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Composite assessment of human health risk from potentially toxic elements through multiple exposure routes: A case study in farmland in an important industrial city in East China

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ABSTRACT

Accumulation of potentially toxic elements (PTEs) in farmland soils and agricultural products is an issue of considerable concern related to food safety and human health. Assessment of composite health risks caused by exposure to PTEs through different pathways and the spatial probability of these risks could help in soil management and reduction in the corresponding human health risk. In this study, we collected 932 soil samples and corresponding samples of rice planted at the same locations in an industrial city in eastern China. The composite human health risk, including the health risks caused by Cr, Pb, Cd, Hg, As, Cu, Zn, and Ni from soil inhalation, ingestion, and dermal contact combined with the consumption of rice were assessed. Sequential Gaussian stochastic simulation and probability kriging were employed to explore the spatial pattern of the composite health risk. The results showed that 13.52%, 5.47%, 2.68%, 2.58%, 1.61%, 0.86%, and 0.21% of soil samples collected from the study area were polluted by Hg, Cd, Pb, Cu, Zn, Ni, and Cr, respectively. Furthermore, 65.02%, 20.28%, 10.94%, 4.72%, 0.75%, 0.11%, and 0.11% of rice samples were polluted by high levels of Ni, Cr, As, Cd, Pb, Hg, and Zn, respectively. Children had a higher hazard index than adults for non-carcinogenic health risks. Both children and adults had potential carcinogenic risks. The largest contributor to non-carcinogenic health risks was As, whereas Ni was the largest contributor to carcinogenic risk. Consumption of contaminated rice accounted for > 90% of the total non-carcinogenic and carcinogenic health risks, suggesting that PTEs accumulation in rice could exert harmful effects on human health. In terms of their spatial patterns, both non-carcinogenic and carcinogenic risks were associated with areas with a high density of anthropogenic activities. Residents in most areas in the study region have a high probability of experiencing significant carcinogenic and non-carcinogenic health effects caused by exposure to PTEs. Children have a higher probability of non-carcinogenic health risk than adults across the study area. The results revealed that consumption of contaminated crops poses essential potential health risks to humans. Measures should be undertaken to reduce the content of contaminated PTEs in farmland soils and rice, and children should be listed as a priority for protection from exposure to PTEs.

1. Introduction

Environmental pollution caused by potentially toxic elements (PTEs) is a global concern owing to their toxicity to humans and animals and their tendency to bio-accumulate (Duffus, 2002; Hodson, 2004; Lacarce et al., 2012; Lequy et al., 2017; Marchant et al., 2017; Schneider et al., 2016; Villanneau et al., 2011). With rapid industrialization, farmland soils and agricultural crops in China are

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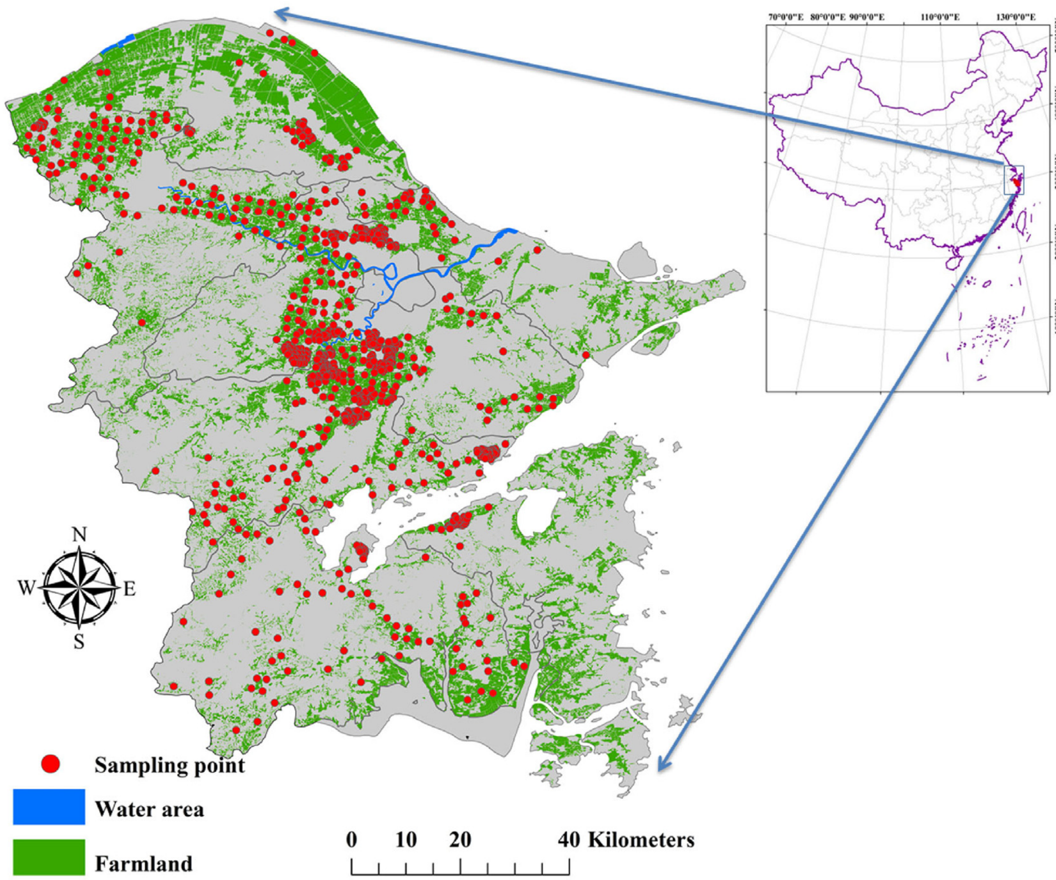

Fig. 1. Map of sampling locations in the study area.

increasingly polluted by PTEs resulting from anthropogenic activities (Hu et al., 2018; Huang et al., 2018; Liu et al., 2019; Lu et al., 2018; Xia et al., 2019). Humans are exposed to PTEs through inhalation, ingestion, dermal contact, and consumption of contaminated food leading to adverse effects on human health (Liu et al., 2013; Obiora et al., 2019; Song et al., 2018; Tepanosyan et al., 2017a; Wu et al., 2018). For example, excessive intake of Pb can lead to harmful effects on the immune, nervous, and endocrine systems (Hu et al., 2017a; Jiang et al., 2017). Bladder, kidney, and liver cancers can be caused by continuous exposure to As (Patlolla et al., 2012; Tan et al., 2016). Long-term exposure to high doses of Zn, Cr, and Cu can lead to impairments in cholesterol, fertility, and liver function (US EPA, 2000).

Rice is the fundamental and staple food in the daily diet of individuals in many countries such as China, South Korea, Thailand, Vietnam, Indonesia, Japan, and India. Accumulation of PTEs in agricultural soils is ascribed to different sources including smelting, atmospheric deposition, automobile exhaust fumes, fertilizer use, and sewage irrigation (Chen et al., 2015; Hu et al., 2017b; Jia et al., 2019; Li et al., 2018a, b, c; Marchant et al., 2010). Accumulation in soil accelerates the uptake of PTEs in crops such as rice (Mao et al., 2019; Wang et al., 2005) and then pose a threat to human health. Accumulation of PTEs in soil and agricultural products has prompted much research (Marchant et al., 2011; Rambeau et al., 2010; Saby et al., 2006; Shi et al., 2018; Zhao et al., 2018). The model recommended by the US Environmental Protection Agency is widely used to determine the risks caused by PTEs in soil (Li et al., 2018a, b, c; Tepanosyan et al., 2017b; Zhong et al., 2018). Rice is the staple food in eastern China, which is the most developed region in China with intensive industrial, business, and traffic activities that may present a threat to the ecosystem and food safety (Mao et al., 2019; Wu et al., 2019). Therefore, it would be meaningful to assess the potential health risk caused by different pathways, especially through the consumption of rice (Chang et al., 2019; Fu et al., 2013; Li et al., 2018a, b, c). In addition, the spatial pattern and probability of occurrence of potential health risks owing to exposure to PTEs is also critical for controlling pollution by PTEs and reducing related health risks. Many studies have been conducted to explore the spatial features of health risks owing to exposure to PTEs. Chen et al. (2015) analysed the spatial features of health risks of PTEs in China at a provincial scale. Hu et al. (2017c) mapped spatial distribution of potential health risks in case of exposure to PTEs in a coastal industrial region of the Yangtze River Delta with ordinary kriging. Rehman et al. (2018) analysed the spatial distribution of assessments of human health risk via toxic elements in soil and surface water ingestion in the vicinity of the Sewakht mines, district Chitral, Northern Pakistan. Sawut et al. (2018) studied the spatial distribution of assessment of health risks owing to PTEs in the vegetable production areas of Urumqi City in northwest China. However, to the best of our knowledge, there has been no study reported as yet that has analysed spatial variation in the occurrence probability of potential health risks owing to exposure to PTEs.

To address this gap, we collected 932 pairs of soil and rice samples from farmlands in an important industrial city in eastern China to assess the total non-carcinogenic and carcinogenic risks from PTEs via inhalation, soil ingestion, dermal contact, and the consumption of rice. We also produced a map of the spatial pattern of non-carcinogenic and carcinogenic risk values and probability of non-carcinogenic and carcinogenic risks to children and adults across the study area. This study aims to (i) assess the current status of PTEs pollution in the soil-rice system in the study region; (ii) analyse the potential health risks as total carcinogenic and non-carcinogenic risks through soil ingestion, inhalation, dermal contact, and the consumption of rice; (iii) evaluate the spatial distribution of total carcinogenic and non-carcinogenic risks in the study area, and (iv) map the spatial probability of non-carcinogenic and carcinogenic health risks exceeding the threshold value in children and adults.

2. Materials and methods

2.1. Study area and sampling

The study area is located in eastern China and belongs to the Yangtze River Delta, with an area of approximately 9800 km^2 (120°55′–122°16′ E and 28°51′–30°33′ N) (Fig. 1). In the past three decades, the area has been experiencing rapid and intensive industrialization and urbanization. In this study, 932 soil samples (0–0.2 m) and 932 rice samples in the same location were collected in 2013 (Fig. 1). Five unique samples were collected from five different locations within a radius of 5 m at each site and then mixed as a composite soil sample. A detailed description of the sampling method is provided in our previous work (Hu et al., 2017a) and in the Supplemental Material.

2.2. Chemical analysis

The pH of soils was measured using a pHS-3C digital pH meter (Shanghai REX Sensor Technology Co., Ltd., Shanghai, China) according to the national standard (NY/T1377-2007). The total concentration of PTEs (Cr, Pb, Cd, Hg, As, Cu, Zn, and Ni) in soil and rice samples was measured using the national recommended methods (see the Supplemental Material). Detailed information on the chemical analysis of soil and rice samples is described in our previous study (Hu et al., 2017b) and provided in the Supplemental Material.

2.3. Human health risk assessment

Target hazard quotients (HQs) and the hazard index (HI) recommended by the US Environmental Protection Agency were used to determine the potential non-carcinogenic health risks from exposure to PTE (US EPA, 1989, 1992, 2000, 2001). The HQ is prescribed as the ratio of chronic daily intake (CDI, mg/kg/day) to reference dose (RfD, mg/kg/day). The HI denotes the total potential non-carcinogenic health risk of the various PTEs and exposure routes and is defined as the sum of HQs. The CDI from certain exposure routes is calculated based on Eqs. (1)–(4), and the HI is determined using Eq. 5 (US EPA, 1991, 2000, 2002, 2011).

$$\mathrm{CDI}_{Ingestion-soil} = \frac{C_{soil} \times IR_{soil} \times EF \times ED}{BW \times AT} \times CF \tag{1}$$

$$\mathrm{CDI}_{Dermal-soil} = \frac{C_{soil} \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF \tag{2}$$

$$\mathrm{CDI}_{Inhalation} = \frac{C_{soil} \times IR_{air} \times EF \times ED}{BW \times AT} \tag{3}$$

$$\mathrm{CDI}_{Diet-rice} = \frac{C_{rice} \times IR_{rice} \times EF \times ED}{BW \times AT} \tag{4}$$

$$\mathrm{HI} = \sum HQ_{ij} = \frac{CDI_{ij}}{RfD_i} \tag{5}$$

The potential non-carcinogenic risk caused by exposure to PTEs will occur when HI > 1 and there is no risk when HI < 1 (RAIS, 2017; US EPA, 2000). The C_{soil} represents the content of PTEs in soil: mg/kg; The C_{rice} denotes the content of PTEs in rice: mg/kg; The IR_{soil} indicates ingestion rate of soil: 100 mg d^{-1}; The IR_{rice} indicates ingestion rate of soil: g d^{-1} (SBZJP, 2013); The IR_{air} denotes the inhalation rate of air (m^3 day^{-1}) (USEPA, 2011). The EF denotes exposure frequency: 350 d y^{-1} (USEPA, 2011) and ED exposure duration: 30 y (USEPA, 2011). The BW represents body weight (NHWC, 2018); The AT is the averaged exposure time for non-carcinogens: 365 × ED (USEPA, 2011); The CF is the units conversion factor: 10^{-6} kg mg^{-1} (USEPA, 2002); The SA denotes the skin surface area for soil contact (cm^2 day^{-1}) (USEPA, 2011); The AF is the soil adherence factor: 0.07 mg cm^{-2} (USEPA, 2011); and ABS indicates the dermal absorption fraction: 0.03 for As and 0.001 for other PTEs (USEPA, 2011).

Carcinogenic risk (CR) represents the lifetime probability of developing cancer caused by exposure to the contaminant and is calculated based on Eq. (6). The total carcinogenic risk (TCR) from the sum of the PTEs is determined using Eq. (7).

$$\mathrm{CR} = \mathrm{ADI}_i \times SF_i \tag{6}$$

$$\mathrm{TCR} = \sum_{j}^{m} \sum\nolimits_{i}^{n} CR_{ij} \tag{7}$$

The risk values exceed 1 × 10^{-4}, it indicates an unacceptable carcinogenic health risk; risks below 1 × 10^{-6} indicate unlikely significant carcinograceenic health effects; risks ranging from 1 × 10^{-6} to 1 × 10^{-4} imply an acceptable total risk (US EPA, 2001); SF_i is the slope factor for substance k (kg d^{-1} mg^{-1}) (USEPA, 2011). As the reference doses or conversion factors for Hg, Cu, or Zn are unavailable, we calculated carcinogenic risks for Cr, Pb, Cd, As, and Ni. The details of the parameters and corresponding values are provided in Table S1.

2.4. Spatial analysis

Sequential Gaussian stochastic simulation (SGSS) was used to assess the spatial pattern of health risk in the study area. The main advantage of SGSS over the traditional and widely used geostatistical methods such as kriging is that it can overcome the smoothing effect (Afrasiab and Delbari, 2013; Deutsch and Journel, 1992; Webster and Oliver, 2007), especially when high or extreme values are included, and it can obtain more detailed patterns of the spatial distribution of the target variable. Soil and plants are easily polluted by point and non-point pollution sources, making SGSS a suitable method for mapping the spatial distribution of health risks (Castrignano and Buttafuoco, 2004; Delbari et al., 2009; Goovaerts, 1997). More details of the SGSS are provided in the Supplemental Material. Probability kriging, which is a method similar to indicator kriging, was used to estimate spatial distribution of the probability of non-carcinogenic risks exceeding the regulated value. The detailed description of probability kriging has been reported by Goovaerts (1997) and Journel and Deutsch (1997).

3. Results and discussion

3.1. PTE characterization in soil and rice

The summary statistics, local background values, and Chinese national standard values of the content of PTEs in soil are listed in Table S2 (CEPA, 2018). The content of PTEs in the soil-rice system samples varied strongly. The mean content in soil decreased in the order of Zn > Cr > Pb > Cu > Ni > As > Hg > Cd. Compared with background values, all PTEs in the study area were enriched except As. Furthermore, 13.52%, 5.47%, 2.68%, 2.58%, 1.61%, 0.86%, and 0.21% of soil samples exceeded the national threshold values for the content of Hg, Cd, Pb, Cu, Zn, Ni, and Cr, respectively, in soil (CEPA, 2018).

Referring to the Chinese national maximum permissible limits for food, the mean concentrations of all PTEs in rice were within the safety level except for As and Ni (Hu et al., 2019; MHPRC, 2017). However, 65.02% of rice samples exceeded the threshold value for Ni, whereas the proportion of rice samples polluted by Cr, As, Cd, Pb, Hg, and Zn was 20.28%, 10.94%, 4.72%, 0.75%, 0.11%, and 0.11%, respectively. Concentration of Cu in all rice samples was at safe levels.

Our results revealed the inconsistency in the rank order of PTEs enrichment in the soil-rice system which has also been confirmed by many other studies (Hu et al., 2017b; Hu et al., 2019; Liu et al., 2013).

Therefore, we should also pay attention to the accumulation and transfer characteristics of PTEs in the soil-crop system. Many previous studies have proved that the transfer of PTEs from soil to crops was not only affected by the content of PTEs in soil but also may be affected by some other factors such as soil properties and crop types (Adriano, 2013; Hu et al., 2017b; Hu et al., 2019; Kabata-Pendias, 2000; Mao et al., 2019; Wen et al., 2018). In addition, agricultural management measures including irrigation methods, irrigation time, fertilization practices, and field management could also affect the transfer of PTEs in soil-rice systems (Hu et al., 2019; Yang et al., 2014).

3.2. Composite non-carcinogenic risk evaluation

3.2.1. Composite non-carcinogenic risk for adults and children

The non-carcinogenic risk was calculated for adults and children (Fig. S1). For children, the non-carcinogenic risk varied significantly by element, in the decreasing order of As (4.36) > Zn (0.35) ≈ Cd (0.34) ≈ Cu (0.33) > Ni (0.15) > Pb (0.10) > Hg (0.051) > Cr (0.040). The non-carcinogenic risk for adults was of the order As (0.27) > Cr (0.082) > Zn (0.022) ≈ Cd (0.021) = Cu (0.021) > Pb (0.010) = Ni (0.010) > Hg (0.0035). Arsenic was the dominant contributor to non-carcinogenic risks for both adults and children.

Fig. 2 shows the proportion of HI caused by different PTEs for children and was in the order of As (76.17%) > Zn (6.21%) > Cd (5.99%) > Cu (5.82%) > Ni (2.54%) > Pb (1.67%) > Hg (0.90%) > Cr (0.70%). The proportion of HI caused by different PTEs for adults was in the order of As (62.08%) > Cr (18.53%) > Zn (4.94%) > Cd (4.71%) > Cu (4.68%) > Pb (2.17%) ≈ Ni (2.11%) > Hg (0.79%). The predominant contributor to the HI was As. This was also confirmed by a study conducted by Mao et al. (2019). The elevated levels of As in the soil-rice system could originate from multiple sources, such as irrigation with As-contaminated water or application of As-enriched fertilizers and pesticides (Gan et al., 2019; Wang et al., 2019; Xiao et al., 2019). Therefore, priority measures related to agricultural practices should be undertaken to mitigate the accumulation of As in farmland soils and rice in the study area.

3.2.2. Composite non-carcinogenic risk by exposure pathway

The non-carcinogenic risk was calculated for various exposure pathways (Fig. S2). The non-carcinogenic risk for children followed the order of rice consumption (5.62) > soil ingestion (8.9×10^{-2}) > dermal contact (9.1×10^{-3}) > inhalation (4×10^{-4}). The HQ for adults was of the same order: rice consumption (4.14×10^{-1}) > soil ingestion (2.18×10^{-2}) > dermal contact (6.1×10^{-3}) > inhalation (3×10^{-4}).

The HI for children ranged from 0.21 to 102.49 (mean: 5.72) and that for adults varied from 0.03 to 6.34 (mean: 0.44). The mean value of the HI was significantly higher for children and exceeded the threshold value of unity, indicating that PTE pollution is likely to exert adverse effects on children in the study area. Some of the HI for adults also exceeded the threshold value.

The exposure pathway of non-carcinogenic risk for children was of the order rice consumption (93.61%) > soil ingestion (4.93%) > dermal contact (1.38%) > inhalation (0.08%) (Fig. 3), whereas the non-carcinogenic risk for adults was of the order rice consumption (98.28%) > soil ingestion (1.56%) > dermal contact (0.16%) > inhalation (0.01%) (Fig. 3). Most non-carcinogenic risks resulted from consumption of rice, followed by soil ingestion and dermal contact. This is consistent with the results reported by Cao et al. (2010) that potential health risks from exposure to self-planted rice were of major concern. Inhalation of contaminated air made only a negligible contribution to non-carcinogenic risks.

3.3. Composite carcinogenic risk evaluation

3.3.1. Composite carcinogenic risk for adults and children

The carcinogenic risks posed by PTEs are listed in Fig. S3. Carcinogenic risk in children was of the order Ni (3.87×10^{-4}) > Cd (1.63×10^{-4}) > As (1.53×10^{-4}) > Cr (1.40×10^{-4}) > Pb (2.47×10^{-7}). Risks from Ni, Cd, As, and Cr (but not Pb) were higher than the threshold value of 1×10^{-4}. Carcinogenic risk in adults was of the order Ni (1.25×10^{-4}) > Cd (4.97×10^{-5}) > As (4.80×10^{-5}) ≈ Cr (4.78×10^{-5}) > Pb (1.68×10^{-7}). The risk was high for Ni; medium for Cd, As, and Cr; and low for Pb. Overall, the carcinogenic risk was higher for children. These findings are consistent with some previous studies by Hu et al. (2017b), Jiang et al. (2017), and Wu et al. (2018).

The contribution of Ni, Cd, As, Cr, and Pb to the target carcinogenic risks for children was 45.85%, 19.37%, 18.16%, 16.61%, and 0.03%, respectively (Fig. 4). The survey conducted by Cao et al. (2010) also found that Cd and As occupy a large proportion of the potential carcinogenic risks. For adults, the contribution of different PTEs had a similar order: Ni (46.23%) > Cd (18.33%) > As (17.72%) > Cr (17.63%) > Pb (0.06%). The differences between element-specific carcinogenic and non-carcinogenic risks are likely the reflection of the differing mechanisms of toxicity from these elements (Wu et al., 2018).

3.3.2. Composite carcinogenic risk by exposure pathway

The carcinogenic risks for the various exposure routes are shown in Fig. S4. For children, carcinogenic risk (8.33×10^{-4}) was mainly attributable to consumption of rice, followed by soil ingestion (1.07×10^{-5}), dermal contact (7.48×10^{-7}), and inhalation (9.30×10^{-8}). Carcinogenic risk for adults followed the same order of rice consumption (2.55×10^{-4}) > soil ingestion (1.32×10^{-5}) > dermal contact (2.50×10^{-6}) > inhalation (2.21×10^{-7}). The major portion of carcinogenic risks in the study area was attributable to consumption of rice, consistent with the findings of Liu et al. (2013). In contrast, inhalation of PTE-contaminated air accounted for only a negligible proportion of the carcinogenic risk. Overall, both children and adults in the study area had notable

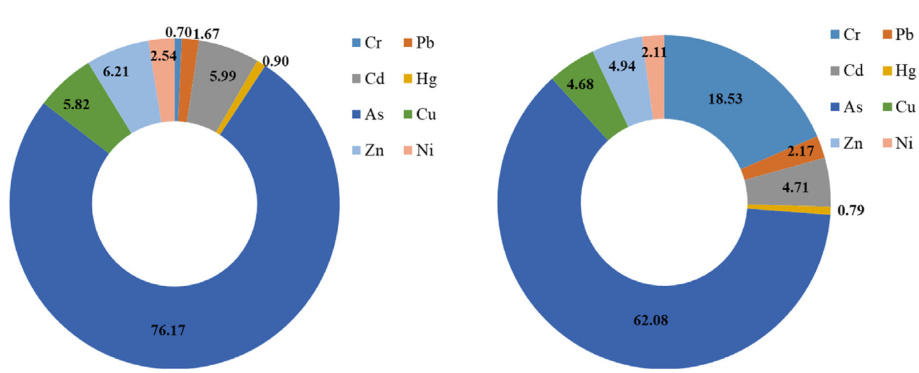

Fig. 2. Proportion of HQ (target hazard quotients) for children (left) and adult (right) caused by different PTEs (%).

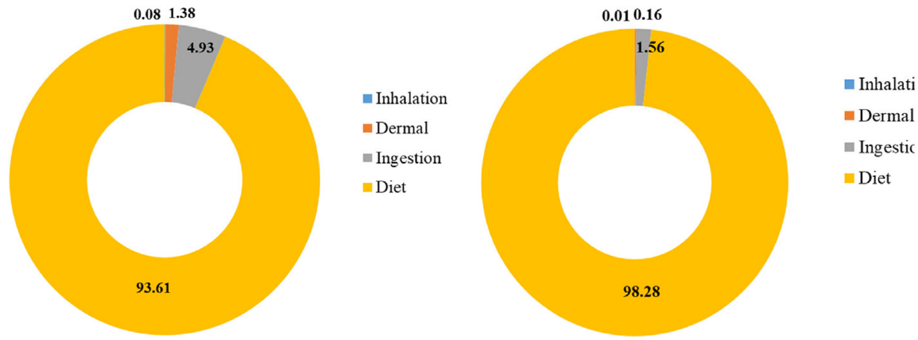

Fig. 3. Proportion of HQ (target hazard quotients) for children (left) and adult (right) caused by different pathways (%).

potential carcinogenic risks from metal and As pollution.

Fig. 5 shows the total carcinogenic risks by exposure route. The total carcinogenic risk for children varied by exposure route in the order of rice consumption (98.63%) > soil ingestion (1.27%) > dermal contact (0.09%) > inhalation (0.01%). The total carcinogenic risk for adults was of the same order: rice consumption (94.14%) > soil ingestion (4.86%) > dermal contact (0.92%) > inhalation (0.08%). Consumption of rice was the dominant contributor to the health risks caused by exposure to PTEs; however, most previous studies did not take dietary rice into consideration.

According to the results of this study, consumption of rice was the dominant exposure route for both non-carcinogenic and carcinogenic health risks in both children and adults. Furthermore, as rice is the staple food in eastern China, this result highlights the vital importance of ensuring food security and reducing transfer of PTE from soil to rice. Measures such as the application of lime in acidic soil is an effective method of adjusting soil pH and lowering the bioavailability of PTEs to reduce accumulation of PTEs in rice and other crops (Chen et al., 2016). In addition, severely polluted farmland or agricultural land could be transferred to other types of land uses such as forest, grassland, or industry. Furthermore, a spatial buffer around PTEs pollution sources like mining areas, industrial land, and large thermal power plants could also be created to protect people from exposure to PTE (Hu et al., 2017c).

3.4. Spatial pattern of composite health risk

3.4.1. Spatial distribution of composite non-carcinogenic risk

The non-carcinogenic health risks for children and adults had a similar spatial trend, although the risk for children was significantly higher (Fig. 6). This is consistent with our previous findings (Hu et al., 2017c). We considered only rice for the assessment of dietary intake of PTEs in this study, and the actual HI value may be higher with concurrent consumption of vegetables, meat, and other food. The adult HI in the study area was lower than the threshold value, although the HI in specific sampling sites was close to the value one. The non-carcinogenic risk hotspot was located in the central region of the study area for both children and adults, which contains industrial enterprises and mines and has high traffic movement (Shao et al., 2018; Wang, 2018). There were also several discrete hotspots in the north-west region of the study area, which are most likely owing to the presence of industrial and mining enterprises (Huang et al., 2018; Shao et al., 2018).

3.4.2. Spatial distribution of composite carcinogenic risk

The spatial pattern of carcinogenic health risk for children and adults is shown in Fig. 7. The spatial pattern was almost identical for the total carcinogenic risks for children and adults, and the area of highest risk for both groups was situated in the central part of the research region, coinciding with high HI values and confirming the negative effects of anthropogenic activities. The total carcinogenic risk for children was remarkably higher than the risk for adults. Hotspots of total carcinogenic risk were observed in the south-western part of the study area, where the calculated values exceeded the acceptable total risk of 1×10^{-4} (USEPA, 2010), presumably owing to traffic emissions, industrial waste, mining, and smelting (Li et al., 2019; Shisia, 2013). The area around the hotspots within the regulatory range (1×10^{-6} to 1×10^{-4}) had acceptable target carcinogenic risks (USEPA, 2010). The carcinogenic risk for children and adults in the coastal area and in the northern and southern parts was lower than the regulatory threshold. These regions had relatively lower soil pH, as the soil parent material in this area is mainly alluvial river deposits, which could bind to PTEs and disrupt uptake by plants (Chen et al., 2016; Wang et al., 2016). Our findings indicate that PTEs accumulation in rice could pose serious health effects given that rice as the staple food is consumed in most places of southern China and therefore, measures should be immediately undertaken to control and mitigate PTEs pollution to reduce health risks.

Overall, the areas with a high value of carcinogenic risk and non-carcinogenic risk overlapped considerably with areas having high population density and human activities such as industrial, commercial, and mining activities. This demonstrates that the high value of potential

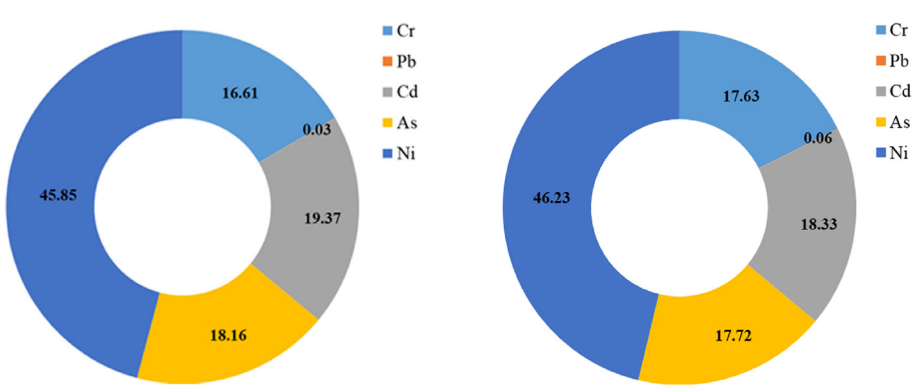

Fig. 4. Proportion of TCR (the total carcinogenic risk) for children (left) and adult (right) caused by different pathways (%).

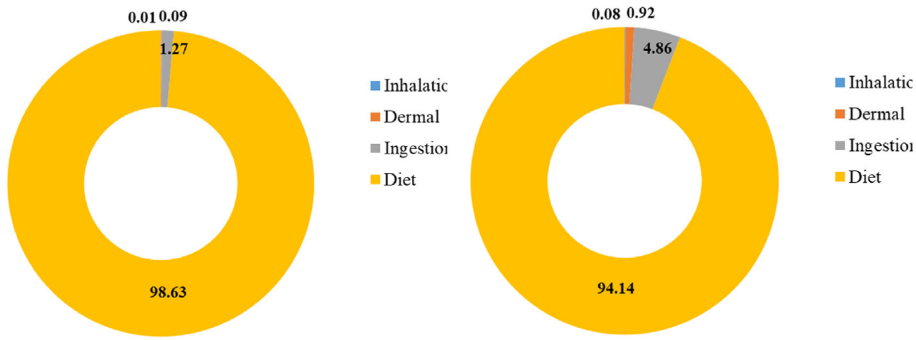

Fig. 5. Proportion of TCR (the total carcinogenic risk) for children (left) and adult (right) caused by different pathways (%).

carcinogenic risk and non-carcinogenic risk was probably related to anthropogenic activities. More information on industrial, traffic, and mining activities is needed to confirm this through further research.

3.4.3. Spatial probability distribution of composite non-carcinogenic risk

The use of probability kriging allows the mapping of the probability of certain variable values exceeding a cut-off value. These probability maps are very useful for decision-making, as they are easy to interpret (Antunes and Albuquerque, 2013). In this study, the cut-off value for non-carcinogenic risk (HI) was set as one. When the value is larger than 1, it indicates the potential non-carcinogenic risk caused by exposure to PTEs. The probability maps for HI $>$ 1 produced by probability kriging are displayed in Fig. 8. The probability for HI $>$ 1 for children and adults showed clearly different spatial patterns. For children, the probability of exceeding the threshold level was very high across farmland soils in the central and southern regions of the study area. This confirmed the results shown in Fig. 6 that people living in these areas were facing considerable non-carcinogenic risks caused by exposure to PTEs. The northern part represents a low probability of exceeding the threshold value. For adults, the highest probability was located in the central and western regions while the low probability areas were distributed in the northern and southern regions of the study area. Overall, the probability for HI $>$ 1 for children was higher than that for adults across the study area. This indicates the urgency in implementing measures to protect children from exposures to PTEs.

3.4.4. Spatial probability distribution of occurrence of composite carcinogenic risk

The probability for TCR $> 10^{-4}$ for children and adults showed a similar spatial trend (Fig. 9). The high probability area was located across the study area while low probability was mainly distributed in the northern coastal area. This revealed that the children and adults living in most areas in the study area were experiencing significant carcinogenic health effects owing to exposure to PTEs. This area is close to the Hangzhou Bay. The soil mainly comprises river alluvial deposits, and the land use type is mainly agricultural land containing some wastes produced by industrial activities (Li et al., 2019; Shao et al., 2018).

4. Conclusion

Pollution from potentially toxic elements in soil and crops including rice was widely observed in China, which poses a great threat to local residents. In this study, a human health risk assessment model issued by the US Environmental Protection Agency was used to determine the health risk posed by exposure to PTEs, and geostatistical models were used to explore spatial patterns and spatial probability of health risks in farmland in an important industrial city located in eastern China. The results showed that the proportion of soil samples polluted by Hg, Cd, Pb, Cu, Zn, Ni, and Cr is 13.52%, 5.47%, 2.68%, 2.58%, 1.61%, 0.86%, and 0.21%, respectively. The mean content of all PTEs in the rice samples were at safe levels except for As and Ni. However, 65.02% of rice samples exceeded the threshold values for Ni and 20.28% of rice

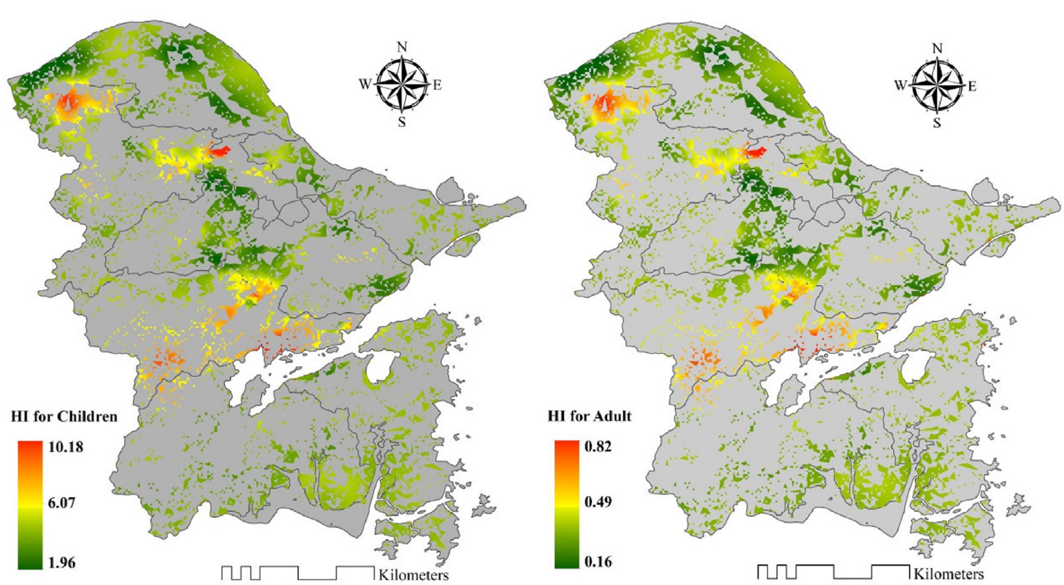

Fig. 6. Spatial distribution of non-carcinogenic healthy risk (HI) for children (left) and adult (right) in farm land in our study area.

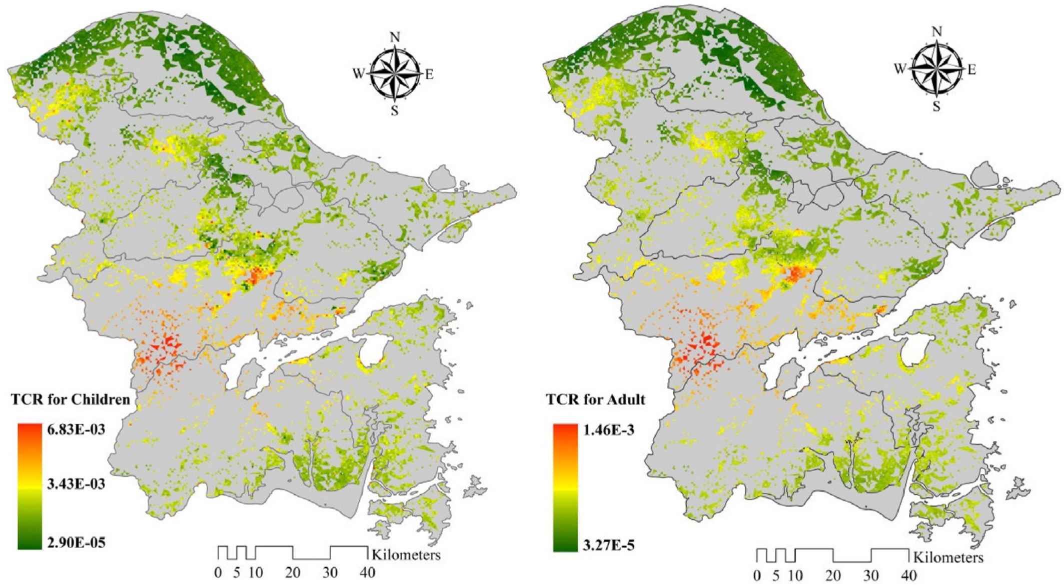

Fig. 7. Spatial distribution of total carcinogenic healthy risk (TCR) for children (left) and adult (right) in farm land in our study area.

samples exceeded the threshold value for Cr.

Regarding potential health risks caused by exposure to PTEs, children in the study area are experiencing potential non-carcinogenic risks owing to exposure to PTEs. In addition, both children and adults are experiencing potential carcinogenic health risks caused by PTEs pollution. The major cause of non-carcinogenic risk is As, whereas Ni is the major cause of carcinogenic risk. Consumption of contaminated rice accounted for > 90% of the total non-carcinogenic and carcinogenic health risks.

Overall, residents of the study area have substantive health risks from exposure to PTEs through multiple routes. Citizens, especially children, in most parts of the study region have a high probability of experiencing significant carcinogenic and non-carcinogenic health effects caused by exposure to PTEs. Measures should be undertaken to remediate the soil and improve crop safety and systematic studies are required to define the sources of PTEs in soil and control future contamination. Moreover, in present work we merely measured the total content PTEs, and probably not all PTEs total concentration is accounted to the bioavailable fraction and this would lead to certain bias on the results. Our conclusions could provide inputs for regulating the health risk owing to exposure to PTEs in other regions that have similar natural and socio-economic environments.

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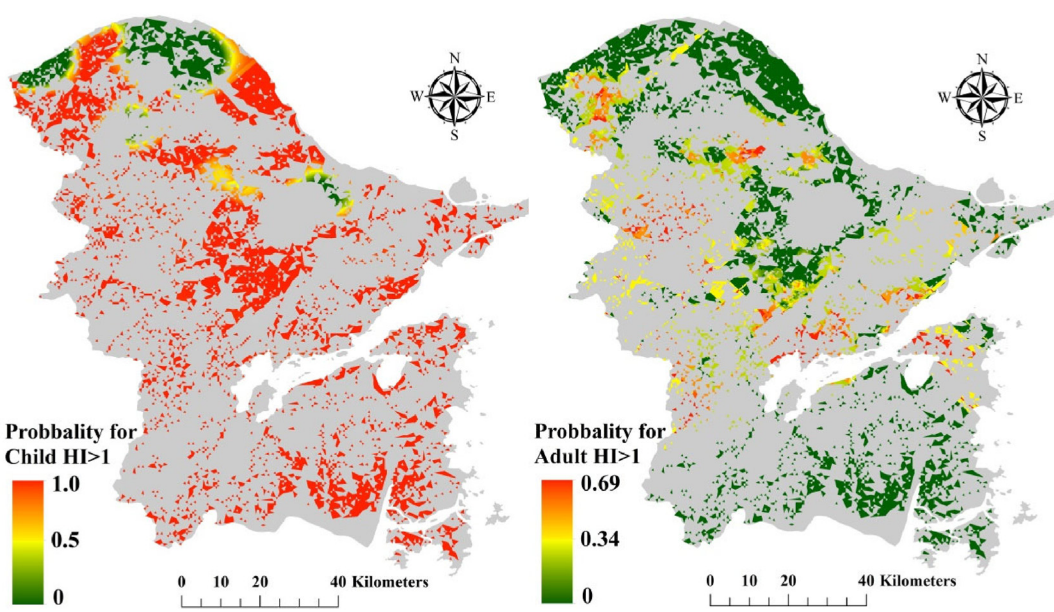

Fig. 8. Maps of the probability that HI (the hazard index) for Child (left) and Adult (right) exceeds the limit value (1.0).

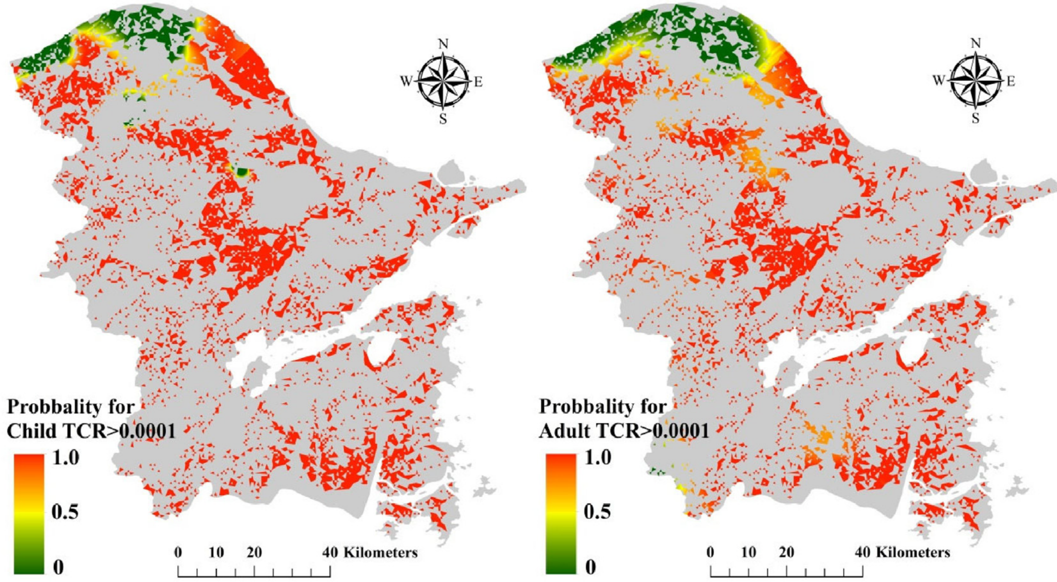

Fig. 9. Maps of the probability that TCR (total carcinogenic healthy risk) for Child (left) and Adult (right) exceeds the limit value (1 × 10–6).

Declaration of competing interest

The authors have declared that no competing interests exist. The founding sponsors had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript or in the decision to publish the results.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.gexplo.2019.106443.

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