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Predicting the probability distribution of Pb-increased lands in sewage-irrigated region: A case study in Beijing, China

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ABSTRACT

Assessment of pollution risk and spatial distribution for pollutant are important in the research of regional soil environmental quality. The paper provides a case of geostatistical method for predicting the probability of exceeding a Pb threshold and tracing the hotspots in sewage-irrigated land. The hotspots of Pb pollution which indicated higher risk than other areas were identified to be mainly distributed in the regions of soils irrigated with sewage. Sewage irrigation and parent material are the main sources for Pb concentration in soils of the study area, which obviously affect the distribution of Pb. The direction of high spatial continuity is consistent with the main direction of the river (Yongding River). Moreover, the Pb concentrations showed more extended transportation along the main direction of Yongding River. The results of disjunctive kriging showed that, if we are only willing to accept land for vegetable production if the probability that soil lead concentrations exceed the contamination threshold is 50% or more, then 6.2% of the study area (1932 km²) would be classified as unsafe for this purpose. That avoids the difficulty in selecting the proper criterion for the probability threshold.

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1. Introduction

Agricultural practices, such as irrigation using wastewater, insecticides, fertilizers and sewage sludge, are usually regarded as the main sources of heavy metals in agricultural soils (Chen et al., 1997; Vidal et al., 2004; Huang et al., 2007). As one of the most toxic elements to human health, Pb has attracted increasing amount of researchers (Mirlean et al., 2005; Hooker and Nathanail, 2006). Unhealthy effects caused by Pb include neurological impairment and deficits in the functioning of the central nervous system (Counter et al., 2005; Toscano and Guilarte, 2005). Therefore, many surveys which mainly focused on specific locations such as industrial sites, parks and soils along roadside, and soil Pb concentrations have been conducted in different countries (Ferreira et al., 2004; Chen et al., 2005; Cloquet et al., 2006).

Due to water shortage in arid and semi-arid regions, soils in parts of these regions are irrigated with sewage, which may have both positive and negative consequences (Yadav et al., 2002). The positive aspect of wastewater irrigation is that nutrients and organic matter are provided to soils (Horswell et al., 2003). However, as a result of long-term irrigation with domestic sewage or industrial effluent, the Pb concentrations in soils of the irrigated area were found significantly

higher than the background values (Bourennane et al., 2006; Dere et al., 2007; Rodriguez et al., 2008).

Geostatistics is based on the theory of a regionalized variable which is distributed in space and shows spatial autocorrelation (Webster and Oliver, 2001). One of the most important applications of geostatistics on environmental geochemical data is to provide an advanced methodology which facilitates quantification of the spatial features of soil parameters and enables spatial interpolation. In addition, geostatistics has become a useful tool for the study of spatial uncertainty and hazard assessment (McGrath et al., 2004; Rawlins et al., 2005), which is increasingly being used in studies for contaminated sites in recent years (Steiger et al., 1996; Juang and Lee, 1998; Lee et al., 2006).

The objective of this study demonstrated a case in predicting the probability of exceeding a Pb threshold and tracing the hotspots of study area with geostatistical method.

2. Materials and methods

2.1. Description of the study area

The study area of Tongzhou and Daxing Districts was located in the Southeast of Beijing Municipality with an average annual precipitation of about 620 mm, where several rivers form a fan-shaped alluvial plain. The main soil types are fluvo-aquic soil (Calcaric Cambisol) and cinnamon soil (Anthrosol). Most of the agricultural soils had been

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irrigated with sewage from Beijing city since the 1960s (Yang et al., 2005). There are three distinct irrigation areas as follows: Yongding River Irrigation Area, Liangshui River Irrigation Area, and Fenggang River Irrigation Area (Fig. 1). The Liangshui River received treated or untreated sewage from Gaobeidian Wastewater Treatment Plant; The Yongding River mainly received mixed domestic and industrial wastewater discharged from the Shijingshan District; The Fenggang River is a natural river, and soils in the Fenggang River Irrigation Area are mainly irrigated with rainfall or groundwater. The soil pH of the study area was 7.8–8.6. The study area was the major agricultural area and largest sewage-irrigated area in Beijing, with its dominant crops such as corn and wheat.

2.2. Sampling and chemical analyses

A total of 360 surface soil samples (0–20 cm in depth) were taken from the study area (Fig. 1). The sampling density was approximately 1 sample per 0.56 km². The coordinates for each sampling location were recorded using a global positioning system (GPS) receiver.

About 1 kg of each sample was collected using a stainless steel spade. It was air-dried, grounded, screened through a sieve of 2 mm meshes size, and then digested with HNO₃ and H₂O₂ under the method of 3050B recommended by USEPA (USEPA, 1996). The concentration of Pb was analyzed with a flame atomic absorption spectrometer (Vario 6, Jena Co., Ltd., Germany). The standard reference material of GSS-1 for soils, obtained from the Center of National Standard Reference Material of China, was applied for quality assurance and quality control (QA/QC) procedures. The ratio between the measured and reference value of Pb varied between 95 and 108%.

2.3. Geostatistics

In order to reduce the cost of sampling and analysis for mapping pollutants, a spatial interpolation technique called kriging is widely applied in soil studies. It uses the semi-variogram to quantify the spatial variation of a regionalized variable. The fitted function to the experimental variogram provides the input parameters for spatial prediction by kriging (Krige, 1951).

For observations (e.g. soil samples) made in a study area $Z(x)$, $x=1, 2, \dots, n$, the relationship between pairs of observations located at h interval apart, the lag distance, can be expressed as the variance of the differences between all such pairs (Cliff and Ord, 1981; Goovaerts, 1999):

$$\gamma(h) = \frac{1}{2} E[Z(x) - Z(x+h)]^2 \tag{1}$$

This function is called the semi-variogram, and it is a measure of the differences, on average, between points at a given distance h apart. The more alike the points are, the smaller $r(h)$ is. For data collected at discrete sampling locations, the following equation is generally used:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i+h)]^2 \tag{2}$$

where $Z(x_i)$ is the value of variable Z at the location of x_i , $Z(x_i+h)$ is the value of Z located at a lag distance of h apart from x_i , and $N(h)$ is the total number of sample pairs separated by lag distance h .

Disjunctive kriging is the principal technique to estimate the probability that the true values at an unsampled location exceeding a specified threshold. It is based on the assumption that data are a realization of a process with a second-order stationary bivariate distribution. To obtain the DK estimator, the original data must be transformed into a new variable, $Y(x)$, with a standard normal distribution where pairs of sample values are bivariate normal.

$$\phi[Y(x)] = Z(x) = \sum_{k=0}^{\infty} C_k H_k[Y(x)] \tag{3}$$

The value for $Y(x)$ is obtained by taking the inverse, $Y(x) = \phi^{-1}[Z(x)]$; $H_k[Y(x)]$ is a Hermite polynomial of order k , and the C_k is the Hermite coefficients.

$$Z_{DK}^*(x_0) = \sum_{k=0}^k C_k H_k^*[Y(x_0)] \tag{4}$$

$$H_k^*[Y(x_0)] = \sum_{i=1}^n b_{ik} H_k[Y(x_i)] \tag{5}$$

where the $H_k^*[Y(x_0)]$ represents the estimated value of the k th Hermite polynomial at the estimation site. The b_{ik} are the DK weights and the series in Eq. (5) is truncated to k terms. The sum of these estimated multiplied by the coefficient, C_k makes up the DK estimated at x_0 . To obtain an estimated value for the Hermite polynomial, the DK weights (b_{ik}) must be found by solving the linear kriging equation for each k

$$\sum_{i=1}^n b_{ik} (\rho_{ij})^k = (\rho_{oj})^k; j = 1, 2, 3 \dots n. \tag{6}$$

The conditional probability is obtained by defining an indicator variable that is equal to unity if $Y(x_i) \geq y_0$ and zero otherwise. This allows the conditional probability to be written in terms of the conditional expectation and gives the estimator of the conditional probability as

$$P_{DK}^*(x_0) = 1 - G(y_c) + g(y_c) \sum_{k=1}^K H_{k-1}(y_c) H_k^*[Y(x_0)] / k! \tag{7}$$

where $G(y_c)$, $g(y_c)$ are the cumulative and probability density functions for a standard normal variable respectively.

In this study, the fitted exponential model was selected. The exponential function is:

$$\gamma(h) = C_0 + C \left(1 - e^{-(h/a)}\right) \quad h > 0 \tag{8}$$

where C_0 is the nugget variance ($h=0$), it represents the experimental error and field variation within the minimum sampling spacing. Typically, the semivariance increases with increasing lag distances to approach or attain a maximum value or sill ($C_0 + C$) equivalent to the

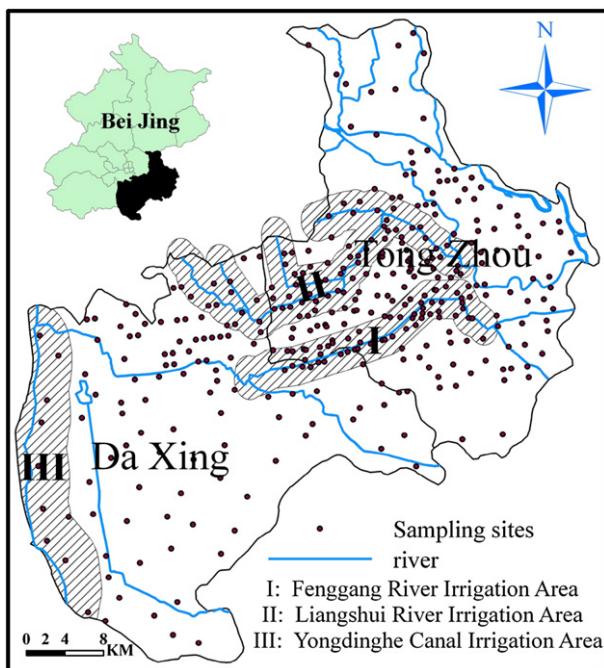


Fig. 1. The sketch map showing the sampling sites in Tongzhou–Daxing irrigation areas, Beijing, China.

Table 1
Comparison between Pb concentrations in the soils of Tongzhou–Daxing irrigation areas and the background concentration of soil Pb in Beijing

	No. of samples	Pb concentration (mg kg ⁻¹)				
		Min.	Max.	Arithmetic mean	Standard deviation	Geometric mean
All samples in the study	360	12.5	58.2	28.7	7.03	27.4
Background concentrations of Pb in soils of Beijing*	101	11.5	38.2	25.1	5.08	24.6

*Chen et al. (2004).

population variance. C is the structural variance and a is the spatial range across which the data exhibit spatial correlation.

3. Results and discussion

3.1. Pb concentrations in soils of the study area

The Pb concentrations in soils of the study area follows a lognormal distribution with the specific significance value 0.269 and the skewness and kurtosis values were -0.059 and 0.977, respectively. The Pb concentrations in soils of the study area varied between 12.5 and 58.2 mg kg⁻¹. Compared with the background level (25.1 mg kg⁻¹) of soil Pb in Beijing (Chen et al., 2004), the Pb concentrations (log-transformed data) of study area were significantly higher than the reference value (log-transformed data) based on one-sample t test ($P < 0.01$) (Table 1).

Baseline concentration measures an expected range of elemental concentrations around a mean and is defined as 95% of the expected range of background concentrations. Based on the normal distribution theory, the expected range can be expressed as the average ± 2 standard deviation (AVG $\pm 2SD$) (Dudka et al., 1995). To obtain a better understanding of the suspected Pb pollution in the study area, the baseline value of soil in Beijing is used for comparison, and it was found that 51 samples had Pb concentrations exceeding the baseline value (35.3 mg kg⁻¹). Therefore, Pb was obviously enriched in some parts of soils of the study area.

3.2. Spatial structure analysis of Pb in the soils

Since the Pb concentrations data do not follow normal distribution pattern, the normal score transformation was applied to turn raw data into Gaussian values before geostatistical analysis (Emery, 2006). Omnidirectional, together with directional semi-variograms along four standard directions and four secondary directions were calculated by formula (1). It was found that exponential models with zonal anisotropy fitted well to the calculated experimental variograms of Pb. Key parameters of suitable models, including nugget value C_0 , sill ($C_0 + C$), and range value a , were compared (Table 2) and analyzed to clarify differences in spatial structures.

Low nugget/sill ratio indicates high spatial autocorrelation or spatial continuity over short distances. The high spatial continuity were observed in N22.5°W \pm 22.5° direction, and relatively poor correlations

Table 2
Key parameters of fitted exponential models of Pb in soils of the study area

Direction	C_0	C	$C_0 + C$	$C / C_0 + C$	Range(km)	Model
N90°E	0.43	0.52	0.95	0.45	22.8	Exponential
N67.5°E	0.44	0.54	0.98	0.45	18.9	Exponential
N45°E	0.47	0.5	0.97	0.48	17.4	Exponential
N22.5°E	0.48	0.49	0.97	0.49	17.1	Exponential
N0°W	0.43	0.69	1.12	0.38	27.3	Exponential
N22.5°W	0.4	0.76	1.16	0.34	21.7	Exponential
N45°W	0.25	0.94	1.19	0.21	21.3	Exponential
N67.5°W	0.49	0.57	1.06	0.46	19.8	Exponential
Omnidirectional	0.47	0.56	1.03	0.46	23.3	Exponential

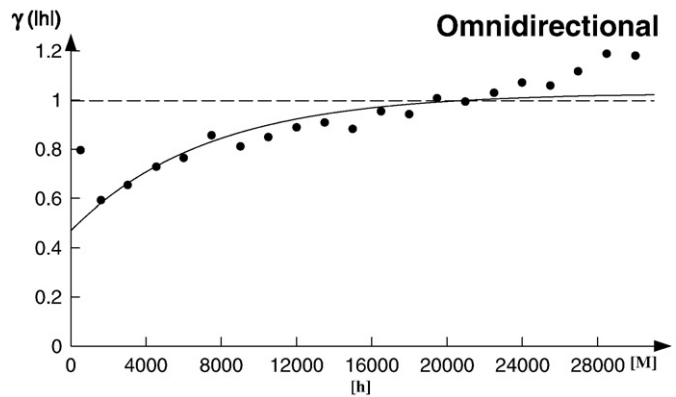


Fig. 2. Omnidirectional variogram model of Pb concentrations in the soils of Tongzhou–Daxing irrigation areas, Beijing, China.

were in N67.5°E \pm 22.5° direction. The Pb concentrations in soils of the study area showed the lowest ranges in the N45°E direction and the highest ranges in the N0°W direction, along which there may be relatively more extended transportations. Anisotropy is significant for the Pb concentrations in soils of the study area. It is not occasional that the main direction of Yongding River in Daxing district is consistent with the direction of best correlation. Moreover, the Pb concentrations showed more extended transportation along the main direction of Yong Ding River. The Pb concentrations in soils of the study area were the result from agricultural activity and parent material, which can be characterized with randomness and spatial continuity. The land of the study area ever irrigated with wastewater from rivers usually accumulated abundant heavy metals. Wastewater irrigation could contribute to the spatial structure of Pb in soils of the study area.

3.3. The distribution of Pb in soils of the study area

The disjunctive kriging, according to the omnidirectional exponential model (Fig. 2), was adapted to map the concentrations of Pb in soils of the study area. In the map of Fig. 3, the ranges of Pb concentrations were decided with reference to the background Pb concentrations in soils in Beijing Municipality. Four distinct levels with different shading and class-

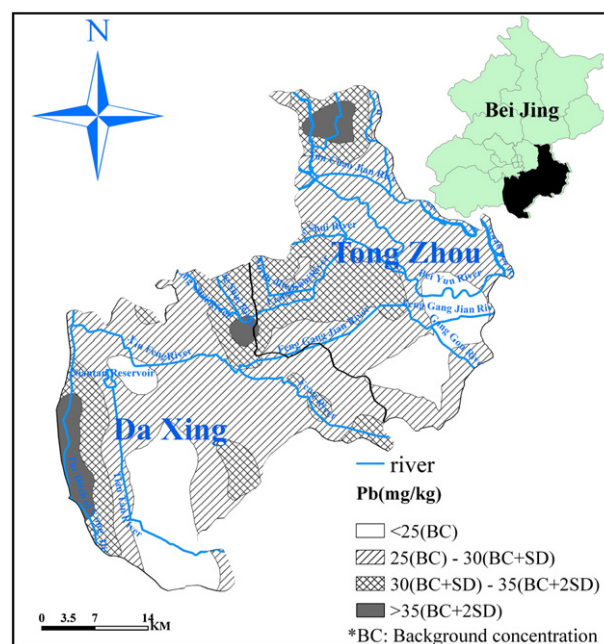


Fig. 3. Spatial distribution map of Pb concentrations in the soils of Tongzhou–Daxing irrigation areas, Beijing, China.

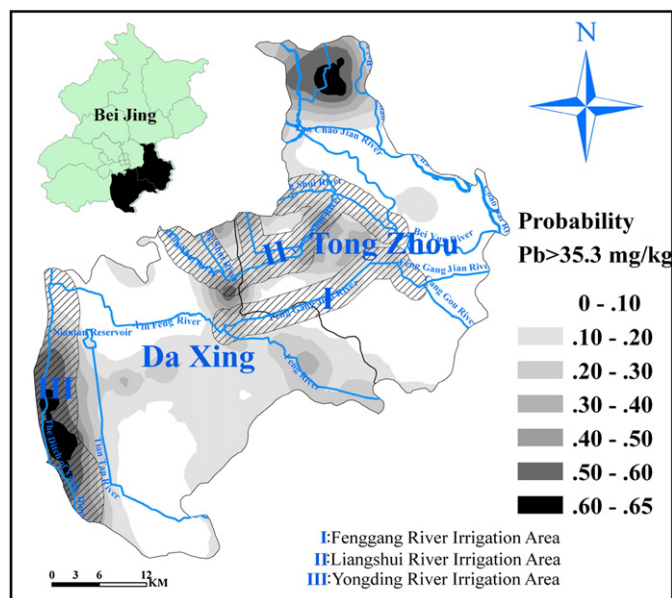


Fig. 4. The map of estimated probability distribution of Pb pollution in soils of Tongzhou–Daxing irrigation areas, Beijing, China.

intervals were decided based on the background concentration, standard deviation and taking the lowest and highest values into consideration. In the South of Daxing District and Southeast of Tongzhou District, Pb concentrations were below the background value (25.1 mg kg^{-1}) of Beijing soils. High concentrations of Pb were observed in the west of Daxing District, the middle-north and northeast of Tongzhou District.

The lands of the study area usually irrigate with water from nearby river. An investigation in 1980s indicated that sediments from the Liangshui River showed higher Pb concentrations, with 185 mg kg^{-1} , than that of the Fenggang River (SGPICSB, 1980). Therefore, the Pb concentrations in soils of the Liangshui River Irrigation Area were significantly higher ($P < 0.05$) than those in soils of the Fenggang River Irrigation Area irrigated with rainfall or ground water. Steel plant of Beijing and other factories in Shijingshan District in the upper reaches of the Yongding River were possible sources for Pb pollution in the soils of the west part of Daxing District. These factories discharged their effluents into the Yongding River, which was used for irrigation in the

west Daxing District partly. High concentration of Pb was usually found in industrial wastewater (Mapanda et al., 2005). Therefore, the Pb concentrations in the soils of Yongding River Irrigation Area were significantly higher ($P < 0.05$) than other irrigation areas.

It seems that sewage irrigation is a source for the Pb concentrations in soils of the study area. The result of spatial structure analysis also shows that sewage irrigation plays an important role for the Pb distribution of the study area. However, the Pb concentrations in some areas of the study area were still under control and comparable with the Pb background value, which indicates that parent material is another source to control the distribution of Pb in soils of study area. There were relatively high Pb in the soils of northeast of Tongzhou District, and such isolated pollution hotspots may imply local pollution sources that need further investigation.

3.4. The probability of exceeding Pb baseline value

Before an agency attempts to reclaim land that appears to be contaminated, it may wish to know the likelihood that the true concentrations in the soils have exceeded the threshold. It will not want to spend money unnecessarily on reclamation (Yates and Yates, 1988; Oliver et al., 1998). The probability of Pb concentrations exceeding the baseline value (35.3 mg kg^{-1}) was estimated by disjunctive kriging (Fig. 4). It was shown that the areas with high risk were mainly located in the west of Daxing District, and northeast of Tongzhou District, with the estimated probability reaching 0.6–0.65.

Any probability threshold p_c can be considered in decision-making. For example, if we would consider the 50% probability of exceeding contaminated threshold as a critical probability level p_c , then this would result in a classification of 6.2% of the study area (covering 1932 km^2) as unsafe to be used for vegetable where Pb concentrations exceeds the contaminated threshold. On the other hand, other field can be regarded as safe for vegetable growth. That avoids the difficulty in selecting proper criterion for the probability threshold.

3.5. The accuracy of the interpolation assessed by cross-validation

By excluding the measured value from the kriging system, the estimated value can be compared with the measured one in the process of cross-validation and the accuracy of the spatial interpolation can be evaluated for those values at sampled locations. It is clear that the estimated values with large errors detected by cross-validation were found for the values lower than 5th percentile (Table 3), especially at locations where Pb concentrations contrasted with other nearby locations. Actually, the large errors are expected as the results of smoothing effect of the kriging procedure (Tao, 1995). The models used for kriging were constructed based on the data structure over the whole study area, and thus over-estimation or under-estimation could not be avoided. However, the cross-validation showed a minimum relative error of 0.1% and maximum relative error of 72.9% for the large proportion of the data set (between 5th percentile and 95th percentile) with an average relative error of 11.1%.

Table 3
The estimated relative errors of Pb concentrations between measured values and estimated values by cross-validation in the Tongzhou–Daxing irrigation areas

Probability distribution	Number of sample points with relative error						Relative error (%)		
	≤0.1	≤0.25	≤0.5	≤0.75	≤1	>1	Min.	Max.	Box–Cox mean
<5 th percentile	0	2	8	12	14	5	15.9	121	60.7
5 th –95 th percentile	159	273	321	324	324	0	0.1	72.9	11.1
>95 th percentile	0	1	17	17	17	0	9.8	50.3	36.3

Table 4
Geostatistical surveys of Pb concentrations in soil

Location	Land use	No. of samples	Area (km ²)	Range	Mean	Method	Variogram model	Nugget/Sill	Range (km)	Reference
Tongzhou and Daxing Districts, Beijing	Complex	360	1932			Disjunctive kriging	Exponential	0.44	23.3	This study
Hangzhou–Jiaxing–Huzhou Plain, China	Rural soil	450	6391	1.1–85.9	8.51	Disjunctive kriging	Exponential	0.29	13.5	Liu et al. (2006)
Weinfield, Switzerland	Rural soil	204	50	6.7–110	23.3	Disjunctive kriging	Pentaspheical	0.16	1.29	Steiger et al., 1996
Ebro river basin, Spain	Rural soil	624	9300	4–61	17	–	–	–	–	Rodriguez et al. (2008)
Pierrelaye–Bessancourt, Paris	Wastewater irrigated fields	87	0.15	118–649	321.6	Ordinary kriging	Spherical	0.40	0.1	Bourennane et al., 2006
Mezquiyutal Valley, Mexico	Wastewater irrigated fields	16	–	–	49.5	–	–	–	–	Ortega-Larrocea et al. (2001)

3.6. Geostatistical surveys of Pb concentrations among different regions

The Pb concentrations in soils of the study area were higher than the value of rural soil and lower than the values of raw wastewater irrigated fields (Table 4). The key parameters of semi-variogram for Pb in soils of the study area were different with the result of other regions. Notably, the distribution of Pb pollution in soils of the study area were the result from sludge irrigation and parent material; the sources of soil Pb in Pierrelaye-Bessancourt, Paris and Mezquital Valley, Mexico mainly come from raw wastewater irrigation (Bourennane et al., 2006; Ortega-Larrocea et al., 2001). However, it is difficult to compare the Pb concentrations among different regions owing to divergent sources. Moreover, the different mensuration leads to different analysis results. Different sources for pollutant show various spatial features, meanwhile, the research scales have obvious effects on the characters of spatial structure for pollutants.

4. Conclusion

Wastewater irrigation and parent material are the main sources for Pb concentration in soils of the study area, which obviously affect the distribution of Pb. The results of disjunctive kriging showed that, if we are only willing to accept land for vegetable production if the probability that soil lead concentrations exceed the contamination threshold is 50% or more, and then 6.2% of the study area (1932 km²) would be classified as unsafe for this purpose.

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