Science of the Total Environment 543 (2016) 609–619

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Surface modeling of soil antibiotics

Wen-jiao Shi [a], Tian-xiang Yue [b,*], Zheng-ping Du [b], Zong Wang [a,d], Xue-wen Li [c,*]

[a] Key Laboratory of Land Surface Pattern and Simulation, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, 11A, Datun Road, Anwai, Beijing 100101, China

[b] State Key Laboratory of Resources and Environment Information System, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, 11A, Datun Road, Anwai, Beijing 100101, China

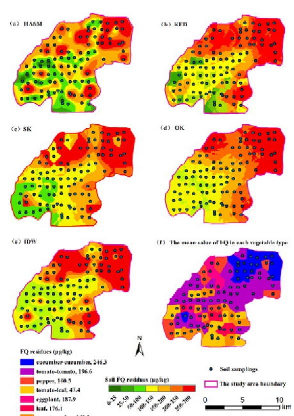
[c] Department of Environment and Health, School of Public Health, Shandong University, Jinan 250012, China

[d] School of Earth Sciences and Resources, China University of Geosciences, Beijing 100083, China

HIGHLIGHTS

- An effective method — HASM, was developed for the interpolation of soil antibiotics.
- The accuracy of HASM is higher than the classical methods.
- HASM can make the map more consistent with the true spatial distributions.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 17 May 2015
Received in revised form 16 November 2015
Accepted 16 November 2015
Available online xxxx

Editor: D. Barcelo

Keywords:
HASM
Soil antibiotics
Spatial distributions
Accuracy

ABSTRACT

Large numbers of livestock and poultry feces are continuously applied into soils in intensive vegetable cultivation areas, and then some veterinary antibiotics are persistent existed in soils and cause health risk. For the spatial heterogeneity of antibiotic residues, developing a suitable technique to interpolate soil antibiotic residues is still a challenge. In this study, we developed an effective interpolator, high accuracy surface modeling (HASM) combined vegetable types, to predict the spatial patterns of soil antibiotics, using 100 surface soil samples collected from an intensive vegetable cultivation area located in east of China, and the fluoroquinolones (FQs), including ciprofloxacin (CFX), enrofloxacin (EFX) and norfloxacin (NFX), were analyzed as the target antibiotics. The results show that vegetable type is an effective factor to be combined to improve the interpolator performance. HASM achieves less mean absolute errors (MAEs) and root mean square errors (RMSEs) for total FQs (NFX + CFX + EFX), NFX, CFX and EFX than kriging with external drift (KED), stratified kriging (StK), ordinary kriging (OK) and inverse distance weighting (IDW). The MAE of HASM for FQs is 55.1 μg/kg, and the MAEs of KED, StK, OK and IDW are 99.0 μg/kg, 102.8 μg/kg, 106.3 μg/kg and 108.7 μg/kg, respectively. Further, RMSE simulated by HASM for FQs (CFX, EFX and NFX) are 106.2 μg/kg (88.6 μg/kg, 20.4 μg/kg and 39.2 μg/kg), and less 30% (27%, 22% and 36%), 33% (27%, 27% and 43%), 38% (34%, 23% and 41%) and 42% (32%, 35% and 51%) than the ones by KED, StK, OK and IDW, respectively. HASM also provides better maps with more details and more consistent

* Corresponding authors.
E-mail addresses: yue@lreis.ac.cn (T. Yue), lxw@sdu.edu.cn (X. Li).

http://dx.doi.org/10.1016/j.scitotenv.2015.11.077

maximum and minimum values of soil antibiotics compared with the measured data. The better performance can be concluded that HASM takes the vegetable type information as global approximate information, and takes local sampling data as its optimum control constraints.

1. Introduction

Because of the high nutrients requirement for vegetable growth and the restricted use of chemical fertilizers for organic vegetable productions, large numbers of livestock and poultry feces were applied into the soils of the intensive vegetable cultivation areas (Bound and Voulvoulis, 2004). Due to the widely use of antibiotics in treating disease, protecting animal health and improving the feeding efficiency of animals, some of the antibiotics via the manure application were imported in soils and been strongly adsorbed (Sarmah et al., 2006; Martínez-Carballo et al., 2007; Aust et al., 2008). Further, soil antibiotics can be transported to surface and ground water, and also can be taken up by plants (Kuchta and Cessna, 2009). The spatial distributions of soil antibiotics are complex, because they are all from the external sources, and have irregular transportation into water and the irregular absorption by different plants.

Given that soil application of animal manure as a supplement to organic fertilizer is a common practice in many countries, there is a growing international concern about the spatial distributions of antibiotic residues in the environment (Sarmah et al., 2006). Because an inaccurate spatial pattern estimation of the soil antibiotics will result in considerable bias in risk assessment, the spatial interpolation of soil antibiotics is vital to evaluate the potential risks to human and environment (Xie et al., 2012). Therefore, it is needed to develop an effective surface modeling method which is suitable to interpolate the spatial patterns of soil antibiotics.

Several interpolators, such as kriging (Stein et al., 1988; Stein and Corsten, 1991; Goovaerts, 1999; Webster and Oliver, 2001), inverse distance weighting (IDW) (Weber and Englund, 1992; Weber and Englund, 1994; Gotway et al., 1996; Panagopoulos et al., 2006) and splines (Webster and Oliver, 2001), can be used for the estimation of the spatial distributions of soil properties, and each one has its own limitations (Shi et al., 2009; Shi et al., 2011; Shi et al., 2012). Some previous studies about soil antibiotics showed that the distributions of antibiotic residues in soil follow a spatial stratification pattern in the east of China (Xie et al., 2012), so the spatial predictions of soil antibiotics are more difficult than other soil properties due to the diverse sources of soil antibiotics.

Soil sampling analysis can provide highly accurate data of soil antibiotics at sampling sites, but these sampling points are sparsely to satisfy the interpolation requirement. The previous studies show that the vegetable type is the major determinant of the spatial stratification of FQs in the soil, and different vegetables also have different accumulation ability. These findings provide us clues to interpolate soil antibiotics more accurately by combining the interpolators with vegetable types.

High accuracy surface modeling (HASM) is a spatial interpolation technique which can be combined with other information to achieve better interpolating accuracy (Yue, 2011). HASM combined with ancillary information, like land use, soil and parent rock types, has been successfully used in the surface modeling of soil properties (Shi et al., 2009; Shi et al., 2011; Shi et al., 2012). However, the previous studies have not considered the statistical characters in each categorical variable type and the spatial variation degree between the neighbor samplings, which should be involved in the interpolation of the soil antibiotics.

Fluoroquinolones (FQs), which are a group of widely prescribed antibiotics and have higher concentrations and longer residence time in soils (Zhao et al., 2010), are selected as the target substances in this study. The objectives of this study are i) to develop an interpolator of HASM combined with vegetable types which are suitable for the prediction of the spatial distributions of soil antibiotics, and (ii) to assess the performance of HASM in improving the soil antibiotic interpolation compared with kriging with External Drift (KED), stratified kriging (StK), and also ordinary kriging (OK) and inverse distance weighting (IDW).

2. Materials and methods

2.1. The study area

The study area is located in the east of China (Fig. 1a). It is a typical vegetable planting area, covering 160 km^2. The main soil type is cinnamon soil, and followed by fluvo-aquic soil. According to the questionnaires investigated during the soil sampling collection, there are seven vegetable types in the study area, including cucumber–cucumber (growing cucumbers after cucumbers, 16% of the study area), tomato–tomato (growing tomato after tomato, 39%), pepper (11%), tomato-leaf (growing tomato after leaf, 13%), eggplant (6%), leaf (5%) and tomato–melon (growing tomato after melon, 10%) (Fig. 1b). The main manure applied into the soils was chicken dung, and the application quantities in most areas were from 3 kg/m^2 to 9 kg/m^2 (Li et al., 2013). The elevation decreases from the south to the north with altitude ranging from 34 m to 5 m, and the river distributions were shown in Fig. 1c. More information about the manure application categories and quantity, the spatial distributions of area of greenhouse and planting age were shown in the previous studies (Xie et al., 2012; Li et al., 2013; Li et al., 2014).

2.2. Soil samples and analysis

In order to compare the performance of different interpolation techniques, a total of 100 greenhouses were selected and in each of them the surface soil were collected following an S-shape in November 2010 (Fig. 1b). The interval of the soil samples in different greenhouses was approximately 1 km. The sampling sites were designed to cover evenly the whole area and to include different vegetable planted types, different manure types, different planting years, etc. Three types of FQs, including ciprofloxacin (CFX), enrofloxacin (EFX), and norfloxacin (NFX) were analyzed using high-performance liquid chromatography and fluorescence detection (HPLC–FD). The molecular formulas of the three antibiotics were shown as Table 1. The reagents and sample analysis methods were given in the former studies (Xie et al., 2012; Li et al., 2013; Li et al., 2014). The physical and chemical properties of soils in the study area can also be found in Li et al. (2013).

2.3. HASM

For the surface modeling of soil antibiotics, we propose a modified HASM combined with the information of vegetable type which is more suitable for the prediction of the spatial patterns of soil antibiotics. Because the detailed equations and the explanations of HASM have been published in the previous references (Shi et al., 2009; Shi et al., 2011; Shi et al., 2012), we only listed the main equations and the modified part in this study for HASM.

The matrix formulation of HASM master equations can be respectively expressed as (Yue, 2011; Yue et al., 2013, 2014, 2015),

$$\begin{cases} AF^{n+1} = c^n \\ BF^{n+1} = d^n \end{cases} \quad (1)$$

where A and B represent coefficient matrixes of the first equation and