POPULATION EXPOSURE TO α-HCH SOIL RESIDUES IN CHINA

Tian CG[1,2], Ma JM[3], Li YF[3,1]

[1]International Joint Research Center-For Persistent Toxic Substances (IJRC-PTS), Harbin Institute of Technology, Harbin 150090,China; [2]School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China; [3]Science and Technology Branch, Environment Canada

Abstract

Based on the concept of intake fraction, a method for assessing population exposure was developed in this study. A coupled atmospheric transport, soil-air, and water-air exchange model was applied to estimate intake fractions of α-HCH across China. Results showed that the spatial trend of annual average α-HCH intake fraction was correlated inversely with its soil residues in latitudes and positively correlated with the residues in longitudes. Annual average values of α-HCH intake fraction were the highest in the provinces of Liaoning, Hebei and Shandong, associated well with the population density, α-HCH residues in soil and the proximity to the location of the major sources of this toxin.

1. Introduction

1,2,3,4,5,6-Hexachlorocyclohexane (HCH) is an organochlorine pesticide, and formed in two formulations: technical HCH and lindane. Due to its effectiveness and low price, technical HCH was one of the most widely used insecticides in the world. The total usage to date has been estimated to be as high as 10.0 million t.[1] During the 1970s and the beginning of the 1980s, China was the largest producer and user of technical HCH in the world. The total amount of technical HCH used in China is 4.5 million t. Technical HCH was mainly used in agriculture, although a small apportion was also used in forestry and public health.[2] Southern and southeastern China have been identified to be major users of technical HCH from the 1950s to 1980s. Although the use of this insecticide was banned on April 1, 1983 by the Chinese government, HCH residues are still found in air, soil, water, food, and living organisms. They still impose threat to human health and the ecosystem in China.[3]

Technical HCH produced in China contains 65-70% α-HCH, 5-6% β-HCH, 13% γ-HCH, and 6% δ-HCH.[4] Although γ-HCH exhibits also significant insecticidal activity, α-HCH is the dominant substance in the pesticide products. After technical HCH was banned in China, the composition of HCH isomers in air was changed with the composition of technical HCH isomers.[5] The composition of α-HCH in air was 5-65% in 2005.[6,7,8] While inhalation exposure dose of γ-HCH was 0.55% of total potential dose, α-HCH enters through the route of inhalation and is absorbed more easily. α-HCH has become a great threat to human health.[9] In the present study we used a new approach to estimate the inhalation exposure and health impact of α-HCH soil residues in 2005 in China based on the concept of intake fraction and a coupled atmospheric transport, soil-air, and water-air exchange model.

2. Methods

2.1 Concept of intake fraction

Intake fraction (IF) is a dimensionless parameter describing the fraction of material released from a pollutant source that is eventually inhaled by population. It is also called exposure efficiency, exposure effectiveness and

population inhalation transfer factor.[10] Annual average intake fraction can be written as:

$$IF = \frac{I}{Q} = \frac{3.65 \times C_{i,j} \times P_{i,j} \times B}{Q \times 10^{13}}, \quad (1)$$

where I is the intake rate of a pollutant and Q is pollutant emission rate; $c_{i,j}$ is the annual average air concentration at a grid cell (i,j) of model domain; $P_{i,j}$ is the population at the grid cell (i,j) ; B is the average breathing rate. In this research, a nominal breathing rate of 20 $m^3 d^{-1}$ was used. This value was commonly adopted by researchers.[10,11,12]

From the perspective of the health impact assessment, the concept of intake fraction is most useful for pollutants whose adverse health effects are proportional to cumulative exposure and whose health effects do not depend substantially on intake dosage rate. In these cases, the health impact cost is proportional to the cumulative exposure (or intake dosage) that is directly linked to emissions by intake fraction (Fig. 1).

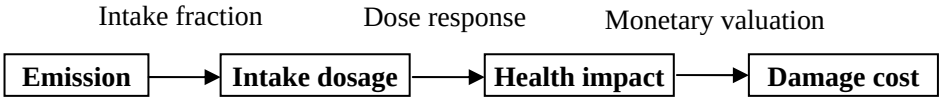

Fig.1 Application of intake fraction to health damage cost valuation

Once the intake fraction is obtained, the number of health effects HE (cases) can be readily estimated by the product of intake fraction , emission rate Q , and dose response coefficient b (case, pg m^{-3}) divided by the average breathing rate B,

$$HE = \frac{b}{3.65 \times 10^{-13} \times B} \times IF \times Q, \quad (2)$$

where Q is the annual emission rate, defined in the present study as annual α-HCH soil residues, IF is the annual intake fraction at each receptor cells, defined in Eq. (1).

2.2 Coupled Atmospheric Transport, Soil-Air, and Water-Air Exchange Model

The coupled model employed in this investigation is a three dimensional atmospheric transport model able to simulate transport and loadings of organochlorine pesticides at any model grid. The meteorological data required to drive the model can be either measured or predicted. The model has been used in previous numerical studies of lindane budget and toxaphene in the North American Great Lakes basin.[13,14,15,16] Briefly, it is a three-dimensional regional scale dispersion model coupled with a dynamic, three soil layer, fugacity-based soil-air exchange model, and a two-film model to estimate water-air gas exchange. In the present study the model has a horizontal resolution of 24 km × 24 km with 270×210 grids and the model domain covers China, Korea, and a large portion of Japan. The coupled model has 12 vertical levels from the surface to 7000 m height. The meteorological data (wind, air temperature and precipitation) at each time step are obtained by interpolating the 6-hourly objectively analyzed data from the United States National Center for Environmental Prediction (NCEP) reanalysis. The model is run from January 1 to December 31, 2005.

2.3 α-HCH Residues in China Soils

A α-HCH soil residues inventory in China in 2005 on a 1/6°×1/4° latitude and longitude grid system was used. These gridded residues were then interpolated to the model grids. Fig.2 shows interpolated α-HCH soil residues in the model domain. At the beginning of 2005 it was estimated that a total of 19548 kg of α-HCH residues were

still left in China agricultural soils due to accumulation from past use of this pesticide. In this study, α-HCH is introduced into the air only by reemission (volatilization) of its historical accumulated residues in the soil (no background air concentrations).

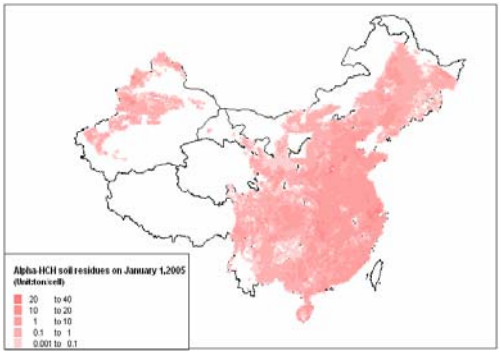

Fig.2 α-HCH soil residues on January 1, 2005 in China

2.4 Population distribution

Geographical population distribution was produced by a GIS system—MapInfo 8.0. The population was from officially reported China's county-level statistical data and was illustrated in an electronic map that consists of boundaries for all counties and urban areas. The whole domain was split into 24 km^2 grid cells along the x- (west-east) and y-axis (south-north), and population was allocated to the cells under the assumption that population in each county is evenly distributed across the county area.

3. Results and discussions

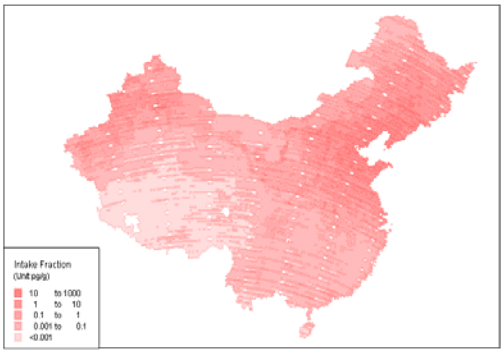

Fig.3 Molded α-HCH intake fraction in China in 2005

As shown in Fig 2, α-HCH soil residues were higher in southern China than northern China and higher in eastern China than western China. The population density was also greater in southern China as compared to northern China. However, the spatial pattern of α-HCH intake fraction (Fig 3) suggests that the annual intake fraction averaged over 2005 in China was correlated inversely with its soil residues in latitudes and positively correlated with the residues in longitudes. The spatial pattern of α-HCH intake fraction appears to be more uniformly distributed compared with α-HCH soil residues. This suggests that that emission of α-HCH residues in soil should not be overlooked even in those regions with a low population density, because atmospheric circulation would make a significant contribution to the spatial distribution of the intake fraction.

The top 3 provinces with the highest intake fraction of α-HCH are Liaoning, Hebei and Shandong. α-HCH intake fraction in these provinces was 51.1% of total annual average intake fraction in China (Fig. 4(a)). A nonlinear regression analysis was conducted using a statistical analysis package, SPSS13.0. The result reveals that population density, α-HCH soil residues and the proximity of receptors to the sources were key parameters in determining intake fraction in these three provinces, defined in Eqs.(3-5).

$$IF_{LIN} = -4.0 + 1.23\times10^{-6}\left(d(i,j)\times\sum\left(Q_{m,n}\times\ln r_{m,n}(x,y,z)\right)\right), \quad (3)$$

$$IF_{HEB} = -387.834 + 9.14\times10^{-6}\left(d(i,j)^{0.67}\times\sum\left(Q_{m,n}\times\ln r_{m,n}(x,y)\right)\right) + 0.005\times d(i,j), \quad (4)$$

$$IF_{SHD} = -371.956 + 0.003\left(d(i,j)^{0.1} \times \sum \left(Q_{m,n} \times \ln r_{m,n}(x,y)\right)\right), \quad (5)$$

where $d(i,j)$ is density of population at cell(i,j), $Q_{m,n}$ is α-HCH soil residues at cell (m,n), $r_{m,n}(x,y,z)$ and $r_{m,n}(x,y)$ are distance between cell (i,j) and cell(m,n).

Figure 4a illustrates α-HCH intake fraction and total population in each province and autonomous region in China, implying that the intake fraction for this toxic compound appears not only associated with the population, but also with factors, such as soil residues and the proximity to the sources, as we have discussed previously. Comparison between the predicted (using Eqs.(3-5)) and modeled intake fraction (using the model and Eq. 1) over the three provinces, Liaoning, Hebei and Shandong, is illustrated in Fig. 4 (b-d). As seen, the modeled intake fractions agree well with their predicted values by Eqs. (3-5).

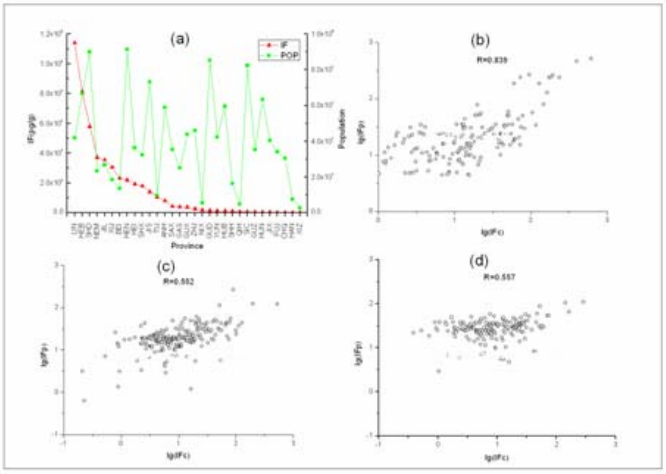

Fig.4 (a) α-HCH intake fraction and total population over each provinces and autonomous regions in China; (b) , (c) and (d) comparison of intake fraction between modeled IF and predicted IF using Eqs (3-5) in Liaoning, Hebei and Shandong, respectively.

References:

1. Li YF. *Sci Total Environ* 1999; 232:123.
2. Li YF, Cai DJ, Singh A. *Arch Environ Contam Toxicol*1998; 35:688.
3. Li YF, Cai DJ, Shan ZJ, Zhu ZL. *Arch Environ Contam Toxicol* 2001; 41: 261.
4. Cai DJ, Sun LJ, Ke JL, Tang GC. *Pesticide usage in China.* Prepared for Environment Canada, 1992, Downsview, Ontario, Canada.
5. Chen SJ. *Environ. Sci. Technol* 1987; 36:5.
6. Jiang K. *Master dissertation, the Graduate School, Qingdao University* 2006:31.
7. Geng CZ. *Master dissertation, the Graduate School, Qingdao University* 2006:53.
8. Li J. *Master dissertation, the Graduate School, the Chinese Academy of Sciences* 2005:66.
9. Dou Y. *Master dissertation, the Graduate School, Beijing University* 2006:77.
10. Hao JM, Li J, Ye XM, Zhu TL. *Journal of Environmental Sciences* 2003; 15:611.
11. Zhou Y, Levy JI, Hammitt JK, Evans JS. *Atmospheric Environment* 2003; 37:815.
12. Li J, Hao JM, Ye XM, Zhu TL. *Environ Sci*2003; 24:16.
13. Ma JM, Daggupaty SM, Harner T, Li YF. *Environ. Sci. Technol*. 2003; 37:3774.
14. Ma JM, Daggupaty SM, Harner T, Blanchard P. *Environ. Sci. Technol*. 2004; 38:984.
15. Ma JM, Venkatesh S, Li YF, Daggupaty S. *Environ. Sci. Technol.*2005; 39:8123.
16. Ma JM, Venkatesh S, Li YF, Cao Z, Daggupaty S. *Environ. Sci. Technol.* 2005; 39:8132.