



## Review

# Inventories of heavy metal inputs and outputs to and from agricultural soils: A review

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## ABSTRACT

Heavy metal pollution of agricultural soils is an important issue around the world. To understand the overall pollution process, accurate determination of every input and output pathway of heavy metals to and from soils is essential. Hence, input and output inventory, a quantitative analysis method of heavy metals balance in agricultural soils, has been widely used. However, due to differences in geography, climate, socioeconomic factors, industrial and agricultural production, substantial variation exists among existing input and output inventories for different countries and regions. In this study, we systematically analyzed these differences and the findings will improve the compilation of inventories worldwide.

## 1. Introduction

The accumulation of heavy metals in agricultural soil has become an important issue around the world (Bigalke et al., 2017). Great efforts have been made to alleviate heavy metal pollution in agricultural soils. First, accurate determination of the input and output pathways of heavy metals to and from soil is needed. Heavy metals can enter an agroecosystem through natural and anthropogenic processes. The application of fertilizers, agrochemicals and irrigation water, as well as atmospheric deposition are recognized as main anthropogenic heavy metal sources to agricultural soils (Hou et al., 2014). In addition, heavy metals in soils have several output pathways, such as biomass exportation, leaching and surface water runoff (Salman et al., 2017). To accurately quantify heavy metal inputs and outputs to and from agricultural soils, the concept of an inventory was developed. An inventory is a method of source apportionment that can be used in conjunction with source (i.e., diffusion) and receptor models. The main areas studied in inventory research include heavy metal inputs to agricultural soils (Nicholson et al., 2003; Luo et al., 2009; Jiang et al., 2014), inputs of heavy metals into soils by atmospheric deposition, fertilizer application, organic waste disposal (Lopes et al., 2011), heavy metal balances of agroecosystems (Moolenaar and Lexmond, 1998), and an emission inventory of heavy metals from the iron and steel industries (Wang et al., 2016a).

The quantification of an inventory can provide a cheap, quick and early diagnosis of agricultural soil contamination by heavy metals (Steiger and Obrist, 1993). The first such inventory was established in the Netherlands to periodically record environmental pollution in 1974 (Misseyer and Van der Most, 1993). Much of the subsequent research has been conducted in European nations, such as France and Germany (Belon et al., 2012), in the United States (Mcgrath et al., 1994) and in China (Luo et al., 2009). An inventory of heavy metal inputs and outputs gives a good indication of the accumulation or depletion of certain elements in selected agroecosystems. It also provides a valuable knowledge base for the improvement of agricultural soil management and development of policy recommendations. Physical geography, climate, economic development, industrial production and citizens' lifestyles differ markedly among countries and regions, resulting in differences in heavy metal inventories and the relative contributions of the input and output pathways to and from soils. However, limited information about these differences in soil inventories among countries and regions is available. In this research, we retrieved totally 260 concerned literatures corresponding to the keywords: inventory, heavy metals, agricultural soil, fluxes, mass balance from databases like web of science, Elsevier ScienceDirect (SDOL), Science Online, Chinese periodical full text, China national knowledge infrastructure (CNKI).

In this research, scattered literatures were utilized to answer the

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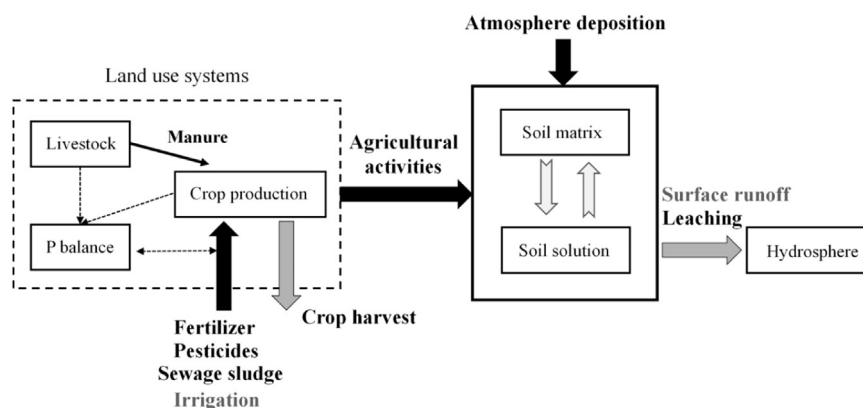


Fig. 1. General scheme for the regional metal balance model PROTERRA-S (Irrigation and Surface runoff are complementary to the PROTERRA-S model).

following questions: How to establish an inventory? How are inventory studies going? Are there differences in main input and output pathways of heavy metals among different regions or countries? What needs to be improved of inventory in the future? To our knowledge, this review is the first to examine heavy metal input and output inventories for agricultural soils.

## 2. Inventory methodology

### 2.1. Determine input and output pathways

In order to develop a quantitative inventory, firstly, we need to determine the main input and output pathways. It is need to determine the scale and relative importance of different sources and sinks of heavy metals. Dominant soil heavy metal input and output pathways in agroecosystem as shown in Fig. 1.

Heavy metal inputs into agricultural soils occur due to atmospheric deposition; the application of commercial fertilizers, animal manure and irrigation water; and the use of sewage sludge and other wastes as soil amendments. Pesticides containing Hg and As were banned in the United Kingdom since 1983 (Nicholson et al., 2006). The agricultural use of Hg-, As- and Pb-containing pesticides has been totally prohibited in China since 2002, and only a small number of approved pesticides contain other trace elements such as Cu and Zn (Luo et al., 2009). So the dominant soil heavy metal input pathways exclude pesticides in some studies.

Atmospheric deposition of particles to agroecosystems occurs via wet and dry processes. Heavy metal inputs by atmospheric deposition are obtained by direct collection using various sampling devices or indirect calculation with models (Dulac et al., 1989). The application of fertilizer can be a significant contributor of potentially hazardous heavy metals (Belon et al., 2012). For example, phosphate fertilizers are generally the major sources of trace metals among all inorganic fertilizers, and much attention has been given to the concentration of cadmium (Cd) in phosphate fertilizers (Bigalke et al., 2017). For agricultural soil to which sufficient nutrients cannot be supplied with commercial fertilizers alone, livestock manure is often used (Nicholson et al., 2003). Sewage sludge is also identified as a significant source of heavy metals where the material is excessively applied (Nicholson et al., 2006). Industrial byproducts and composts have often been neglected in soil inventories due to the lack of required information. Some other potential sources of heavy metal inputs to agricultural soil (e.g., flooding events, accumulating waste and spray) have been identified as being of local importance, but were not included in the previous inventories because of difficulties in estimating their contributions to overall heavy metal inputs.

Pathways of soil heavy metal outputs mainly include crop harvesting, leaching water and surface runoff (Fig. 1). Crop harvesting, one

of the major sinks, has been observed and evaluated. It is well-known that the accumulation of heavy metals in plants, particularly for hyperaccumulators, is in accordance with a potential risk of transferring them to people. Cd contamination in rice in Hunan, China is a major environmental health concern (Wang et al., 2016b). Moreover, with end uses of harvested residues (such as straw) as fertilizers, animal feed, building material and fuel, they play an important role in heavy metal cycling within arable land systems with resulting consequences for human exposure (Williams et al., 2009). The heavy metal output through crop harvesting has included the grazing of agricultural soil in some studies (Ahmadi et al., 2016). Generally, plant samples from a specific area can be sampled and chemically analyzed to calculate their output fluxes (Xia et al., 2014). The soil output fluxes of heavy metal from harvesting can be calculated by summing the heavy metal stocks in the seedling and seed parts of each sample, with that of the root excluded as in Vries and Mclaughlin (2013). Intensively cultivated agricultural soils always have a high potential for losses of water and nutrients by leaching. Therefore, leaching may be an important pathway for soil heavy metal output, which may be related to precipitation (Dobson et al., 2012). Soil heavy metals can also be removed by surface water runoff, with levels calculated by consideration of the dissolved and particulate forms (Dorioz, 2013).

### 2.2. Data collection

Secondly, in order to develop a quantitative inventory, we need to collect relevant data. The compilation of an inventory depends on the availability of appropriate data and its robustness. Databases used to develop inventories have been obtained from statistical yearbooks, scientific literature, regional agricultural statistics, soil information systems and model calculations. The data typically collected include the area of cultivated land, consumption of chemical fertilizers, precipitation, sown area of crops, irrigation volume, and output of major farm products in the target areas. Data for heavy metal concentrations in crops, fertilizers and manures are typically obtained from previous studies and reports, or by sample analysis. Data sources for input and output pathways differ primarily with respect to the spatial scale, which influences the precision of the simulation results. When developing inventories, such differentiation has to been typically processed. To make the estimates comparable, all input and output fluxes are given as rates per unit area of arable land for the entire study region.

### 2.3. Principal models applied in establishing inventories

Various models have been proposed to assess heavy metal balances in agricultural soils. Steiger and Obrist (1993) developed PROTERRA model to assess phosphorus and heavy metal balances in regional agroecosystems. Keller et al. (2001) extended this model to incorporate

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