



# Temporal–spatial loss of diffuse pesticide and potential risks for water quality in China



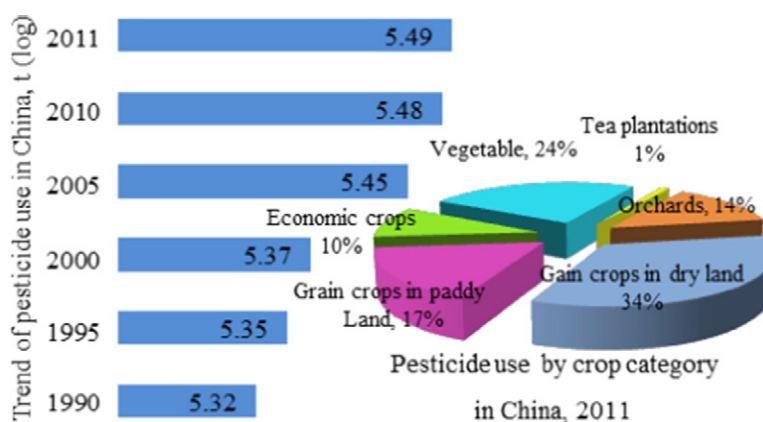
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## HIGHLIGHTS

- Pesticide uses of six crops in China over two decades were summarized.
- Temporal-spatial differences of three kinds of pesticides were highlighted.
- Spatial pattern of diffuse pesticide loss and potential risk for water was figured.
- Estimation uncertainties of pesticide use and loss were quantified.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 21 August 2015  
 Received in revised form 23 September 2015  
 Accepted 23 September 2015  
 Available online 3 October 2015

Editor: D. Barcelo

### Keywords:

Pesticide  
 Temporal–spatial pattern  
 Agricultural development  
 Diffuse pollution  
 Water risk  
 Uncertainty

## ABSTRACT

Increasing amount of pesticide has been used in Chinese agricultural system with effects on environmental quality and human health. The comprehensive inventory of pesticide use in six main crop categories over the period from 1990 to 2011 in China was conducted. The national average pesticide use intensity was estimated  $1.74 \text{ kg} \cdot \text{ha}^{-1}$  for grain crops in paddy land,  $1.31 \text{ kg} \cdot \text{ha}^{-1}$  for grain crops in dry land,  $1.38 \text{ kg} \cdot \text{ha}^{-1}$  for economic crops,  $3.82 \text{ kg} \cdot \text{ha}^{-1}$  for vegetables,  $1.54 \text{ kg} \cdot \text{ha}^{-1}$  for tea plantations, and  $3.49 \text{ kg} \cdot \text{ha}^{-1}$  for orchards. The pesticide use was estimated to be approximately  $5.24 \times 10^4 \text{ t}$  for grain crops in paddy land,  $1.05 \times 10^5 \text{ t}$  for grain crops in dry land,  $3.08 \times 10^4 \text{ t}$  for economic crops,  $7.51 \times 10^4 \text{ t}$  for vegetables,  $3.26 \times 10^3 \text{ t}$  for tea plantations, and  $4.13 \times 10^4 \text{ t}$  for orchards. Based on the pesticide use and loss coefficients for each category, the distribution of pesticide loss in China was calculated. Total pesticide loss in China was estimated about  $4.39 \times 10^3 \text{ t}$  in 2011. The pesticide loss from six main crop categories was about 14.84% for grain crops in paddy land of total pesticide loss, 33.31% for grain crops in dry land, 10.47% for economic crops, 26.37% for vegetables, 1.08% for tea plantations and 13.93% for orchards. The results indicated that the highest pesticide use intensity and highest pesticide loss rate occurred in China's eastern and central provinces. The Monte Carlo simulation was used to quantify the uncertainties associated with estimation of pesticide use and loss rate for the six types of crops. The potential risk to national water quality was assessed and the water in the provinces of Henan, Shandong, Hebei, Beijing and Shanghai was at high risk for pesticide pollution. The implication for the future agricultural and environmental policies on reducing the risk to environmental quality was also summarized.

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## 1. Introduction

The pesticides play important role in safeguarding crop yields, which are used in agricultural tillage around the world and contribute to global food safety (Carvalho, 2006). However, the excessive application of pesticides over the past half century has posed serious risks to human health and water quality (Kolpin et al., 1998). Pesticide residues in water and soil are the significant environment threats and have been classified as carcinogens pollutants in many countries (Dich et al., 1997; Bressa et al., 1997). In China, the food security is a national priority due to the big population and lead to widespread application of pesticides on farmland. Consequently, the pesticides have been detected in some Chinese rivers (Feng et al., 2003; Zhou et al., 2006). Thus, it is necessary to determine the temporal–spatial patterns of pesticide use and loss and to assess risk for water quality.

The total cultivation area in China increased to  $1.63 \times 10^7$  ha in 2013, and the agricultural planting structure has shifted in response to recent economic development (Zhao et al., 2008). The cultivated area devoted to grain and economic crops remain stable, but the areas sown with vegetables and orchards increased (Liu and Savenije, 2008). Given the differences in pesticide use for various crop categories, the variation in the spatial pattern of agricultural planting directly impacts pesticide use. It is well known that grain crops in paddy land, tea bushes and orchards are commonly planted in southern China, and the grain crops in dry land are concentrated in northern China (Ministry of Agriculture, 2011). However, there is limited understanding of the detailed temporal–spatial characteristics of pesticide use with crop categories.

Some parts of the pesticides sprayed on crops will remain in farmland, but some of them will enter the surrounding air, soil and water (Malone et al., 2004; Lefrancq et al., 2013). As the artificial organic compounds, the pesticides can remain in the environment for many years and may be transported over long distance (Scholtz et al., 2002). The potential environmental risks posed by pesticide loss during runoff and erosion are of considerable concern and have become a priority issue for the Chinese Environmental Protection Agency (Bao et al., 2012). The National Agricultural Diffuse Pollution Action Plan, which was issued in 2015, set detailed diffuse pollution control goal for the near future (Ministry of Agriculture, 2011). To effectively prevent water pollution from diffuse pesticide loss, the detailed information concerning the main types of pesticides used on crops at provincial scale is required for subsequent action.

Some methods have been used to assess pesticide loss at the watershed level, and the field monitoring of pesticide discharge is the basic method for determining pesticide concentrations on-site. The measurements also provide sound data for the assessment of larger areas (Müller et al., 2006). On the watershed scale, diffuse pollution modelling has gained popularity in recent decades (Bannwarth et al., 2014). However, these distributed models cannot provide reliable results on the national scale because of complicated territorial dynamics and limited monitoring data. Therefore, a multiple-year comprehensive inventory of pesticide use is employed in this study, which has been applied in diverse case studies (Sarigiannis et al., 2013).

To improve the understanding of diffuse pesticide pollution, we analyzed the temporal–spatial dynamics of Chinese pesticide use from 1990 to 2011. The potential pesticide loss due to runoff and erosion at a provincial scale was determined and used to evaluate the risk to water quality. The pesticide use and loss was investigated by crop categories, which provided information about the response to agricultural structure change and had implications for national water management. Finally, the uncertainty analysis was conducted to improve the accuracy and applicability of the estimation.

## 2. Methodology and key parameters

### 2.1. Pesticide use calculations

Three main types of pesticides (insecticides, herbicides and fungicides) account for more than 95% of the total use, which were calculated separately. The pesticide use amounts were estimated for the following crop categories: grain crops in dry land (GCDL), grain crops in paddy land (GCPL), economic crops (EC), vegetables (VG), tea plantations (TP) and orchards (OC). The GCDL referred to cereals (wheat and corn), soybeans and tubers. The GCPL was rice. The EC covered the oil-bearing crops, cotton, fiber crops, sugar crops and tobacco. The pesticide use amounts for these six categories were calculated using the bottom-up approach with a refined pesticide type database and agricultural planting structure database (Tian et al., 2014). The basic equation for pesticide use inventory was followed:

$$U_{Total}(n) = \sum_i U_i(n) = \sum_i \sum_m P_{i,m}(n) \times A_i(n) \times 0.001 \quad (1)$$

where  $U$  is the annual pesticide use amount for a crop category in China (t/year);  $P$  is the pesticide use intensity of crops (kg/ha);  $A$  is the cultivated crop area (ha/year); and  $i$ ,  $m$ , and  $n$  indicate the province (autonomous region or municipality), pesticide type, and crop category, respectively. The pesticides were counted based on their active ingredients.

The pesticide use intensity was defined as the annual pesticide use amount per unit of cultivated area (kg/ha). There are large differences in pesticide use intensity among crop types because of different growing conditions and the crop's resistance to disease, pests or weeds (Lucas, 2011). The pesticide use intensity factor was defined as the ratio of the pesticide use amount for a specific crop to that of grain crops, which was used to evaluate the pesticide use intensity for different crops. Based on previous studies, the pesticide use intensity factor was set at 1.0 for grain crops, 1.0 for economic crops, 2.5 for vegetables, 1.0 for tea plantations and 2.5 for orchards (Kolpin et al., 1998; Fu and Qi, 1998; General Administration, 2007; Short and Colborn, 1999). Pesticide use intensity was calculated as Eq. (2):

$$P_{i,m}(n) = G_{i,m} \times F(n) \quad (2)$$

where  $P$  is the pesticide use intensity of crop (kg/ha);  $G$  is the pesticide use intensity of grain crop (kg/ha); and  $F$  is the pesticide use intensity factor of crop.

The pesticide use intensity for grain crop was calculated as Eq. (3):

$$G_{i,m} = \frac{U_{i,m} \times 1000}{\sum_n A_{i,m} \times F_n} \quad (3)$$

where  $U$  is the pesticide use amount by province (t);  $F$  is the pesticide use intensity factor of crop; and  $A$  is the annual cultivated crop area (ha).

The planting structure and cultivated area data of crops in each province from 1991 to 2011 was obtained from the China Statistical Yearbook. The pesticide use data at a provincial scale over the same period was referred to the Development Center of Science and Technology, Ministry of Agriculture, China.

### 2.2. Pesticide loss calculation from farmland

The representative chemicals of insecticide, herbicide and fungicide categories were chosen to define the pesticide loss coefficient. The chlorpyrifos and carbendazim were chosen as the representatives for insecticides and fungicides, respectively. The acetochlor and butachlor were selected as the representatives of herbicides used on crops in dry land and paddy land, respectively. The use amounts of these four

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