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A comprehensive framework for assessing the impact of potential agricultural pollution on grain security and human health in economically developed areas☆

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A B S T R A C T

Agricultural pollution poses a considerable challenge to grain security and human health, especially in economically developed areas. Mineral exploitation, chemical enterprise operation, pesticide and fertilizer application, sewage discharge, and vehicle emissions are the pollution sources of agricultural land. Identifying and assessing potential agricultural pollution (PAP) is, therefore, the most urgent task to achieve grain security and the human health. Large-scale (e.g., regional or national) PAP assessment can be very expensive, which could also generate a certain amount of information that usually discourages evaluation by decision-makers. To identify areas for regional priority investigation, here we proposed an assessment framework for PAP in economically developed areas. The framework consisted of PAP assessment, vulnerability assessment, hazard assessment, and socio-economic assessment. Then, we conducted a case study by using the proposed framework in one of China's economically developed areas, Zhejiang Province. The results showed that PAP, especially soil heavy metal pollution, soil acidification, and surface water pollution involved almost the entire study area. High-vulnerability high-hazard areas were mainly associated with high socio-economic development or high grain yield. These areas had negatively affected grain security and increased carcinogenic risk, potentially contributing to the formation of cancer villages. Based on the results, we proposed measures for environmental risk managers to alleviate the impact of PAP on grain security and human health in economically developed areas.

1. Introduction

Grain security and human health are important public concerns (Liu et al., 2013). If grain security is to be achieved, priority should be given to agricultural production (Lu et al., 2015a). As China's per capita arable land is less than half of the world's average, the country cannot withstand the negative impact of agricultural pollution on grain security and human health (Fu et al., 2013; Lu et al., 2015b). It is estimated that more than 10 million tons of grains contain pollutants that exceed the national standard every year in China (Liu et al., 2013; Rai et al., 2019). Therefore, it is crucial to identify areas with potential agricultural pollution (PAP) and apply control measures (Hu et al., 2019). In many cases, however, it is unfeasible to investigate all types of PAP because of limited financial resources; instead, attention should be given to those areas with high vulnerability and high hazard of pollution (Pizzol et al., 2011). Therefore, there is an urgent need to identify PAP and its priority investigation areas (Agostini et al., 2012; Li et al., 2014).

Soil heavy metal pollution, soil acidification, surface water pollution, and air pollution may give rise to PAP (Guo et al., 2010;

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Du et al., 2013; Wei et al., 2020). For example, large amounts of heavy metals have entered the farmland due to the non-standard production process of mining and chemical enterprises (Augustsson et al., 2018). The over-standard rates of heavy metals at soil sampling points around mining areas and polluting enterprises is 33.4% and 36.3%, respectively (MEE&MNR, 2014). In addition, the overuse of fertilizers and pesticides has caused serious agricultural pollution, and chemical fertilizers are identified as the chief culprit of soil acidification in China (Guo et al., 2010). The average fertilizer application rate in several provinces of China even exceeded the internationally recognized upper limit (225 kg/hm^2) by up to 2–3 fold (Jiang et al., 2018), resulting in serious soil acidification in farmlands over the past 40 years. Moreover, soil acidification can enhance the bioavailability of soil heavy metals (Zeng et al., 2011). For example, a reduction in pH of one unit increases soil Cd or Pb availability by 4–5 fold (Wang et al., 2019), which in turn enhances crop uptake of these metals and endangers human health through the food chain (Qian et al., 2010; Åkesson et al., 2014). Therefore, PAP caused by soil acidification is generally regarded as the most serious concern. Notably, soil pollutants accumulate to the extent that they may hinder soil function and pollute groundwater and surface water (Pizzol et al., 2011). Sewage irrigation would last for a long time when there are no alternative water sources, which could cause PAP (Jiang and Guo, 2019; Rai et al., 2019; Shi et al., 2019). There is no doubt that air pollution has the same effect (Xu et al., 2017; Wei et al., 2019).

According to Vegter et al. (2003), the management of polluted land should follow the concept of risk-based land management. In this context, regional risk assessments have been carried out by using the geographical information system. For example, Pizzol et al. (2011) conducted a regional risk assessment for polluted sites in Poland. Subsequently, Hu et al. (2017) assessed the regional risks of soil heavy metals in China. More recently, Teng et al. (2019) developed a regional risk assessment method for nitrate pollution of groundwater in China. Although previous studies have assessed pollution in soil, groundwater, surface water, and air (Hu et al., 2017; Teng et al., 2019; Wei et al., 2019, 2020), a comprehensive assessment framework for PAP (including soil, water, and air) has not been established. Therefore, a new assessment framework for PAP is required, which in addition to this must include vulnerability assessment, hazard assessment, and socio-economic assessment. From this perspective, estimating the vulnerability of receptors and assessing the hazard of pollution sources are of utmost concern (Zabeo et al., 2011), and they represent two of the initial steps in PAP assessment at a regional scale (Vegter et al., 2003).

The guarantee of grain security and human health strongly requires ranking information of PAP, based on which the local government could assess the areas for priority investigation (Agostini et al., 2012; Li et al., 2014). In this study, we proposed a comprehensive framework for assessing PAP using backpropagation neural network (BPNN) and K-means clustering algorithm. Then, we conducted a case study by using the framework in one of China's economically developed areas, Zhejiang Province. On this basis, we identified the impact of PAP on grain security and human health.

2. Assessment framework for PAP

2.1. PAP assessment

Soil heavy metal pollution, soil acidification, surface water pollution, and air pollution are the four pathways of PAP (Fig. 1). In general, the spatial distribution of soil heavy metal pollution can be obtained by spatial interpolation methods based on field sampling data. For example, inverse distance weighted interpolation precisely predicts the level of soil pollution under uniform sampling. Subsequently, the PAP areas can be identified according to the technical regulations of heavy metal pollution assessment provided by regional authorities (Hu et al., 2019; Jia et al., 2019). Similarly, soil acidification can be obtained from temporal changes in soil pH by soil observations and spatial interpolation techniques (Chen et al., 2019). Surface water quality of different water systems and at different intervals has been determined by river sampling and the results of subsequent laboratory analysis (Grant et al., 2012). Usually, the buffer zone of surface water can be conducted to determine the areas involved (Pizzol et al., 2011). Air pollution could be assessed by spatial interpolation techniques based on air

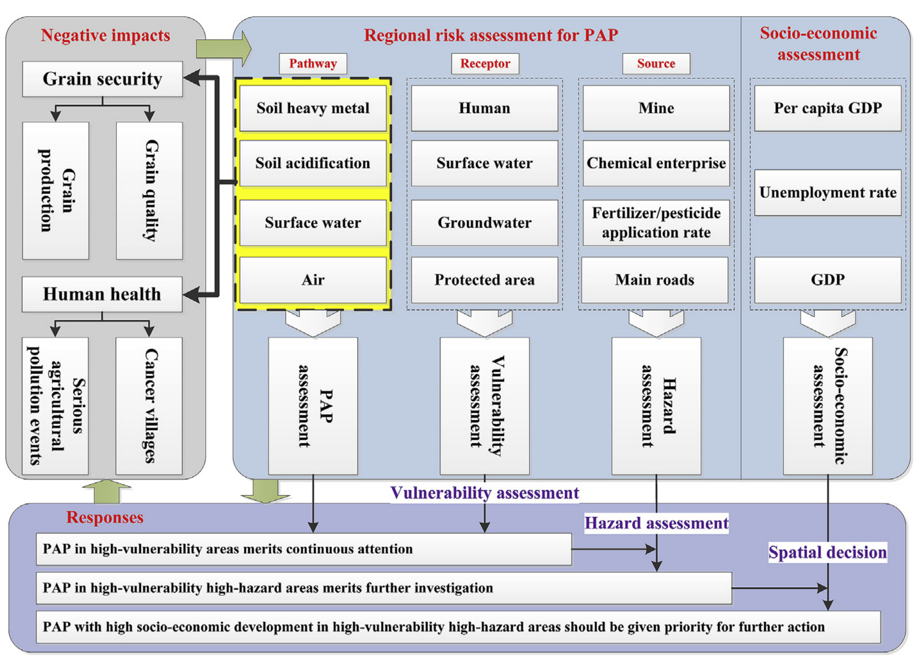

Fig. 1. A comprehensive framework for assessing the impact of potential agricultural pollution (PAP) on grain security and human health in economically developed areas.

quality monitoring data from meteorological sites (Wei et al., 2019, 2020), such as $PM_{2.5}$, PM_{10}, or NO_2. Finally, PAP can be evaluated by spatial summarization of the pollutants from these four pathways.

2.2. Vulnerability assessment

The objective of vulnerability assessment is to estimate the receptor's sensitivity by processing the spatial relationships and properties of the receptors (Fig. 1). The assessment can be divided into the following five steps (Zabeo et al., 2011): 1) identify regional-scale receptors; 2) distinguish the vulnerability of receptor-related attributes; 3) assign attribute values to receptors; 4) normalize attribute values; and 5) apply BPNN to aggregate attribute values for estimation. BPNN consists of an input layer, a hidden layer, and an output layer (Olawoyin, 2016). BPNN does not need to determine the mathematical equation of the mapping relationship between the input and output layer in advance; instead, it only obtains the closest result to the expected output value when given the input value by learning a certain rule. The most commonly used function f is the sigmoid function, and the weight of the net is corrected by the back propagation of the error signal and the forward propagation of the working signal. When the net minimizes the total mean-square error, a mapping relationship is established using Eq. (1):

$$net : y = f(x) \tag{1}$$

In this study, we used the stochastic gradient descent to optimize the network according to non-monotonic measure questionnaires completed by experts (Table S1). The trained network is used to simulate for human, protected area, and surface water data (Eq. (2)), with a hidden layer, six neurons configuration, and learning rate factor $\eta = 0.00001$, respectively.

$$y = sim(net, x), x = human,\ surface\ water,\ or\ protected\ area \tag{2}$$

Receptors are required to assign relevant attributes as characterization; however, understanding the specific background and accessibility of data is necessary. While receptors containing all relevant attribute values must be aggregated to estimate their vulnerability scores, the normalization process should be performed so that all attributes can enter the same spatial domain before the aggregation. According to experts (Pizzol et al., 2011; Zabeo et al., 2011; Agostini et al., 2012), the simplest way to define this function is to use a continuous piecewise linear function whose segmentation points are defined by experts, such as the closed interval [0,1]. Finally, vulnerability assessment is aggregated by the assessment results of these receptors' vulnerability using the K-means clustering algorithm.

The K-means clustering algorithm is a widely used classical clustering method involving the following steps (Li et al., 2018): 1) determine the number of clusters; 2) input raw samples $x = (x_1, x_2, \cdots, x_n)$ and randomly select k samples as the initial cluster center $z = (z_1, z_2, \cdots, z_k)$; 3) divide samples by calculating the Euclidean distances between samples x and cluster centers z (Eq. (3)), and assign each sample to the closest cluster; 4) recalculate the center of each cluster after all samples are assigned; and 5) repeat steps 3) and 4) until the criterion functions E begins to converge (Eq. (4)).

$$d(x, z) = \sqrt{\sum_{i=1}^{n} (x_i - z_j)^2}, (j = 1, 2, \cdots, k) \tag{3}$$

$$E = \sum_{j=1}^{k} \sum_{i=1}^{n_j} d(x_i, z_j), x_i = C_j \tag{4}$$

where $d(x_i, z_j)$ is the Euclidean distance; x_i is the attribute of samples; and Z_j is the cluster center of the subclass C_j in Eq. (3); E is the sum of all with-class distances; n_j is the sample size of subclass C_j; and $d(x_i,\ z_j)$ is the European distance between sample x_i and cluster center Z_j in Eq. (4). The smaller the value of E, the greater the clustering effect.

2.3. Hazard assessment

The objective of hazard assessment is to estimate the hazard scores of pollution sources, including mines, chemical enterprises, main roads, and fertilizers (Fig. 1), which suggests that more than one potentially polluting activity may have acted on the same area. Therefore, the hazard assessment was obtained by aggregating the scores of each activity. Specifically, kernel density was used to determine the density value of each pollution source and the hazard scores were estimated by the K-means clustering algorithm after normalization of density values. Detailed description of kernel density and bivariate local indicators of spatial association have been reported by Jia et al. (2019).

2.4. Socio-economic assessment

The objective of socio-economic assessment is to rank local administrative units by their economic development (Fig. 1). It mainly includes the following steps (Agostini et al., 2012): 1) selecting proper socio-economic units of assessment; 2) determining specific socio-economic parameters (e.g., gross domestic product [GDP], per capita GDP, and unemployment rate) for estimating economic development level; and 3) applying the K-means clustering algorithm to aggregate the parameter values. Socio-economic units can be directly determined using administrative boundaries.

2.5. Responses

Using the assessment framework for PAP, we can identify areas where regional authorities should give priority for further action (Fig. 1). First, the results of PAP assessment are combined with vulnerability assessment data to identify areas with high vulnerability to PAP. Second, the hazard assessment results are superimposed based on the previous step; high-vulnerability high-hazard areas are worthy of further investigation. Finally, areas with high socio-economic values should be given priority for further action by regional authorities. This simple spatial visualization may motivate decision-makers to investigate and manage PAP in economically developed areas.

3. Case study

3.1. Study area

Zhejiang Province (27°02′-31°11′ N, 118°01′-123°10′ E) is located in Southern China and has a total area of 1.055×10^5 km^2. Zhejiang is one of the most developed provinces in China. By the end of 2018, the GDP of Zhejiang ranked fourth in China with an increase of 7.1% from 2017. According to a 2015 report, forest, agricultural land, grassland, and water body are the major land-use types in Zhejiang. The areas of forest and agricultural lands are 5.69×10^4 km^2 and 2.62×10^4 km^2, respectively, accounting for 53.9% and 24.8% of the total land area. According to the Department of Eco-Environment of Zhejiang Province, a total of 3710 key pollutant discharge units have been identified in 2019. Moreover, there are many types of minerals in Zhejiang, with more than 2000 mines included in the statistics. The reserves of alunite rank first in the world, and the reserves of fluorite rank second in China.

3.2. Dataset

The following data were collected in the study area (Table 1). First, 14,801 topsoil samples (0–20 cm) were collected in 2013 by systematic grid sampling (1 km × 1 km; Fig. S1). Soil As, Pb, Cd, Cr, Hg, and pH levels were determined by standard testing methods. Detailed sampling and laboratory testing methods have been reported by Jia et al. (2019). Second, the division of environmentally functional zones of surface water (2011–2015) and the types and ranges of the protected area were obtained from the Department of Eco-Environment of Zhejiang Province. Third, soil acidification assessment was based on the spatial distribution of soil pH in the 1980s (Chen et al., 2019) and 2010s in this study. Fourth, 1-km resolution high-quality daily PM2.5 concentrations from 2015 to 2018 were collected, which were generated by Wei et al. (2019, 2020) using the improved tree-based ensemble learning approaches. Five, population density (1990–2015, 1-km resolution), land-use (2015, 100-m resolution), and major traffic road data were obtained from the Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (http://www.resdc.cn). Six, Zhejiang Statistical Yearbook (1998–2018) was used to obtain the percentage of vulnerable groups (2004–2018), fertilizer application rate (1998–2018), unemployment rate (1998–2018), GDP (1998–2018), per capita GDP (1998–2018), and grain production (1998–2018). Seven, a total of 2308 mine point data was obtained, mainly, from the Ministry of Natural Resources of the People's Republic of China (http://geoglobal.mnr.gov.cn/). Eight, a total of 20,531 potentially polluting enterprise points were corrected by the web crawler and partially verified by field survey. Finally, 24 cancer villages were derived from the latest Chinese government-supported survey data (Gong and Zhang, 2013) and reports by mainstream media. The data were projected to a unified coordinate system for subsequent analysis.

3.3. Results and discussion

3.3.1. PAP assessment results

3.3.1.1. Heavy metals. According to the Chinese official assessment criteria (Tables S2 and S3), the assessment results of soil heavy metal pollution (Fig. S2) showed that pollution by these elements occurred across the study area. In fact, 26.5% (Cd) and 44.1% (Hg) of the soil samples exceeded the risk screening values. The maximum contents of Cd and Hg were 114 and 7 mg kg^{-1}, respectively, which exceeded the risk screening values by 10-fold each (Jia et al., 2019). In 2010, the provincial government of Zhejiang conducted a field survey across 2.365 million hectares of agricultural land, nearly 48.8% of which was polluted by heavy metals and 20.0% was not suitable for growing green agricultural products (Wu et al., 2015).

3.3.1.2. Soil acidification. Soil acidification occurred in the entire study area (Fig. S2). In 1980 and 2010, more than half of the soil had pH values < 6.5 (Fig. S1). Soil acidification was the second most important reason for the high accumulation of heavy metals in agricultural land across Zhejiang (Zhao et al., 2015). Even though the local soil is naturally acidic, anthropogenic acidification occurred due to the excessive use of nitrogen fertilizers and intensification of agricultural production (Wang et al., 2019). Compared with fertilization, the atmospheric deposition was a secondary factor resulting in the decline of soil pH in agricultural land across China over the past 30 years (Guo et al., 2010). According to Zhejiang Statistical Yearbook (1998–2018), the fertilizer application rates (324–888 kg/hm^2; Fig. 2a), which were similar to those in other provinces of China (Lu et al., 2015b), exceeded the internationally recognized upper limit.

3.3.1.3. Surface water pollution. Based on the results of surface water quality (Fig. S1), a 100-m buffer zone was selected. According to China's environmental quality standards for surface water (Table S4), pollution was observed in the entire study area (Fig. S2). Based on the survey of surface water quality (Table S5), the total probability of water quality meeting the national standard in Zhejiang was only 61.2%, and the water in several areas was almost completely polluted. According to Zhejiang Statistical Yearbook (1991–2018), the discharge of industrial wastewater in the study area increased >3-fold from 1990 to 2017, and the discharge of wastewater in 2017 was >4.5 billion tons (Fig. 2b). However, sewage treatment rate reached 70.0% in urban areas, while it was <7.0% in rural areas (Chen, 2007). Massive untreated sewage was directly discharged into rivers and lakes in the study area (Shi et al., 2019).

3.3.1.4. Air pollution. The spatial distribution of $PM_{2.5}$ (Fig. S1) showed that the air quality in the study area was beyond the pollution range according to the Chinese air quality standard (Table S6). From 1990 to 2017, industrial waste gases were continuously discharged (Fig. 2b); however, the air quality did not reach the limit of pollution. A plausible reason for this finding is the implementation of industrial and energy structural optimization policies by the Zhejiang government (Xu et al., 2017). Compared with the 2013 data, the proportion of energy consumption such as coal and oil decreased by 2.8% in 2014, while the proportion of clean energy consumption such as natural gas, water, and nuclear power increased by 1.4% (Xu et al., 2017). Even though other air pollutants such as SO_2, NO_2, and PM_{10} were not involved, the results (Xu et al., 2017) seemed to be consistent with $PM_{2.5}$. Altogether, the assessment results indicate that PAP involved almost the entire study area of Zhejiang (Fig. 3a).

3.3.2. Vulnerability assessment results

The distribution of population density (Fig. S3) showed that the population density was higher in eastern than western Zhejiang. The expert scores of land use (Table 1) revealed that the vulnerability of land-use was relatively high in the study area. Specifically, highly vulnerable land-use (e.g., agricultural land and forest) accounted for 78.7% of the total study area (Fig. S3). The percentage of vulnerable groups was higher than 30.0% (Fig. S3), indicating a wide distribution of the vulnerable groups. Groundwater was not considered in this study. The vulnerability assessment results (Fig. S4) showed that both human and protected areas had a high vulnerability, while surface water had a medium vulnerability. Low vulnerability in Fig. 3b may be lack of assessment results of human and surface water (Fig. S4). Interestingly, the distribution of high vulnerability (Fig. 3b) was similar to the distribution of surface water and protected areas because they were more vulnerable than human (Table S1).

3.3.3. Hazard assessment results

The kernel density results of mines (Fig. S5) showed that the spatial heterogeneity of mine distribution was relatively high, and many hotspots of mine distribution were observed. The kernel density results of the main roads and chemical enterprises shared a similar pattern (Fig. S5), and the high-density areas were mainly distributed along the coastal area with a relatively developed

Table 1
Relevant attributes for the assessment of potential agricultural pollution (PAP), vulnerability, hazard, and social-economy.

Phase		Index	Grade	Description	Scoring by expert	Data Sources
PAP assessment	Soil heavy metal pollution assessment	Heavy metal in soil (including As, Pb, Cd, Cr, and Hg)	No risk	–	–	Field sampling
			Low risk	–	–	
			Medium risk	–	–	
			High risk	–	–	
	Soil acidification assessment	pH[a] drops more than 0.5	Soil acidification risk	–	–	Chen et al., (2019) and This study
		Others	No risk	–	–	
	Surface water quality assessment	Surface water quality	I	–	–	ZJEED[a]
			II	–	–	
			III	–	–	
			IV	–	–	
	Air quality assessment	$PM_{2.5}$ ($\mu g/m^3$)	0–35	–	–	Wei et al., (2019), 2020
			35–75	–	–	
			>75	–	–	
Vulnerability assessment	Human	Population density (200 people/km^2)	<2	Low density	0.2	RESDC[b]
			2–5	Medium density	0.4	
			5–10	High density	0.6	
			10–20	Very high density	1.0	
		Land use	Agricultural land	–	0.7	
			Forest	–	0.8	
			Grassland	–	0.8	
			Waters	–	0.4	
			Construction land	–	0.3	
			Isolated area	–	0.2	
		Percentage of vulnerable Groups (%)[c]	<30	Low	0.3	Zhejiang Statistical Yearbook (2004–2018)
			30–33	Medium	0.5	
			33–36	Medium–high	0.7	
			>36	High	1.0	
	Protected areas	Type	State level	–	1.0	ZJEED
			Province level	–	0.8	
		Extension (100 hm^2)	<1	Tiny extension	0.6	
			1–1.5	Small extension	0.7	
			1.5–10	Medium extension	0.8	
			10–40	Large extension	0.9	
			>40	Huge extension	1.0	
	Surface water	Size	Very large river	–	0.0	RESDC
			Large river	–	0.3	
			Medium river	–	0.5	
			Small river	–	0.8	
		Grade	Primary tributary	–	0.3	
			Secondary tributary	–	0.6	
			Sub-secondary tributary	–	0.8	
Hazard assessment	Mine	Mine density	–	–	–	MNR[d]
	Chemical enterprises	Enterprise density	–	–	–	Web Crawler
	Main roads	Density of railways, highways, and national ways	–	–	–	RESDC
	Fertilizer	Fertilizer application rate	–	–	–	Zhejiang Statistical Yearbook (1998–2018)
Socio-economic assessment	–	Per capita GDP[e] (10^4 RMB yuan)	<2	Low density	0.2	
			2–3	Medium density	0.4	
			3–4	Medium-high density	0.6	
			4–5	High density	0.8	
			>5	Very high	1.0	
	–	Unemployment rate[f]	<0.2	Low	1.0	
			0.2–0.3	Medium	0.7	
			0.3–0.4	Medium-high	0.4	
			0.4–0.5	High	0.2	
			>0.50	Very high	0.0	
	–	GDP[e] (10^8 RMB yuan)	<2	Low	0.2	
			2–3	Medium	0.3	
			3–4	Medium-high	0.4	
			4–5	High	0.5	
			>5	Very high	0.8	

[a] Both pH in two periods is less than 6 involved in the calculation; ZJEED, Department of Eco-Environment of Zhejiang Province.
[b] RESDC, Resources and Environmental Sciences, Chinese Academy of Sciences.
[c] The percentage of vulnerable groups represented the population percentage composed of children (<18) and elderly persons (>60).
[d] MNR, Ministry of Natural Resources of the People's Republic of China.
[e] Divided by the respective purchasing power in Zhejiang and the criteria of the World Bank.
[f] Refers to those people who are not engaged in social work, including students, homemakers, municipal unemployed, retired, lost working ability and so on.

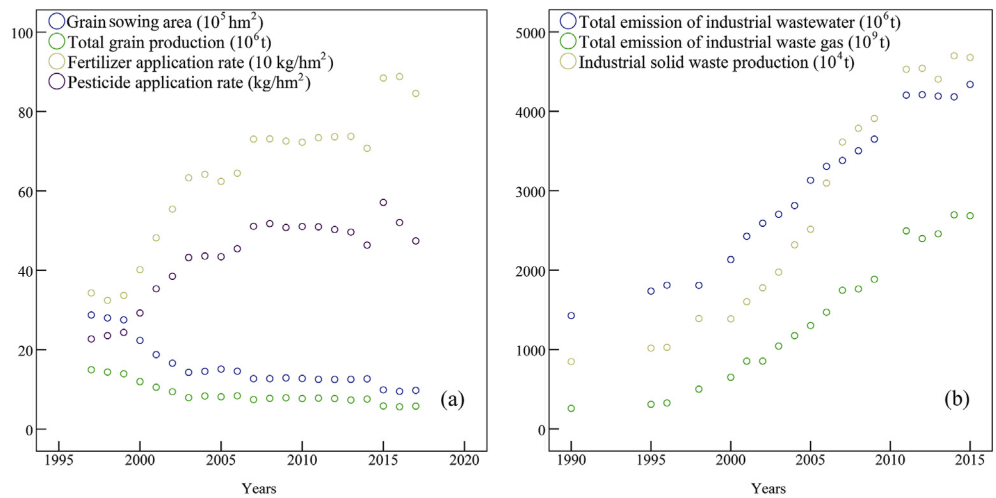

Fig. 2. Grain sowing area, total grain production, and fertilizer/pesticide application rate (a), industrial waste emissions (b) in Zhejiang over the past two decades (Zhejiang Statistical Yearbook, 1991–2018).

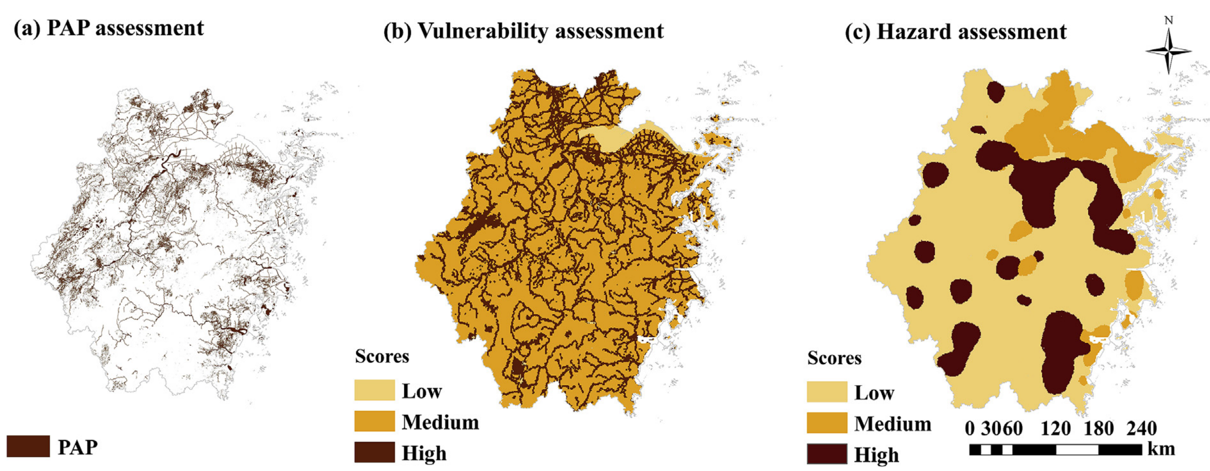

Fig. 3. Results of (a) potential agricultural pollution (PAP) assessment, (b) vulnerability assessment, and (c) hazard assessment in Zhejiang, China.

economy. The hazard assessment results (Fig. 3c) revealed that the distribution of high-hazard areas was similar to the kernel density results of mines and chemical enterprises, which were the two main sources of PAP. According to Zhejiang Statistical Yearbook (1991–2018), industrial wastes increased yearly (Fig. 2b). For example, waste water, gas, and residue increased from 142,717, 2,595, and 847 tons in 1990 to 453,935, 31,310, and 4,828 tons in 2017, respectively; these waste emissions increased by 2–12 fold over the study period. Wastes released by mines were not quantified, but they could be much higher (Li et al., 2013). This growth rate is notable and should be reduced as soon as possible while ensuring grain security.

3.3.4. Socio-economic assessment results and responses

Expert scores were assigned to different parameters (Table 1) following the general rule. In fact, the maximum score of 1 was assigned to high GDP, high per capita GDP, and low unemployment rate, and the lowest score of 0.2 was assigned to the classes corresponding to disadvantaged or underdeveloped social economy. Between the two extremes, other levels and related scores were defined by literature reviews and experts (Agostini et al., 2012; Li et al., 2014). The socio-economic assessment results (Fig. S6) showed that the economically developed areas were mainly located in the northern area and included the cities of Hangzhou and Ningbo, both of which have been ranked among the top 20 cities in China for a long time.

Combined with the results of vulnerability and hazard assessments, we obtained the high-vulnerability areas (Fig. 4a), high-vulnerability high-hazard areas (Fig. 4b), and priority investigation areas (Fig. 4c) across Zhejiang. The high-vulnerability high-hazard areas were mostly associated with very high or high levels of socio-economic development (Fig. S6). This result indicates that high levels of socio-economic development were likely to contribute to PAP.

3.4. Integrated measures for addressing the impact of PAP

3.4.1. PAP in relation to grain security

3.4.1.1. Impact of heavy metals on grain security. In this study, hotspot areas of soil heavy metal pollution were identified across Zhejiang, including high-risk areas, medium-risk areas, and low-risk areas (Fig. S1). Previous studies have reported that a significant proportion of heavy metal in crops exceeds the standard limits when the crops are grown in soil with heavy metals exceeding the risk screening values in Zhejiang (Fu et al., 2013; Hu et al., 2017). For example, Zeng et al. (2011) observed that grain Pb contents in over

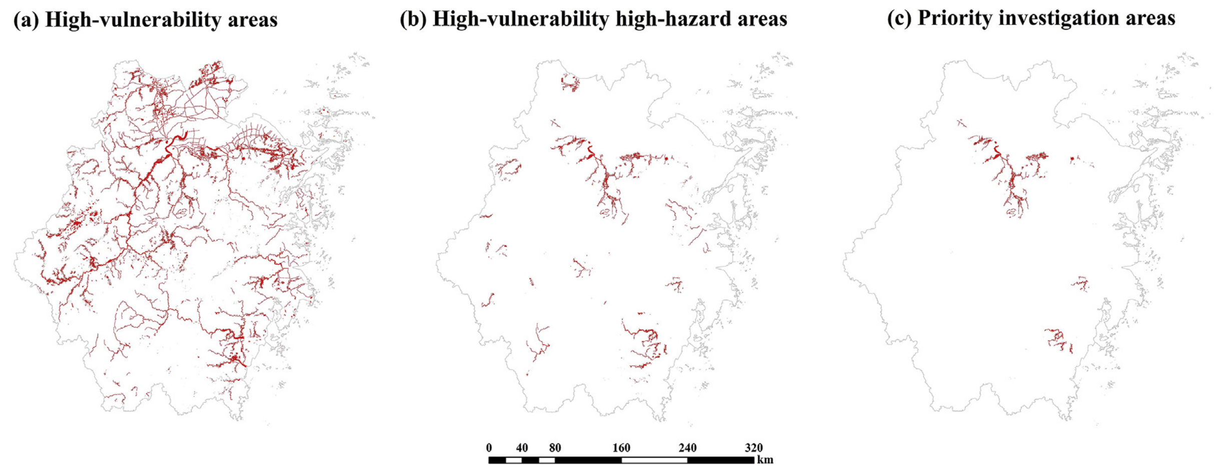

Fig. 4. Identification of (a) high-vulnerability areas, (b) high-vulnerability high-hazard areas, and (c) priority investigation areas in Zhejiang, China.

80.0% crop samples from three counties (districts) of Zhejiang exceeded the maximum allowable level (0.2 mg kg^{-1}) of the World Health Organization (WHO) (JECFA, 2011). The fact is that large amounts of heavy metals and metalloids have been discharged into the soil over the past decades (Fig. 2b), along with rapid industrialization and urbanization in Zhejiang (Shi et al., 2019; Hu et al., 2020a). While the current status of soil heavy metal pollution in Zhejiang may not be as grim as perceived in previous studies, there are still reasons for concern regarding grain security.

Similar results have been reported in other regions of the world (Zhu et al., 2008; Meharg et al., 2013). In southern China, a large proportion of heavy metals in rice exceed the standard limits, which has attracted widespread public attention over the last decade (Williams et al., 2009). Nationally, the Market Basket Survey reported that 2.0%–13.0% of rice samples exceed the limits of heavy metals in China (Qian et al., 2010; Zhao et al., 2015). Based on a survey of 12 countries across four continents, Cd contents in rice grain are the highest in Bangladesh and Sri Lanka, countries that have high per capita consumption of rice (Meharg et al., 2013). Compared with dietary Cd intakes of adult males and children in different countries, the average intake rate is higher in China than in the USA, UK, France, New Zealand, and Australia, but lower than in Japan (Table S7). Mainland China has adopted a 0.2 mg kg^{-1} standard limit of Cd content in rice, which is stricter than the threshold of 0.4 mg kg^{-1} for rice adopted by FAO/WHO and some other rice-consuming nations and regions (Hu et al., 2016), particularly Japan and Taiwan (China). This strict limit is considered necessary because of the high consumption of rice in mainland China, especially southern China.

3.4.1.2. Impact of fertilizers and pesticides on grain security. In general, the application rates of fertilizers and pesticides increased from 1997 to 2017 (Fig. 2a). In contrast, total grain production declined during this period, indicating that excessive application of fertilizers and pesticides did not stimulate grain production and had a negative impact on grain security. First, excessive use of nitrogen fertilizers has accelerated soil acidification in the study area (Fig. S1). Heavy metal content in grain may exceed the standard limits in "unpolluted" soil, and this phenomenon is most likely due to acidic soil pH (Zhao et al., 2015; Hu et al., 2016). In addition, grain can be easily infected by pests and diseases (especially soil-borne pathogens and root nematodes) after soil acidification (Abawi and Widmer, 2000). Second, grain protection by the use of pesticides has made a significant contribution to growth in agricultural productivity (Lu et al., 2015b). However, the average consumption of pesticides over the past 21 years is 43.22 kg/hm^2 (Fig. 2a), much higher than the average levels in China (15 kg/hm^2) and other Asian countries (Lu et al., 2015b). There is no doubt that pesticide residues in grains result from excessive pesticide applications. In 2004, the average detection rate of organic pesticides (OPs) in grains surveyed for grain security across Zhejiang was 26.5%, close to 27.8% in 2003 (Shen et al., 2007). However, more OP species were identified in 2004 than in 2003, and 6.3% of vegetables contained multi-component pesticides. Several banned OPs still remain as residues in the soil, which may continue to affect grain quality due to the long residue time (Shen et al., 2007).

The decrease in total grain production from 1997 to 2017 (Fig. 2a) may be attributed to two factors: the decrease in grain sowing area and the policy or market adjustments. Specific policy or market adjustments may stimulate grain production. For example, the Zhejiang government adjusted a series of policies and the agricultural markets in 2003, such as subsidies for seeds, marketization of grain purchase and sale, and abolished agricultural taxes (Lu et al., 2015b). Therefore, both grain sowing area and total grain production increased in the following three years (Fig. 2a). After 2003, the correlations between pesticide application, fertilizer application, and grain production became lower than those before 2003 (Table S8). This indicates that policy and/or market adjustments reduced the dependence of grain production on fertilizers and pesticides. Furthermore, according to the Integrated Pest Management (FAO, 2010) and Soil Testing and Formulated Fertilization (Ministry of Agriculture and Rural [MAA], 2005), farmers could use fewer pesticides and fertilizers to achieve higher yields. However, the results of Pearson's correlation analysis ($|r| = 0.561–0.996$; Table S8) indicate that the current grain production is still heavily dependent on the use of pesticides and fertilizers, regardless of policy or market adjustments. Despite their decreasing trend over recent years (Fig. 2a), the rates of fertilizer and pesticide application are still very high. The rates of soil acidification and the risks of grain security would substantially increase due to long-term application of fertilizers and pesticides at high rates. The emergence of this situation poses an extreme threat to grain security in Zhejiang and has become a global problem.

3.4.1.3. Impact of surface water pollution on grain security. In China, the soil polluted by heavy metals covers an area of >20 million hm^2; 26.4% of this polluted soil area is caused by sewage irrigation (MEE&MNR, 2014). The double-rice cropping system, a typical planting system in the study area, requires a large volume of surface water for irrigation (Zhao et al., 2015). With increasing sewage discharge and poor surface water quality, sewage irrigation has emerged as an effective measure to alleviate water shortage and increase agricultural production (Jiang and Guo, 2019). However, sewage contains large amounts of potentially hazardous substances; it, therefore, could add pollutants or harmful substances to the soil and reduce grain quality if continuously used in an inefficient irrigation system (Du et al., 2013; Ye et al., 2015).

Sewage emission from mines and chemical enterprises (Fig. 2b) is the major cause of poor surface water quality in Zhejiang (Rai et al., 2019). Du et al. (2013) reported that Cd content in polluted surface water from mining areas and enterprises ranged from 0.02 to 0.04 mg/L, whereas Cd contents in two paddy fields near the polluted surface water ranged from 3.46 to 4.16 mg kg^{-1}. Assuming 1 m of irrigation water per year in the study area, the input of Cd into the paddy system would amount to 50–400 g/ha (Zhao et al., 2015). When local surface water reaches pollution levels, the amount of heavy metals entering the soil and crops may be substantial (Jiang and Guo, 2019). Unfortunately, the relationship between long-term sewage irrigation and grain quality has not attracted much attention from the government, and public awareness of grain quality is relatively low in Zhejiang. In 2013, the Chinese government banned for the first time the application of sewage containing heavy metals and/or persistent organic pollutants for irrigation (The State Council of China, 2013). However, policy implementation remains challenging, especially in areas lacking adequate water quality.

3.4.2. PAP in relation to human health

3.4.2.1. Serious agricultural pollution events. Twenty incidents involved in agricultural pollution in Zhejiang over the past 10 years have been summarized (Fig. S7). These widely reported events (Table S9) had significant effects on public health and resulted in economic losses, and agricultural systems in the affected areas seemed to be more vulnerable, especially soil and surface water. For example, in 2011, the illegal discharge of wastes by Taizhou Battery Company in Zhejiang resulted in the loss of basic functions of 4377.03 m^2 farmland due to excessive Pb content in the soil, and blood Pb levels of 68 people living nearby exceeded the standard levels. It also garnered headlines of local and national media and elicited considerable concern from the public (Shi et al., 2019). In the same year, solid wastes were discharged to nearby rivers through rainwater pipelines following heavy rains, and the physical examination results of 3300 villagers in nearby villages showed that there were six leukemia and 31 cancer cases. Residents living in PAP areas mainly consume locally produced grains, which might contain levels of heavy metals that exceed the standard limits (Zhu et al., 2008; Wang et al., 2019). Consequently, heavy metal intake by residents exceeded the Provisional Tolerable Monthly Intake (PTMI) values recommended by FAO/WHO.

In Zhejiang, 349 g is the average daily per capita intake of rice (adult weight, 65 kg) (Zhu et al., 2017), and the Cd content of rice in the recycling area of electronic wastes is 0.449 mg kg^{-1} (Fu et al., 2013). Therefore, the Cd intake of nearby residents is 72.32 μg kg^{-1} body weight per month, which is 2.89-fold higher than PTMI (25 μg kg^{-1}) of JECFA. At the current rate of dietary Cd intake in China, on average it would take >100 years for the general population to reach a cumulative Cd intake of 2.6 g, which has been reported to cause severe Cd disease onset in 50.0% of Itai-Itai patients (Inaba et al., 2005). In contrast, in the recycling area of electronic waste, it would take only 46 years for the general population to reach a cumulative Cd intake of 2.6 g. This is a conservative estimate as it does not consider the dietary Cd intake from other grains. Furthermore, Nogawa et al. (2017) have shown that the lower limits of the benchmark dose of the Cd concentrations in rice for Itai-Itai disease or suspected disease are 0.27–0.56 and 0.62–0.76 mg kg^{-1} in women and men, respectively, based on a recent study in the Jinzu river basin of Japan. Cd has a long biological half-life (10–35 years) in the human body and the two main accumulating organs are the kidneys and liver (JECFA, 2011). Long-term consumption of Cd-contaminated rice can cause serious human diseases, including anemia, hypertension, neuralgia, soft bone, nephritis, secretion disorders, irreversible renal damage, and cancer (Åkesson et al., 2014; Wang et al., 2019).

3.4.2.2. Cancer villages. Villagers living in PAP areas for a long time face serious health consequences, including cancer (Hu et al., 2020b). For example, a survey on nitrate accumulation across vegetable production areas in Zhejiang showed that the over-standard rates of nitrate levels in vegetables in 2003 and 2004 were 23.8% and 16.5%, respectively (Lu et al., 2006). Nitrate can be reduced to nitrite by microbial activity or react with the amine in the stomach to produce nitrosamines, which have strong carcinogenic effects (Martinoia et al., 1981). A total of 24 cancer villages were mainly located in northern Zhejiang (Fig. 5). Nearly 80.0% of cancer villages were less than 3 km away from polluted rivers, and nearly 90.0% were less than 4 km away from polluted rivers. The probability of surface water meeting the national standard was very low in northern Zhejiang, ranging from 0.8% to 66.2% (Table S5). The results reveal that a high proportion of water was unsuitable for drinking or irrigation. Rural residents rely on untreated domestic water for drinking or grain produced by sewage irrigation for eating potentially contributed to cancer villages. It is noteworthy that approximately 90.0% of the administrative areas harboring these villages were situated in relatively developed areas, rather than in underdeveloped areas (Fig. S6).

Bivariate local indicators of spatial association characterized that the high-high distribution of cancer villages and PAP were mainly concentrated in northern Zhejiang (Fig. S8). This distribution pattern indicates that the impact of PAP on human health in northern Zhejiang is significantly higher than in southern Zhejiang. Moreover, comparing the distribution of grain production and cancer villages across different areas, we observed that cancer villages were mainly located in the high-yield areas of grain, especially in northern Zhejiang (Fig. 5). The directional distribution of cancer villages identified using Standard Deviational Ellipse in ArcGIS revealed the combined impact of various factors on the spread of cancer villages in the study area (Fig. 5). First of all, numerous chemical enterprises were located within the directional distribution (Fig. S5). Moreover, 20 serious incidents of PAP mainly occurred in the directional distribution (Fig. S7). In addition, the spatiotemporal distribution of centers of grain production (including rice, rapeseed, and fruit) and aquatic product output were shifting toward the directional distribution from 1997 to 2017 (Fig. 5). More importantly, PAP areas that required priority investigations were also mainly distributed in the directional distribution (Fig. 5). These results further emphasize the urgency for identifying priority investigation areas of PAP.

3.4.3. Integrated measures for addressing the impacts of PAP

Pollutants can enter the tables of families and restaurants through rice, fruits, and rapeseed grown in PAP areas (Augustsson et al., 2018). If crops are produced from polluted areas, it would be futile for humans to take any action to protect grain security and human health (Liu et al., 2013). To ensure grain security and human health, the government should immediately take measures to prevent PAP. The key question of how to identify priority investigation areas of PAP should be addressed first. In this regard, the proposed assessment framework for PAP provides a possible solution. In addition, a series of policy actions can help address the impact of PAP on grain security and human health in economically

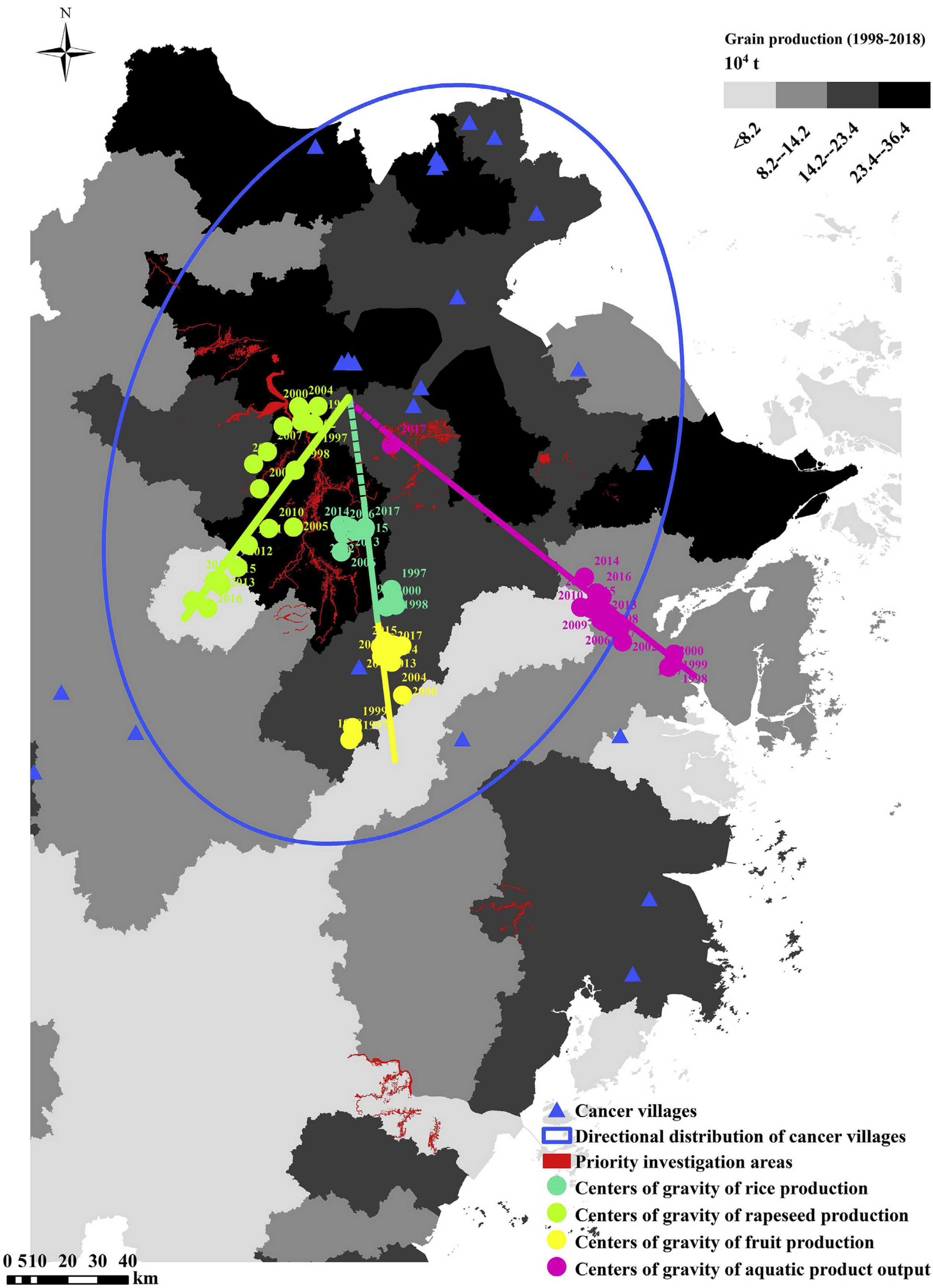

Fig. 5. Spatiotemporal distribution of grains production centers and directional distribution of cancer villages in Zhejiang, China. (Note: Grain production classification using natural breaks).

developed areas in China.

1) The first step in prevention of PAP is to identify and control the main pollution sources, which requires stricter monitoring of pollution sources and effective implementation of environmental protection laws (Brombal et al., 2015), especially in large emission sources such as mining, smelting, and chemical enterprises.
2) There are two alternative solutions to tackle PAP. First, planting low heavy metal accumulated crops or fiber plants represents a viable choice. However, this measure should be implemented only on heavily polluted soils to prevent a significant adverse impact on regional grain production and national grain security, which is a top priority of the Chinese government. Second, it is promising to use low-cost and environmentally friendly technology to clean up pollutants in polluted soil. It must be an efficient and low-cost technology suitable for the treatment of large-scale agricultural pollution because farmers cannot afford expensive investments or stop farming for many years.
3) Local governments should strengthen coordination and cooperation between different departments to reduce the intersection of pollution control work. In China, MEE, MNR, and MAA have their own systems and methods in the soil environment survey. Unfortunately, these departments do not share their survey data, which contributes to duplication and redundancy (Lu et al., 2015a). Therefore, a data-sharing platform should be established between different departments to generate a PAP assessment and provide strong support for agricultural pollution remediation.

4. Conclusions

The comprehensive framework proposed in this study provides a holistic approach to assess PAP in economically developed areas. The results indicate that PAP involved almost the entire study area, which would have a negative impact on food security and consequently human health. This framework can also be used in other places with similar problems to effectively identify PAP areas with high vulnerability, high hazard, or investigation priority. It can be further improved from the following perspectives: first, introducing more potential receptors or pollution sources; second, increasing the number of experts or integrate experts from different fields. These improvements are needed to transform the framework from a study prototype into a practical tool, which could guide regional authority in the ranking of PAP.

Declaration of competing interest

Authors have no conflict of interest to declare.

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CRediT authorship contribution statement

Yefeng Jiang: Conceptualization, Data curation, Formal analysis, Visualization, Writing - original draft, Writing - review & editing. **Songchao Chen:** Writing - review & editing. **Bifeng Hu:** Writing - review & editing. **Yin Zhou:** Project administration. **Zongzheng Liang:** Writing - original draft. **Xiaolin Jia:** Methodology. **Mingxiang Huang:** Investigation. **Jing Wei:** Writing - review & editing. **Zhou Shi:** Conceptualization, Methodology, Resources, Supervision.

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The high-resolution and high-quality PM2.5 data set in China (ChinaHighPM2.5 data set) are available at https://doi.org/10.5281/zenodo.3753614.

Appendix A. Supplementary data

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