annual emission factors for α-HCH have been combined in order to estimate emissions to the atmosphere on a 1°x1° lat/long grid. For highly persistent insecticides, such as HCH, it would be necessary to consider the emissions due to several consecutive years of application. For the present study gridded usage of α-HCH from 1970 to 1980 are used for estimation of α-HCH emissions in 1980, and gridded usage of α-HCH from 1980 to 1990 are used for estimation of α-HCH emissions in 1990. The resulting emission estimates not only account for the present year's application but also any residual pesticide present in the soil from previous 10-year's application.

If we compare these two figures with Figs. 1 and 2, we see that although Japan and the United States banned the use of technical HCH in 1972 and 1976, respectively, Fig. 5 shows that α-HCH residues still remaining in the soil of these two countries, resulted in emissions to the atmosphere in 1980. Figure 6 shows that seven years after China banned the use of technical HCH in 1983, the residual α-HCH in Chinese soil still produced emissions to the atmosphere in 1990.

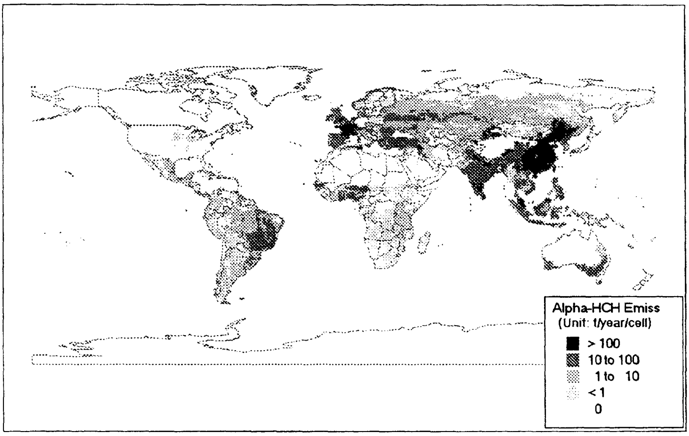

Figure 5. Gridded global annual emissions for α–HCH in1980.

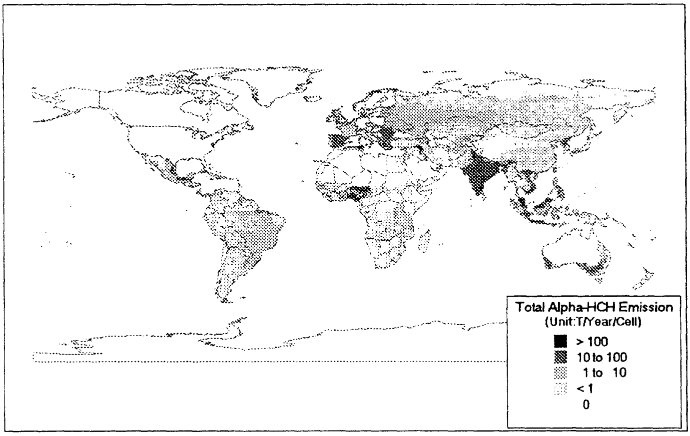

Figure 6. Gridded global annual emissions for α–HCH in1990.

5. Results and Discussion

The total global usage of α–HCH was estimated to be around 312 kt in 1980, and 52.5 kt in 1990. Total global α–HCH emissions in 1980 were 217 kt, among which, 95.5% was contributed by the current use of technical HCH in 1980, and 4.5% by the residues due to usage in previous 10 years. Total global α–HCH emissions in 1990 were 37 kt, among which, 92.9% was contributed by the current use of technical HCH in 1990, and 7.1% by the residues due to usage in previous 10 years. It is clear that as more and more countries banned the use of technical HCH, the portion of α–HCH emissions to the atmosphere to the total emissions due to residues has become larger.

Figure 7 presents the latitudinal distribution of annual α–HCH emissions. Emissions of α–HCH in 1980 concentrate in a latitude band between 20° N to 50° N as shown in Fig. 7 (a), while those in 1990 concentrate in a latitude band between 0° to 45° N as shown in Fig. 7 (b). It indicates that α–HCH emissions moved from higher latitudes towards lower ones, and underwent "southward tilt" (Goldberg[29]) from 1980 to 1990. An inverted latitudinal concentration profile of α–HCH in seawater was found in the late 1980s and early 1990s (Wania & Mackay[16]). The α–HCH concentration gradient in seawater was extending north from the Western Pacific toward the Canadian Arctic. This is called the "global distillation" phenomenon (Goldberg[29]).

Figure 8 summarizes the regional contributions to the global α–HCH emissions in the Northern and Southern Hemisphere. Emissions in the South Polar region were zero in both 1980 and 1990. As shown in Fig. 8 (a), approximately 80% of the global α–HCH emissions originated in the Northern Hemisphere midlatitudes, and followed by around 14% in the Northern Hemisphere tropics in 1980. The regional contributions to the global α–HCH emissions in 1990 are quite different. Fig. 8 (b) indicates that approximately 48% of the global α–HCH emissions originated in the Northern Hemisphere tropics, and followed by around 40% in the Northern Hemisphere midlatitudes in 1990.

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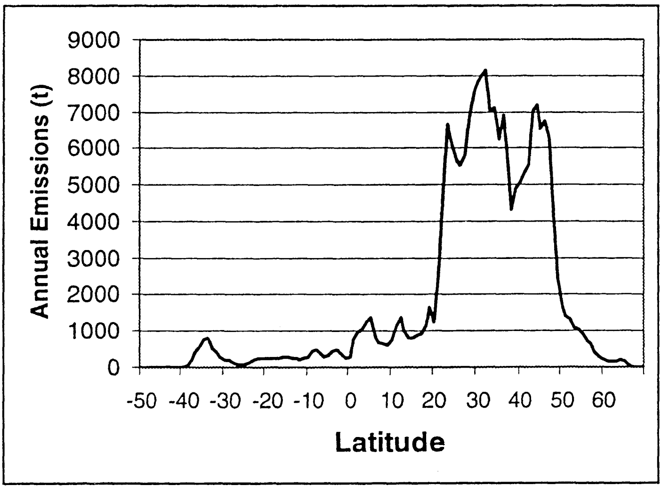
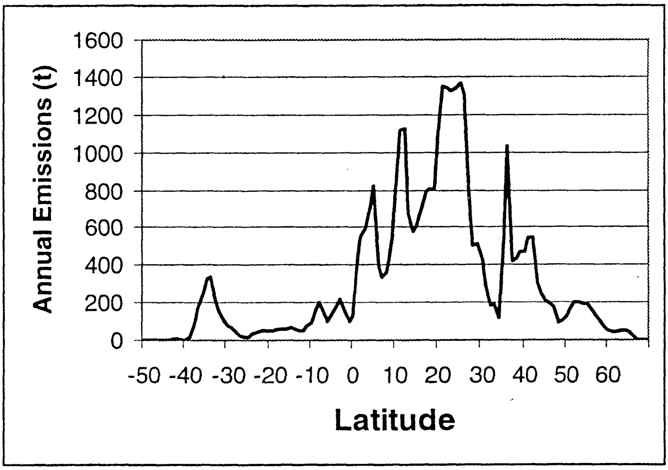
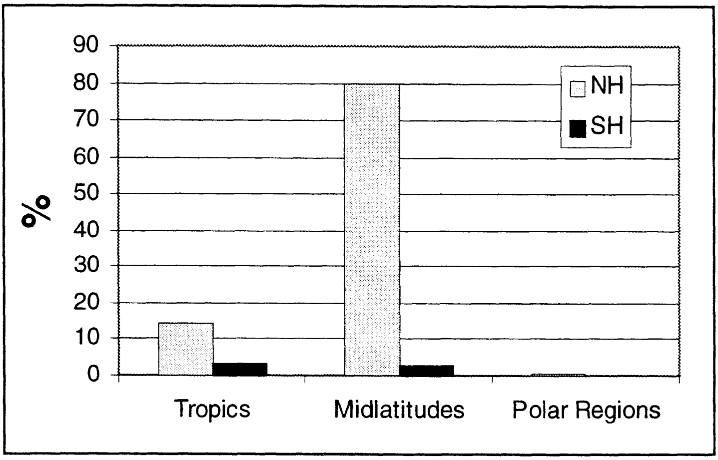


(a)

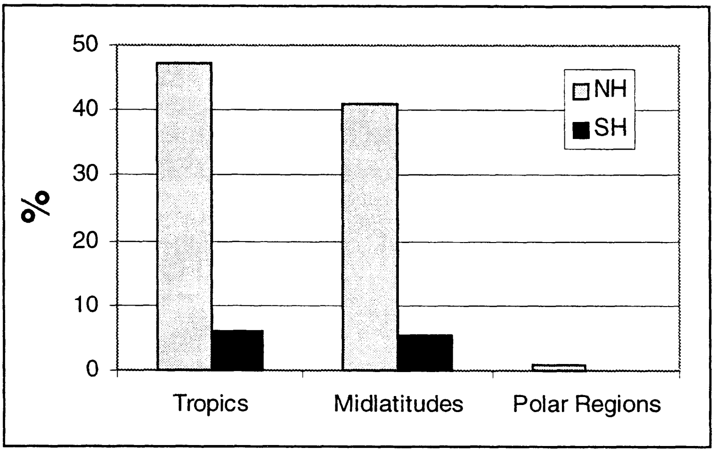


(b)

Figure 7. Latitudinal distribution of annual α–HCH emissions in (a) 1980 and (b) 1990.



(a)



(b)

Figure 8. Regional contributions to the global α–HCH emissions in (a) 1980 and (b) 1990 in the Northern (NH) and Southern Hemisphere (SH). The tropical regions are defined as the latitude band from the equator to 23.5°, the midlatitudes regions between 23.5° and 60°, and the polar regions from latitude 60° to the poles. Emissions in the Southern Hemisphere polar region are zero.

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