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Reduction in the exposure risk of farmer from e-waste recycling site following environmental policy adjustment: A regional scale view of PAHs in paddy fields



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ABSTRACT

Farmland contamination by polycyclic aromatic hydrocarbons (PAHs) has drawn increasing attention across China with enhanced regulations and environmental policies proposed by government to protect soil environment safety. As the informal electronic waste (e-waste) dismantling activities were forbidden under recent environmental regulation, this study compared levels, compositions, spatial distributions, human health risks of PAHs in paddy soil within the vicinity of an e-waste recycling area in southeastern China, with 129 and 150 soil samples collected in 2011 and 2016, respectively. The soil contamination was dominated with high molecular weight PAHs. The mean concentration of EPA 16 PAHs decreased from $590.4 \pm 337.2 \mu\text{g kg}^{-1}$ in 2011 to $407.3 \pm 232.2 \mu\text{g kg}^{-1}$ in 2016. Distribution maps of soil PAHs concentration displayed the temporal change in spatial. Principal component analysis together with diagnostic ratios revealed the combustion of biomass and coal in industrial and unregulated e-waste dismantling were the main sources of PAHs in the study area. Both deterministic and probabilistic assessments demonstrated reduced exposure risk for farmers from 2011 to 2016. Sensitivity analysis revealed that exposure frequency (*EF*) is the most influential parameter for the total variance in the risk assessment model. This study implied that the more stringent environmental policy and regulation can lead reductions in soil contamination with PAHs.

1. Introduction

The quality of farmland soil is of vital importance because it acts as the basic resource that providing life-supporting services of food production as well as habitats for wildlife and humans. China's per capita arable land area is less than one third of the world average (UNESCO, 2012). This sharp contradiction between the large population and limited arable land means farmland soil quality is an on-going concern in China. Severe soil pollution caused by various types of pollutants affects 19.4% of Chinese farmland with PAHs one of the main contaminants (MEP and MLR, 2014; Sun et al., 2018). In response, the government has drafted a series of environmental policies on soil pollution prevention and control over the last decade (Zhang et al., 2012; SCC, 2013; Wang et al., 2016). For example, six pioneering areas were listed in "Action Plan for Soil Pollution Prevention" (SCC, 2016), are taking the leading role in soil pollution remediation and control. In addition, Soil Pollution Control Act was passed by Chinese government, which provides an important legal basis for soil contamination

prevention and control work (SCC, 2018). However, how environmental policy influence soil PAHs remain unclear.

PAHs are organic compounds made up of two or more aromatic rings which exist widely in the environment. Many PAHs are associated with mutagenic, carcinogenic and teratogenic effects, and they are labeled as priority control pollutants by USEPA (Cao et al., 2016; Islam et al., 2017). Paddy field is one type of anthrosol which is greatly sensitive to agricultural activities and environmental changes, it can also serve as a main reservoir and sink for pollutants (Yang et al., 2010). PAHs tend to accumulate in paddy soils not only due to their high hydrophobicity, and difficulty to be degraded (Wang et al., 2010; Daso et al., 2016), but also owing to their relatively high organic matter content of these soils.

Health risk assessment is a process of estimating the possibility of deleterious effects of human beings who may be exposed to contaminants in environment media (USEPA, 2016). Risk-based approaches link the site contamination with the adverse probability of human health, which is one of the most valuable tools to provide

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guidance for pollutant management and remediation practices (Umeh et al., 2017). Traditional deterministic risk assessment was conducted by the most likely (usually the mean or the maximum) value of exposure parameters and single point pollutant contents. However, due to the range of pollutant concentrations, uncertainty of exposure parameters and the inherent variability of the risk assessment model, incorporating statistical distributions of input parameters to conduct probabilistic health risk has been a growing trend (Augustsson et al., 2017; Ginsberg and Belleggia, 2017).

Previous studies were mostly focused on the environmental occurrence, distribution patterns, and related exposure risks of PAHs in the contamination sites at a fixed point in time (Huang et al., 2014; Leung et al., 2015; Devi et al., 2016; Y.J. Wang et al., 2017). While changes in the temporal or spatial distribution, sources as well as exposure health risk of PAHs at a specific region have not been adequately addressed with reference to environmental policy implementation. The goals of this study are: (1) to elucidate the alterations of pollution characteristics of PAHs in paddy soil during after the regulation process; (2) to assess the associated human health risks exposed from soil PAHs by both deterministic and probabilistic risk assessment methods. The results aim to provide a reliable reference for zonal management and risk control of farmland PAHs contamination.

2. Methods and materials

2.1. Description of study area and soil sample collection

Wenling, a prominent rice producing area in southeast of Zhejiang Province of China; it was described in detail in a previous study of Liu et al. (2018). It has been an e-waste disposal center in Taizhou since 1990s. There are abundant contaminants present around the site which are not limited to those released from e-waste, such as heavy metals (HMs) (Zhao et al., 2010), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) (Fu et al., 2011), but also those generated during the unregulated dismantling activities, such as PAHs (Tang et al., 2010). Prior to regulation small family-sized e-waste workshops, open burning and informal hand dismantling activities were prevalent. These unregulated salvaging operations have caused severe soil contamination and high potential risks to inhabitants (Shen et al., 2009; Fu et al., 2013; Lu et al., 2016). The government took action and enforced strict environmental regulation policy in Wenling, especially towards e-waste since the “12th Five-Year Plan (12th FYP, 2011–2015)” (Zhang et al., 2012; The State Council, 2012; SCC, 2013). With increasing strengthened environmental regulations and laws, many private open burning e-waste sites were abandoned, family-sized workshops cease operating and smelting, electroplating and dismantling parks for e-waste were central coordinated. For example, > 9500 small-sized dismantling workshops in Luqiao district were closed down by authority (Fu et al., 2012). Also, many e-waste dismantling enterprises have been transferred to other countries, such as Laos, Cambodia and Vietnam.

Soil samples (0–15 cm in depth) were gathered from rice paddies across different administrative regions in October of 2011 (129) and 2016 (150) (Fig. 1). At least three subsamples were collected within a 10 m radius circular area and then bulked to provide a composite sample. The sampling sites of 2016 followed the history points with increased density in the highly polluted area. All the sampling method and pretreatment of soil samples and chemical analysis were same in both years. Twigs and pebbles were first clear away then soil samples were fully freeze-dried to constantly weight for > 72 h. Finally mixed homogenize fully after passed through a 100 mesh sieve.

2.2. Chemical analysis

2.2.1. Sample extraction and cleanup

Five grams (dry weight) of soil sample was extracted three times

with 10 mL of 1:1 acetone/n-hexane (v/v) using ultrasonic agitation for 30 min. The extraction was centrifuged for 10 min at 3000 rpm, then the supernatant was carefully gathered and rotary evaporated to nearly 1 mL before cleanup. The concentrated extracts were cleaned using Supelco 6 mL C18 SPE cartridge. Prior to elution, the C18 column was activated with 3 mL n-hexane. Then elution extract by 8 mL of 7:3 n-hexane/dichloromethane solvent (v/v) as eluent. After that eluent was finally dried under gentle steam of pure nitrogen and resuspended to 1 mL in n-hexane. Detailed procedures were described in He et al. (2017).

2.2.2. Instrumental analysis

The profiles and concentrations of 16 PAH congeners were analyzed by gas chromatography (GC) coupled with ion-trap mass spectrometer (MS) using a HP-5MS capillary column (0.25 mm × 30 m × 0.25 μm, Agilent 19091S-433). Helium gas was used for the carrier gas with flow rate of 1 mL/min. The program of the oven temperature was set as: began at 60 °C (lasted for 2 min), went up to 120 °C at 30 °C/min and sustained for 1 min, then ramped to 280 °C at the speed of 5 °C/min and kept for 13 min.

The ionization was accomplished in the 70 eV electron shock, and the data were obtained in the mode of selected ion monitoring (SIM). Identification of PAH congeners was depended on the comparison of retention time between selection ions of samples and the standard solution. The 16 PAHs identified by USEPA were detected under SIM mode by the target ion and two reference ions. The specific target and reference ions of 16 PAHs are listed in Table S1 in Supporting Information (SI). Quantification of each target compound was made by external standard method (at least five-point calibration curve).

2.2.3. Quality control and assurance

The analytical processes were performed under strictly quality control measures. All solvents used (n-hexane, acetone and dichloromethane) were HPLC grade (Sigma-Aldrich, Shanghai, China). A procedural and spiked blank were run in every batch of 12 samples to detect potential sample contamination. Results showed that no target compounds were found in the laboratory blanks. The PAHs concentrations variations in duplicates were no > 15%. Daily calibration checks were undertaken with the PAHs reference standard (Supelco Inc., USA). Additionally, 2-fluoro-1,1'-biphenyl and phenanthrene-d10 were spiked prior to evaluate the effectiveness of cleanup and extraction. The surrogate standards recoveries for determination of PAHs were within the ranges of 78.0–108.0% and 82.0–118.0%, respectively. The average recovery rates of the spiked standards of individual PAHs were 70.2–110.8%. The correlation coefficients (R^2) for standard curves were observed in the range of 0.9990–0.9999. The detection limits of PAHs were in the range of 0.022–0.470 μg kg⁻¹. Details about the detection limit for each component of 16 PAHs are listed in Table S2.

2.3. Human health risk assessment

2.3.1. Risk characterization and assessment

We employed the incremental lifetime cancer risk (ILCR) model to assess the carcinogenic risk from exposure to soil-borne PAHs (USEPA, 1991). Adult farmers who spent most of their time in paddy fields have the greatest exposure to contamination and were therefore considered the most sensitive population. ILCR (unitless) mainly considers the following assumption pathways which adult farmers could be exposed to soil PAHs in this study: (a) accidental oral ingestion of soil, (b) dermal contact, and (c) inhalation intake of fugitive soil particle. The corresponding ILCR calculations are as follows (Eqs. (1)–(3)). Total risk (ILCR_T) is the total of individual risk from each exposure pathway.

$$ILCR_{Oral} = \frac{C_s * CSF_{Ing} * IR_{Ing} * \sqrt[3]{\frac{BW}{70}} * EF * ED}{AT * BW * 10^6} \quad (1)$$

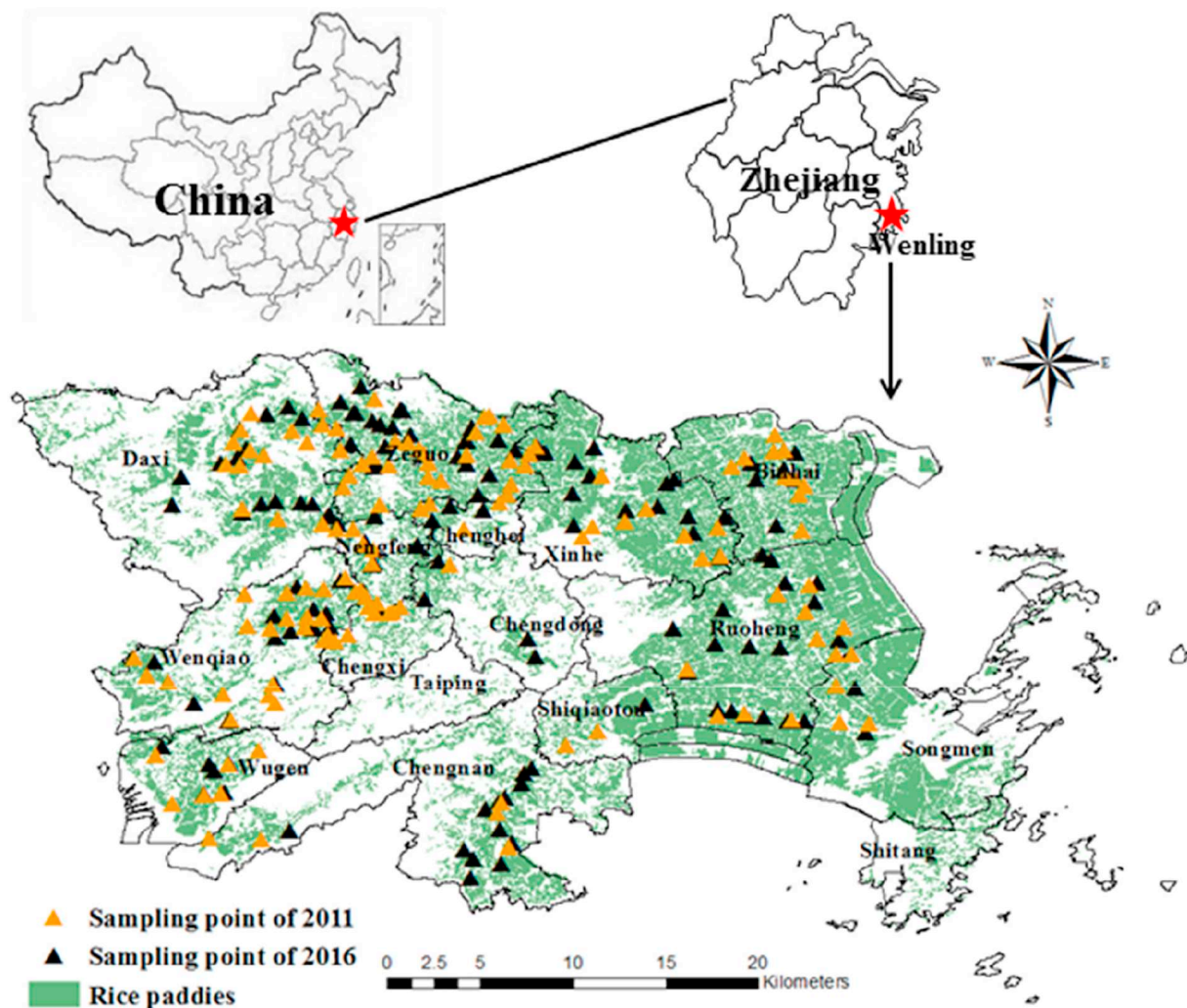


Fig. 1. Location of the study area and distribution of sampling points.

$$ILCR_{Derm} = \frac{C_s * CSF_{Derm} * \sqrt[3]{\frac{BW}{70}} * EF * ED * SA * AF * ABS}{AT * BW * 10^6} \quad (2)$$

$$ILCR_{Inh} = \frac{C_s * CSF_{Inh} * IR_{Inh} * \sqrt[3]{\frac{BW}{70}} * EF * ED}{AT * PEF * BW} \quad (3)$$

$$ILCR_s = ILCR_{Ing} + ILCR_{Derm} + ILCR_{Inh} \quad (4)$$

where $ILCR_{Oral}$, $ILCR_{Derm}$ and $ILCR_{Inh}$ are the risks via oral, dermal contact and inhalation pathways; C_s is the total concentration of transformed individual PAHs by the toxic equivalency factor (TEF) ($\mu\text{g kg}^{-1}$) of BaP. The corresponding TEF of 16 PAHs can be found in Table S3. CSF_{Inh} , CSF_{Ing} and CSF_{Derm} are the carcinogenic slope factor for inhalation, ingestion and dermal contact respectively ($\text{mg kg}^{-1} \text{d}^{-1}$) $^{-1}$; IR_{Ing} is the soil particle intake rate (mg day^{-1}); IR_{Inh} is the inhalation rate of soil ($\text{m}^3 \text{day}^{-1}$); AT represents the average life span (day); PEF is the particle emission factor ($\text{m}^3 \text{kg}^{-1}$); BW means the body weight (kg); ED is the exposure duration (year); EF is the exposure frequency (day year^{-1}); SA is the exposure dermal surface area (cm^2); AF is the dermal adherence factor ($\text{mg cm}^{-2} \text{h}^{-1}$); ABS is the dermal adsorption fraction.

For deterministic risk assessment, concentrations of C_s in each sampling sites were used, and values of each other associated parameter were listed in Table S4.

2.3.2. Monte Carlo (MC) simulation and sensitivity analyses

Probabilistic risk assessment was conducted by Monte Carlo simulation with 10,000 iterations due to the uncertainty of parameters and variety of PAHs concentration range. Concentrations of C_s were chosen from the measured sampling sites of 2011 and 2016 for each run and the extreme values were removed to fit a lognormal distribution. These parameters (BW , EF , ED , IR_{Inh} , IR_{Ing} and SA) were probabilistically treated with the defined distribution listed in Table S5. These parameters (CSF_{Ing} , CSF_{Derm} , CSF_{Inh} , AT , AF , ABS and PEF) were assumed invariable with determinate values (Table S6). Sensitivity analyses were also made to compare the contribution of each input parameters to output results during the MC simulations.

2.4. Data analysis tools

Basic statistic, principal component analysis (PCA) and spearman correlation were made by SPSS 16.0 (SPSS Inc., IL, USA), figures were graphed in Origin 2018 (Origin Lab, Northampton, MA). The spatial distributions maps of PAHs concentration were made by Inverse Distance Weight (IDW) interpolation method with ArcGIS 10.2.2 (ESRI, Redlands, USA). Monte Carlo simulation and sensitivity analysis were achieved by Crystal Ball 11.1.2 software (Oracle, Inc., USA).

3. Results and discussion

3.1. Temporal characteristics of concentration, component and spatial distribution pattern of PAHs in paddy field in 2011 and 2016

The 16 PAHs were frequently detected in paddy fields with different detection frequency (Fig. S1). Table 1 shows the statistics of measured concentrations for each PAH and various groups. The soil was both dominated by high molecular weight PAHs in 2011 and 2016. Compared with other sites, the PAHs concentration found in present study ($\Sigma_{16}\text{PAHs}$ -2011, median $479.6 \mu\text{g kg}^{-1}$, mean $590.4 \mu\text{g kg}^{-1}$; $\Sigma_{16}\text{PAHs}$ -2016, median $355.6 \mu\text{g kg}^{-1}$, mean $407.3 \mu\text{g kg}^{-1}$) was higher than those in farmland soils in eastern China (median $134 \mu\text{g kg}^{-1}$, mean $158 \mu\text{g kg}^{-1}$) (Sun et al., 2017a), Yangtze River Delta (YRD) region (median $166 \mu\text{g kg}^{-1}$, mean $267 \mu\text{g kg}^{-1}$) (Cai et al., 2017) and Tibetan Plateau (mean $60 \mu\text{g kg}^{-1}$) (Wang et al., 2014). While slightly lower than the mean content ($666 \mu\text{g kg}^{-1}$) of PAHs in agricultural soil around an industrial site in Shanghai (Jiang et al., 2011) and another industrial area (mean $471 \mu\text{g kg}^{-1}$) near the YRD region (J. Wang et al., 2017). The threshold concentration of soil PAHs is not yet regulated in China, therefore some relevant guidelines in other countries were employed to assess the pollution extent. The concentrations of Σ_{16} PAHs in all of the sampled sites were above $50 \mu\text{g kg}^{-1}$ of the Dutch government standard (Agarwal, 2009). Additionally, the concentration of 7 carcinogenic PAHs exceeded the Swedish soil limit ($300 \mu\text{g kg}^{-1}$) for sensitive land use in 2011 (mean $324.8 \mu\text{g kg}^{-1}$) and were slightly below in 2016 (mean $262.8 \mu\text{g kg}^{-1}$). The paddy soils in Wenling were regarded as polluted by PAHs.

A contamination classification based on the total concentrations of 16 PAHs provided by Maliszewska-Kordybach (1996) suggests 4 categories: non-contaminated (clean) soil ($< 200 \mu\text{g kg}^{-1}$), weakly contaminated soil ($200\text{--}600 \mu\text{g kg}^{-1}$), moderately contaminated soil ($600\text{--}1000 \mu\text{g kg}^{-1}$) and heavily contaminated soil ($> 1000 \mu\text{g kg}^{-1}$). Based on the criteria, 14.4% of the samples collected in 2011 were categorized as heavily contaminated and 23.5% as moderately contaminated, these were predominantly in the towns of Zeguo and Chengbei in northwestern area (Fig. 2a), where many small family-sized e-waste recycling workshops were located previously (Wang et al., 2011). In these workshops, plastic waste was combusted daily with

emissions discharged directly into atmosphere, with the remaining ash disposed to soil. However, in Binhai town which is far away from the open burning sites were weakly contaminated ($511.5 \mu\text{g kg}^{-1}$), it could be inferred that the contamination of remote, less human influenced sites were probably resulted from the long-range atmospheric transport (Bakker et al., 2001).

For the soil samples in 2016, 4.0% and 12.0% were categorized as heavily contaminated and moderately contaminated sites, respectively; these were located in the south and southwest part of Wenling (Fig. 2b). The percentage of non-contaminated soil samples has increased to 14.0% compared with 2.3% in 2011. These “hot-spots” and temporal change contrast with those of heavy metals which remained constant in our previous study (He et al., 2019), these may indicate their different characteristics of heavy metals and PAHs. The correlation results indicate that soil organic matter (SOM) concentration plays partial role in determining the retention of PAHs ($r^2 = 0.39$, $p < 0.05$) (Tables S8 and S9), which is consistent with other studies (Sun et al., 2017a; Cai et al., 2017). Besides, the emission sources and deposition processes were also the critical factors determining the geographical distribution pattern of contaminants in soil (Wu et al., 2019).

3.2. Potential sources of PAHs

Identification of the potential sources is essential for the understanding of PAHs distribution and prevention. Diagnostic ratios of PAHs are often used to differentiate between petroleum and combustion sources, which have been widely adopted to help identify the potential sources of PAHs (Yunker et al., 2002; Alves et al., 2016; Peng et al., 2016).

In our study, for the samples collected in 2011, the ratios of FLU/(PYR + FLU) ranged between 0.29 and 0.83, with the average value of 0.57. Furthermore, values of IND/(BghiP + IND) varied from 0.13 to 0.84, with mean value of 0.64. With regards to samples of 2016, the ratios of FLU/(PYR + FLU) varied between 0.29 and 0.69, with the average value of 0.53, values of IND/(BghiP + IND) were between 0.31 and 0.70 with mean value of 0.55. As shown in Fig. 3, $> 80\%$ of the ratios of IND/(BghiP + IND) and FLU/(PYR + FLU) of soil samples were all above 0.5, suggesting that the pollution of soil PAHs in Wenling came primarily from mixed sources and dominated by biomass and coal

Table 1
Concentrations of PAHs in paddy fields from Wenling in 2011 and 2016 ($\mu\text{g kg}^{-1}$).

Compounds	Aromatic ring	Guideline ^a	2011 (n = 129)				2016 (n = 150)			
			Range	Mean	Median	SD	Range	Mean	Median	SD
NAP	2	40	0.2–113.4	15.5	9.4	19.6	3.8–158.5	36.7	32.1	22.5
ACY	3	–	1.6–12.7	2.8	2.5	1.5	N.D–114.8	17.4	8.5	28.4
ACE	3	–	0.9–13.8	3.2	2.6	1.9	N.D–15.0	7.3	7.4	3.2
FL	3	–	1.4–51.0	10.1	8.2	6.9	4.9–26.0	13.0	12.2	4.2
PHE	3	40	4.0–240.0	64.5	50.3	44.7	17.3–275.4	63.7	52.6	43.3
ANT	3	40	2.1–103.1	7.5	5.2	11.7	4.4–29.7	11.7	11.0	4.4
FLU	4	50	4.5–251.1	53.9	44.4	37.6	10.7–128.8	42.4	38.4	20.2
PYR	4	–	9.4–272.1	41.0	33.1	32.1	9.6–153.8	37.0	33.8	18.3
BaA	4	5.0	1.3–114.4	23.6	18.0	19.5	1.2–105.2	32.3	24.6	21.1
CHR	4	50	N.D–220.4	35.8	29.1	28.5	N.D–131.5	37.9	29.0	23.4
BbF	5	–	3.1–366.6	61.1	44.5	54.7	5.4–183.8	46.8	33.3	31.7
BkF	5	5.0	N.D–144.7	6.0	N.D	16.4	N.D–218.3	67.1	58.5	43.7
BaP	5	0.5	N.D–98.8	20.4	15.9	15.6	12.1–142.1	48.3	31.4	34.0
IND	6	5.0	8.1–460.0	128.5	90.3	111.8	5.8–362.2	72.9	53.2	59.8
DBA	5	–	6.6–211.6	49.4	36.6	37.4	7.0–89.7	41.8	25.4	28.8
BghiP	6	30	N.D–287.9	67.2	45.9	60.2	N.D–273.8	61.3	41.3	53.1
$\Sigma_{7c}\text{PAHs}$			80.4–1134.0	324.8	241.8	214.2	27.6–1009.8	262.8	214.0	190.4
LMW PAHs			22.4–311.4	103.5	89.3	59.6	30.9–334.9	118.8	103.5	56.4
HMW PAHs			130.6–1610.1	486.9	385.3	304.2	27.6–1111.5	288.5	240.2	209.5
$\Sigma_{16}\text{PAHs}$			190.8–1921.5	590.4	479.6	337.2	58.5–1332.2	407.3	355.6	232.2

N.D: under detection limit. SD: standard deviation. $\Sigma_{7c}\text{PAHs}$: BaA, CHR, BbF, BkF, BaP, IND, DBA Carcinogenic PAH. LMW PAHs: low molecular weight 2–3 ring PAHs. HMW PAHs: high molecular weight 4–6 ring PAHs. $\Sigma_{16}\text{PAHs}$: Total 16 PAHs concentrations. a: Critical exposure values in Soil quality standards of the Netherlands.

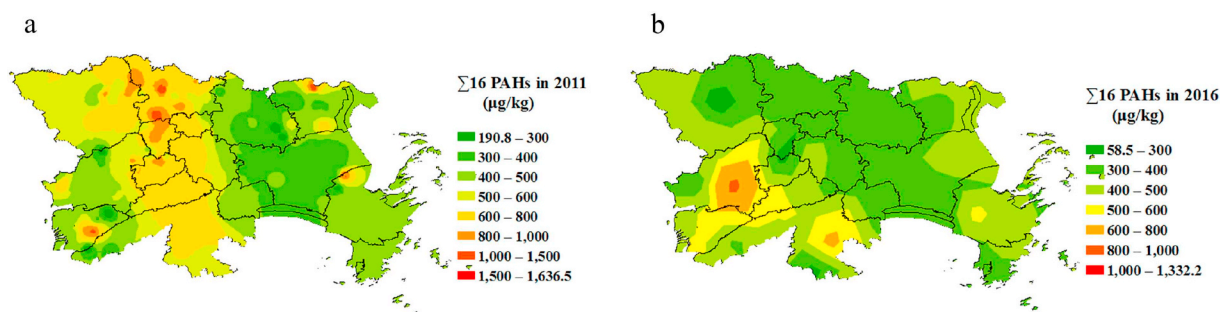


Fig. 2. Spatial distribution of PAHs in paddy soil in 2011 (a) and 2016 (b).

combustion. Those ratios above 0.5 were mostly moderately to heavily contaminated sites.

Furthermore, both three principal components (PC1, PC2 and PC3) were got and explained 60.6% and 63.5% of the total variability for soil PAHs in 2011 and 2016, respectively (Tables S10 and S11). The correlations between individual PAHs congeners in each component are significant (Tables S12 and S13), indicating they were from same source. For soil samples of 2011, PC1(2011) (32.9% of total variance) was dominated by 3–5 ring PAHs, with ACY, PHE, PYR, FLU, BaA, BaP, CHR and BbF. This group included both low and high molecular weight PAHs. ACY and PHE are low molecular weight (LMW) PAHs mainly caused by petrogenic, while high molecular weight (HMW) PAHs are more related to fossil fuel combustion. Fang et al. (2006) reported that high amounts of FLU implied incomplete combustion of fuel and oil burning. Since Wenling was a popular site for e-waste recycling, the open burning of e-waste, combustion of coal and straw to melt circuit boards to release valuable chips in coarse family-sized workshops might be one of the major causes to these congeners (Shen et al., 2009; Nishimura et al., 2017). Besides, the spatial distribution of factor scores of PC1(2011) (Fig. 4a) was higher in west, northwest where many industrial plant and e-waste workshops were exist. Therefore, PC1(2011) seems to represent combustion in e-waste dismantling process. The groups DBA, BghiP and IND, which projected on high values of PC2(2011), explained 15.4% of the total variance. DBA, BghiP and IND

are often associated with traffic emissions (Larsen and Baker, 2003). Moreover, the spatial distribution of PC2(2011) showed higher loadings in central city where many traffic roads densely located (Fig. 4b). Therefore, PC2(2011) was considered as vehicular emission. The third group consisted FL, ANT, ACE and NAP, which accounts for 12.3% of total variance, representing volatile LMW PAHs came from both anthropogenic and biological sources (Dahle et al., 2003; De et al., 2004). NAP and ACE are associated with high vapor pressure, low molecular weight, high fugacity, and relatively high solubility among the 16 PAH congeners. They can be transported for long time and distance in atmospheric and deposited into the soil mainly through gaseous deposition (Bakker et al., 2001). Wenling has a subtropical monsoon climate, the frequent wind is northwest except for in the summer time when the wind blows from the southwest. The high loadings in down wind area (far southeast edge) might cause by atmospheric deposition.

In 2016, PC1(2016) accounts for 29.8% of the total variability with higher loadings of BaA, BbF, CHR, BkF, IND, DBA, BghiP and BaP. When we took soil samples in 2016, some kinds of factories such as shoes making, textile clothing, leather and steel wires were distributed in northwest to west area. As the distribution of factor loadings of PC1(2016) showed, higher values were located in west part and southeast coastal area, PC1(2016) was attributed as mixed sources of combustion of fossil, coal in industrial production and traffic. PC2(2016) dominated by ACY, FLU, PYR and PHE, which explained

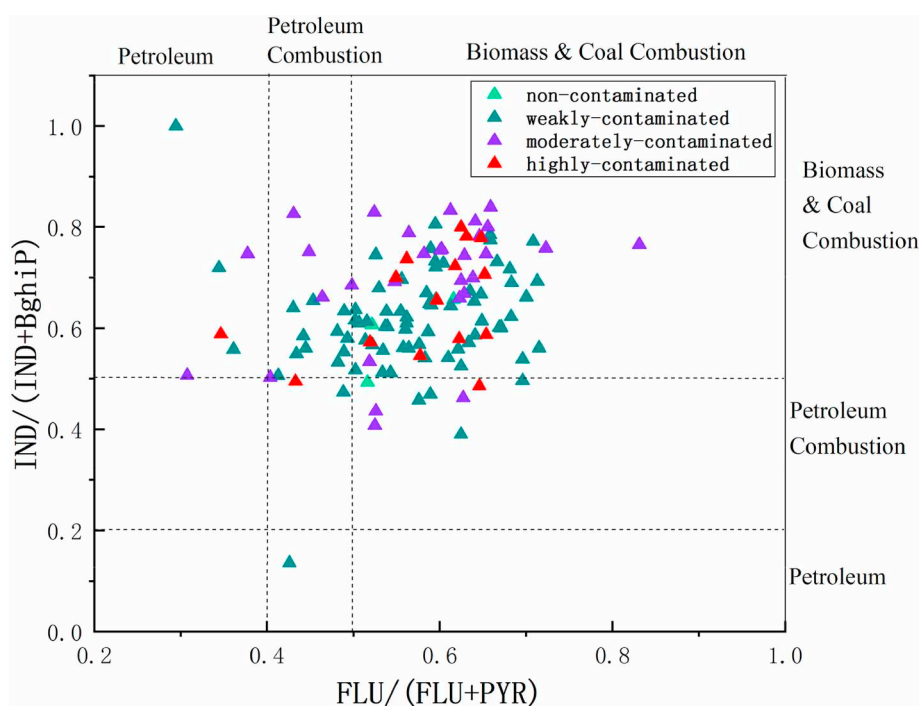


Fig. 3. Plot of IND/(IND + BghiP) vs. FLU/(FLU + PYR) for samples in 2011 and 2016.

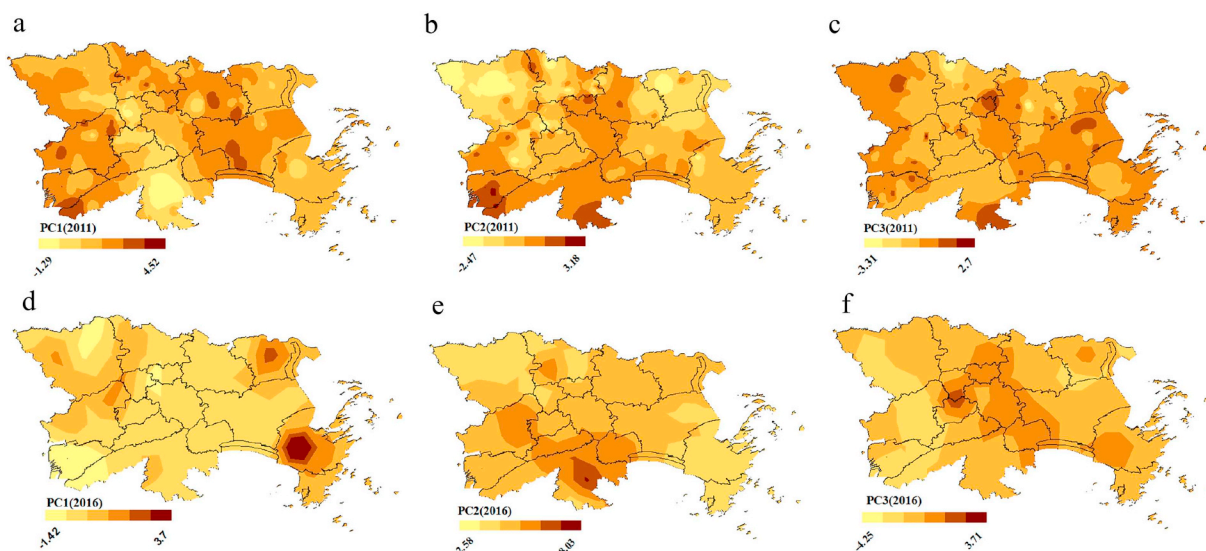


Fig. 4. Spatial distribution of factors scores of principal component analysis of PAHs in 2011 (a, b, c) and 2016 (d, e, f).

18.4% of the total variance, thought to represent the petrogenic sources. For the third factor, which explains 15.3% of total variance, has the high loadings of NAP, ACE, FL and ANT represent atmospheric decomposition as soil samples of 2011.

Combining from molecular weight ratios and principal loadings of the PAH congeners, we could infer that the mixed combustion of fossil fuel, biomass, coal in industrial and open burning of e-waste were the primary emission sources of PAHs in Wenling. And the secondary source might be traffic and deposition from long-range atmospheric deposition.

3.3. Health risk assessment and sensitivity analyses

3.3.1. Risk-based analyses towards the effect of environmental regulation

The estimated deterministic health risks incurring from the three exposure routes in 2011 and 2016 are listed in Table 2. $ILCR$ lower than 10^{-6} is considered to have no adverse effect on human health, and $ILCR$ ranges from 10^{-6} to 10^{-4} is regarded as acceptable level, while those exceeding 10^{-4} are regarded as high risk (USEPA, 2016). In 2011, the total $ILCR_s$ were ranged from 4.07×10^{-7} to 4.55×10^{-6} , with mean value of 1.59×10^{-6} . Further analyses showed that about 66.7% of sites have total risks exceeding the threshold value (10^{-6}), but are acceptable. While in 2016, the total $ILCR_s$ were ranged from 1.07×10^{-9} to 4.63×10^{-6} with mean value of 6.74×10^{-7} and only 16.7% sites exceeding 10^{-6} no adverse effect value. No site or year had the $ILCR_s$ over 10^{-4} . Ingestion is the dominant exposure pathway of $ILCR_s$, while inhalation is negligible in our assessment. The results are equivalent to the non-carcinogenic and carcinogenic risks exposure from PAHs in agricultural soils near an industrial site in Shanghai (Tong

et al., 2018) and the coastal wetland soils in China (Yang et al., 2015). Some researches have focused on risks of pollutants only from single exposure pathway, e.g. inhalation or ingestion (Shen et al., 2014; Zhang et al., 2016; Sun et al., 2017b), considering multi-exposure pathways make the risk assessment results more accurate.

The probabilistic $ILCR_s$ ranged from 3.02×10^{-10} to 3.37×10^{-5} with mean value of 2.90×10^{-7} in 2011 and 2.99×10^{-11} to 2.98×10^{-5} with mean value of 1.26×10^{-7} in 2016 exposure from PAHs through oral ingestion, inhalation and dermal contact pathways. Percentage of $ILCR_s$ exceeding 10^{-6} is 6.1% and 1.9% for the samples of 2011 and 2016, respectively (Fig. 5). In common with deterministic results, there is no risk value exceeding 10^{-4} , indicating that the farmers in study area are exposed to low potential health risks above acceptable levels (10^{-6}) in farmland soil.

The regulations are positive as the pollution degree and exposure risk both have mitigated with the strengthened environmental policy implemented. This reduction mainly due to slight degradation of PAHs in soil (Antizar-Ladislao et al., 2006) and decrease of coal, biomass combustion and primitive e-waste dismantling activities with environmental regulations implemented. Similarly, Fu et al. (2012) reported the significantly decreasing temporal trends of PCBs, PBDEs, PCDD/Fs in rice hulls during 2005 to 2009 with centralized recycling action and enhanced regulations on e-waste in Fengjiang town, indicating the stricter regulation measures and environmental policy led the positive effect of POPs. There are many other e-waste sites facing severe soil pollution and high exposure risk (Osibanjo and Nnorom, 2007; Robinson, 2009). To reduce soil PAHs concentration and mitigate the related exposure risks, more strict environmental regulations on primitive open combustion and associated supervising for e-waste

Table 2

The deterministic $ILCR_s$ via three exposure routes in 2011 and 2016.

Year	Exposure pathway	$ILCR_s$					
		Min	Max	Mean	5%	Median	95%
2011	Ingestion	$1.47E-07$	$1.64E-06$	$5.73E-07$	$2.09E-07$	$4.35E-07$	$1.27E-06$
	Dermal contact	$2.60E-07$	$2.91E-06$	$1.02E-06$	$3.72E-07$	$7.72E-07$	$2.25E-06$
	Inhalation	$1.14E-11$	$1.27E-10$	$4.44E-11$	$1.62E-11$	$3.37E-11$	$9.83E-11$
	Total	$4.07E-07$	$4.55E-06$	$1.59E-06$	$5.81E-07$	$1.21E-06$	$3.52E-06$
2016	Ingestion	$3.86E-10$	$1.67E-06$	$2.43E-07$	$1.69E-09$	$1.03E-07$	$1.19E-06$
	Dermal contact	$6.85E-10$	$2.96E-06$	$4.31E-07$	$3.00E-09$	$1.83E-07$	$2.12E-06$
	Inhalation	$2.99E-14$	$1.29E-10$	$1.88E-11$	$1.31E-13$	$8.00E-12$	$9.26E-11$
	Total	$1.07E-09$	$4.63E-06$	$6.74E-07$	$4.69E-09$	$2.86E-07$	$3.31E-06$

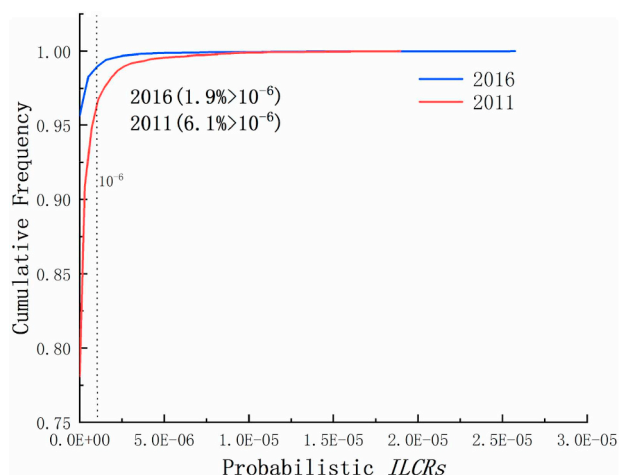


Fig. 5. Cumulative frequency of probabilistic $ILCR_s$ in assessment of 2011 and 2016.

activities could be effective as shown in this study. The combustion of biomass, coal during unregulated e-waste dismantling process was the main source of PAHs in our study area. The concentration and exposure risk of soil PAHs were both reduced with strict environmental management policy towards e-waste, while relatively high level of soil PAHs was still detected. Therefore, more environmental policies and regulations towards other pollution sources are also needed. Besides, target remediation strategies are supposed to be adopted in specific highly contaminated areas to reduce soil PAHs concentration.

3.3.2. Sensitivity analyses

Values that ranged from 0 to 100% in the sensitivity analyses represented the contributions of parameters to the risk assessment results. The most influential input parameter which contributed to the total variance of $ILCR_s$ was exposure frequency (EF), accounting for 72.6% and 53.6% for risk assessment of 2011 and 2016, respectively (Fig. 6). The results are consistent with the previous report in which the EF contributes the most to the total variance (Tong et al., 2018). However, some reports found the contribution of EF to risk is negligible (Yang et al., 2015; Yang et al., 2014). This difference may be ascribed to the different farming habit among farmers in different area. The C_s has the second highest contribution to the total variance of $ILCR_s$ in both assessments of 2011 and 2016, followed by soil intake rate (IR_{Ing}), exposure duration (ED), body weight (BW) and surface area (SA). While inhalation rate (IR_{Inh}) has the least contribution to the total variance of

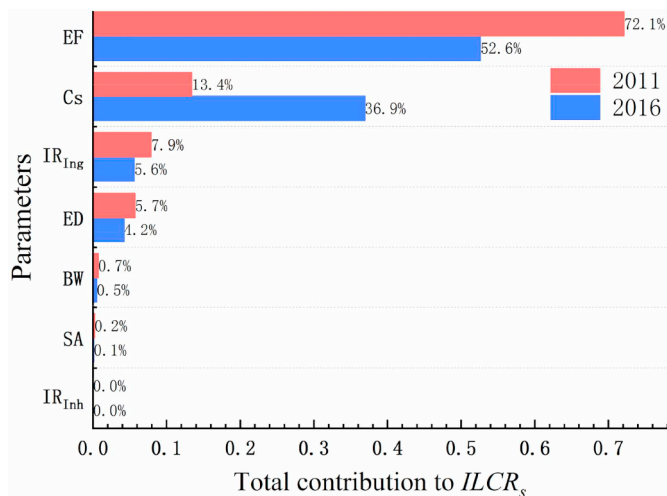


Fig. 6. Sensitivity analyses of parameters for $ILCR_s$.

$ILCR_s$, which confirmed the fact that the risk exposure from inhalation is negligible. The contribution of EF to total variance of $ILCR_s$ in 2016 reduced while C_s increased comparing with 2011, mainly because the difference in probability distribution (mean and Standard deviation values) of C_s in 2011 and 2016. Due to the great contributions of EF , C_s and IR_{Ing} , to reduce soil PAHs concentration by remediation actions and decrease exposure time and soil ingestion by application of mechanical equipment would be effective options to mitigate the exposure risk for working farmers.

3.3.3. Deterministic vs. probabilistic risk assessment

The deterministic method overestimated the risk up to 4–5 fold compared with the probabilistic results when considering variability of PAHs concentration and uncertainty of exposure parameters in the present study. This finding was consistent with the risk exposed to Naphthalene and Benzo(a) pyrene as reported by Jia et al. (2013), who reported the deterministic risk was overestimated 5–6 fold compared with probabilistic risk assessment. While contrary to those results risks were underestimated when parameters were probabilistic treatment for the assessment (Xia et al., 2010; Koupaie and Eskicioglu, 2015). The different results of probabilistic and deterministic risk assessment mainly arise from the single value used in the former either mean or maximum. More realistic via probabilistic risk assessment approach than the deterministic was confirmed, and it is useful to evaluate the importance of exposure parameters to total risks and make remediation strategies. However, the deterministic risk assessment can provide specific risk value for all the sampling sites, allowing for easy screening of potentially harmful sites.

4. Conclusions and implications

The mean concentration of total PAHs and the percentages of heavily and moderately contaminated soil in 2016 both declined relative to 2011. Moreover, the highly contaminated PAHs sites in northwest were lost with the closure of many family sized dismantling workshops. Mixed combustion of coal, petroleum, biomass and open burning of e-waste were the primary source of PAHs in study area. Both deterministic risk and probabilistic risk assessment revealed low potential risks were exposed to working farmers, with the exposure frequency (EF) the most influential parameter in the risk calculation. Overall, the strengthened management of e-waste recycling has been effective in reducing the soil PAHs contamination and related health risk. Further study on the bioaccumulation and biomagnification of PAHs found in rice grains and local population might be needed to better evaluate the risk to human health of contaminated rice intake. Ongoing monitoring of the PAH concentration in paddy soil and strict policy supervision are required.

There is still lack of official soil quality criteria for PAHs in China, the standard from Maliszewska-Kordybach (1996) was mostly adopted to classify the soil PAHs pollution levels in present study. It is essential for Chinese Environment Agency to establish soil PAHs standards. Insufficient and imprecise exposure parameters in the Chinese exposure guidebook add uncertainties for risk assessment. So there is a pressing need to update and establish a database of exposure parameters based on the characteristics of Chinese population to improve accuracy of risk assessment procedures.

Declaration of competing interest

The authors declare that there is no conflict of interests regarding the publication of this article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.105136>.

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