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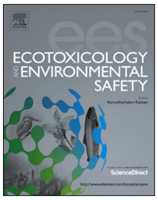

Integrating bioavailability and soil aging in the derivation of DDT criteria for agricultural soils using crop species sensitivity distributions

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ABSTRACT

Although the agricultural use of dichlorodiphenyltrichloroethane (DDT) has been banned for decades in many countries around the world, the detection of DDT and its metabolites in soils is very common due to their persistence. DDTs (sum of DDT and its metabolites) in farmland soils can be absorbed by crops at different levels and accumulate in the edible parts of agricultural products, posing threats to the health of human being. However, no information on the species sensitivity distribution (SSD) of crops with regard to DDTs has been reported due to the lack of enough bioavailability data and models to normalize the bioavailability data from different sources. Based on the bioconcentration factors of 17 crop species in Chinese soils obtained from previous studies, the criteria of DDTs in soils was derived according to the quality standard of agricultural products using the SSD method. Corrections for water content and aging time were conducted to normalize the data from different sources. The risk values of agricultural products at different concentration levels of DDTs in soils were also evaluated. It was found that oil crops are able to take up more DDTs than non-oil crops, so the soil criteria were calculated separately for oil crops and non-oil crops, which were 0.083 mg/kg and 0.29 mg/kg, respectively. With the residual concentrations of DDTs in soils at the range of 0.01–0.5 mg/kg, 0–8% of the agricultural products exceeded the permissible limits for DDTs which were set in the National Food Safety Standard of China. The results also demonstrated the feasibility for applying SSDs to derive the soil criteria of DDTs in order to ensure the safety of agricultural products. This work will provide information for the risk assessment and the establishment of soil environmental quality standards to ensure safe agricultural production.

1. Introduction

Dichlorodiphenyltrichloroethane (DDT) was one of the most widely used organochlorine pesticides throughout the world until the 1970s. p,p′-DDT as well as its two metabolites: 2,2-bis-(chlorophenyl)-1,1-dichloroethane (p,p-DDD) and 2,2-bis-(chlorophenyl)-1,1-dichloroethylene (p,p′-DDE) are classified as persistent organic pollutants, which have posed detrimental effects on the environment due to their bioaccumulation, biomagnification and high persistence properties (ATSDR, 2002). As a result, DDT has been banned in Canada, the United States, China, and many other countries. It is estimated that at least 0.4 million tons of DDTs were produced in China from the 1950s through 1983 when their production was banned, accounting for 20% of the global production (Hua and Shan, 1996).

Although the agricultural use of DDT has been banned for decades, the occurrence of DDT and its metabolites in soils is very common due to their persistence. Moreover, recent inputs of DDT to soils were also reported due to its usage as an anti-malaria agent or as an impurity in other pesticides such as dicofol (Yi et al., 2013). A few ng/g to several hundreds ng/g of DDTs have been detected in agricultural soils from different countries (Manirakiza et al., 2003; Pan et al., 2017). As hydrophobic organic compounds, DDTs have a strong affinity to soil organic matters, leading to half-lives of up to 30 years (Mitton et al., 2014). DDTs in soils can be taken up by crop roots, then translocated to the aerial portion via transpiration stream and eventually accumulated in the edible part of the products, posing threat to the health of human beings (Gaw et al., 2008). Agricultural products contained levels higher than the maximum residue limits were even reported in recent years (Wu et al., 2017). Thus, it is still necessary to implement long-term monitoring and control of soil DDT contamination. Derivation of DDTs criteria is crucial to the prevention and control of soil contamination and the quality security of agricultural products.

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The soil criteria of toxicants can be derived using a method of species sensitivity distribution (SSD), which was first proposed by US EPA to develop water quality benchmarks (Kooijman, 1987). SSDs are models of the variation in sensitivity of species to a particular stressor, and are generated by fitting a statistical or empirical distribution function to the proportion of species affected as a function of stressor concentration or dose (Garner et al., 2015). There are usually three steps for the construction and application of SSD models: 1) obtain the toxicity data and normalize to minimize the effects of soil characteristics on the toxicity values so the resulting toxicity data will reflect more closely the inherent sensitivity of the test species to the contaminant (Heemsbergen et al., 2009), 2) fit the cumulative distribution of the data to get the SSD curve, and 3) estimate the hazardous concentration at which 5% of species are harmed (HC5) and the potentially affected fraction (PAF) of species that will be harmed from exposure to the toxicant. SSD has been widely used to develop environmental quality benchmarks/guidelines for heavy metals in soil and water media. Nowadays, there has been increasing application of SSD in the determination of guidelines for organic pollutants. For example, the predicted no effect concentrations or the soil hazardous concentrations of triclosan and bisphenol A (2,2-bis[4-hydroxyphenyl]propane) were derived using the SSD method (Amorim et al., 2010; Kwak et al., 2018).

There are many factors influencing the uptake and accumulation of DDTs by crops: 1) the crop species (Gaw et al., 2008), 2) the content of DDTs in soil and the aging time (Gaw et al., 2008; Kiflom et al., 1999; Yao et al., 2007), and 3) soil properties and farming conditions, such as organic matter content and irrigation (Kiflom et al., 1999; Yao et al., 2007). Therefore, the specific situation of these factors in Chinese soils should be taken into account in the determination of the threshold of DDTs.

The term of aging was applied to the phenomenon that the availability of certain chemicals that have been in soil for some time is less than freshly added compounds (Alexander, 2000). It has been demonstrated that aging significantly reduced the bioavailability of DDTs (Morrison et al., 2000). Sorption, microbial degradation and chemical reactions might be the important processes leading to the decrease in extractable fraction of organic pollutants. However, after an initial and quick dissipation, it is usually observed that there is a continuing but slow process that organic pollutants can be gradually sequestered into soil nanopores and the organic matter matrices and become less available to soil microorganisms, plants, and animals (ATSDR, 2002). This slow process can occur over weeks to years (Alexander, 2000) and can be described by the simplified Elovich equation (Aharoni and Sparks, 1991).

According to the soil environmental quality standard of China (SEPAC, 1995), the environmental standard for DDTs in agricultural lands is 0.5 mg/kg, which was derived using the model from the former Soviet Union on the basis of soil hygienic standard for pesticides (Xia, 1996). In the draft of the risk control standard for soil contamination of agricultural land, which was released in 2018 (MEPPRC and GAQSIQPRC, 2018a), the risk control standard for DDTs were adjusted to 0.1 mg/kg in accordance with the environmental quality evaluation standards for farmland of edible agricultural products (SEPAC, 2006). No scientific basis for this revision could be found in the explanation document (MEPPRC and GAQSIQPRC, 2018b). Moreover, it remains unknown to what extent these standard values ensure the safety of agricultural products. Although there are some bioavailability data reported on the accumulation of DDTs by crops (Gaw et al., 2008; Kiflom et al., 1999; Tao et al., 2005), how to construct a SSD curve using these data is still a challenge due to the different soil properties and DDT contents which made the comparison of species sensitivity almost impossible. Therefore, the objectives of this study were to: 1) establish a method to normalize the data of bioconcentration factor (BCF, ratio of DDT concentration in plant to that in soil) from different sources and calculate soil DDT thresholds for individual crop species based on the normalized BCFs and the quality standards of agricultural products, 2) construct the SSD curve using the soil DDT thresholds of different crop species and derive the criteria of DDTs in China soils with the SSD method, and 3) evaluate the risks of agricultural products at different concentration levels of DDTs in soils. The results of this study will provide information for the risk assessment and the establishment of soil environmental quality standards to ensure safe agricultural production.

2. Materials and methods

2.1. Establishment of BCF database

The BCFs of DDTs for the edible part of crops are essential to the calculation of soil DDT thresholds of different crop species which were subjected to the construction of SSD curves. The BCF data were mainly obtained through a literature review including research papers, reports, etc. Bibliographic databases that were used for this end included Chinese databases (CNKI, VIP and Wanfang) and an English database (SCIE). BCF data were accepted only if the soil and crop were sampled simultaneously.

Three kinds of data were collected, including the BCFs of DDTs for the edible part of crops, the contents of DDTs in the edible part of crops and those in the corresponding soils, and the graphical data of BCFs or DDT contents.

To the data regarding the contents of DDTs in the edible part of crops (C_p, ng/g) and in the corresponding soils (C_s, ng/g), BCF is calculated using Eq. (1) (Trapp and Legind, 2011)

$$\mathrm{BCF} = \frac{C_p}{C_s} \tag{1}$$

To the graphical data, a digitizer software, Getdata Graph Digitizer (version 2.20), was used to get the BCF data or the contents of DDTs in the edible part of crops and in the corresponding soils and were then processed as described above.

As the maximum residue limits for DDTs set in the National Food Safety Standard (NHFPCPRC and MAPRC, 2016) are measured on the basis of wet weight for vegetables or the grain moisture content for safe storage purpose, the BCFs calculated using DDT contents on dry weight basis in some studies were extrapolated to wet weight or moisture content for safe storage purpose using Eq. (2)

$$BCF_w = BCF_d{*}(1{-}\theta) \tag{2}$$

where θ is the water content of vegetables or the safe storage moisture of grains (in %).

For a given crop species, there may be several BCF values available. In this case, the geometric mean of the BCF values was calculated as the BCF of this species. Besides, if the DDT content data of crops and soils were obtained from artificially spiked soils in laboratory experiments, the aging effect on the bioavailability of DDTs to crops should be incorporated in the determining of soil DDT criteria. The Elovich equation as described below was used to simulate the aging process in the present study due to its goodness in fitting the aging data of DDTs.

$$C_s(t) = A + Blnt \tag{3}$$

Where $C_s(t)$ is the extractable DDTs in soils at time t (mg/kg), t is aging time (d), and A and B are constants. The linear regression analysis was performed using OriginPro 8 (OriginLab Corporation, Northampton, MA). The fitted values of A and B are 26.5 and -1.90, respectively (coefficient of determination: $R^2 = 0.996$, $P < 0.01$).

Then, the content of DDTs in the edible part of crops grown in the soils aged for a time period of t ($C_p(t)$, mg/kg) can be estimated using Eq. (4) (USEPA, 2011)

$$log_{10}C_p(t) = alog_{10}C_s(t){+}b \tag{4}$$

where a and b are constants with fitted values of 2.36 and -1.94 for Chinese cabbage ($R^2 = 0.993$, $P < 0.01$), and 3.31 and -3.04 for Bok