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Groundwater Contamination Site Identification Based on Machine Learning: A Case Study of Gas Stations in China

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Abstract: Accurately identifying groundwater contamination sites is vital for groundwater protection and restoration. This study aims to use a machine learning (ML) approach to identify groundwater contamination sites with total petroleum hydrocarbons (TPH) as target contaminants in a case study of gas stations in China. Firstly, six classical ML algorithms, including logistic regression, decision tree, gradient boosting decision tree (GBDT), random forest, multi-layer perceptron, and support vector machine, were applied to develop the identification models of TPH-contaminated groundwater with 40 features and the performances were compared. The comparison results showed that the GBDT model achieves the best prediction performance, with F1 score of 1 and AUC value of 1. Next, Bayesian optimization optimized GBDT (BO-GBDT) was conducted to further decrease the training time from 19,125 s to 513 s while maintaining the same prediction performance (F1 score = 1, AUC = 1). Finally, Shapley additive explanations (SHAP) analysis was performed on the BO-GBDT model. The SHAP results displayed that the critical feature variables in the BO-GBDT model include wind, population, evaporation, total potassium in the soil, precipitation, and leakage accident. This study demonstrated that BO-GBDT is one satisfactory model to identify groundwater TPH-contamination at gas stations. The method proposed in this study has the potential to be applied to other types of groundwater contamination sites.

Keywords: machine learning; groundwater contamination site; gas station; gradient boosting decision tree; Bayesian optimization; Shapley additive explanations



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1. Introduction

The identification of potentially contaminated sites is crucial for the management of contaminated sites and the protection of public health [1]. Given the mobility of groundwater and the difficult-to-repair nature of contamination, the deterioration of groundwater quality due to point source pollution, represented by various industrial and commercial sites, is attracting widespread concern [2,3]. There are over 2.5 million potentially contaminated sites in Europe [3]. In Asia and the USA, new contamination sites are being identified every day [4]. China is estimated to have over 500,000 contaminated sites [5]. Therefore, identifying groundwater contamination sites has become a research hotspot [6].

Current methods for identifying groundwater contamination sites include site history review, field survey and sampling analysis, and interviews [6]. Site history review refers to the search for documents relating to original buildings on the site from environmental and health agencies, associations, unions, and so on [7]. However, historical documents

are not always available, especially for sites with long histories. In addition, this method is time-consuming for a large number of sites. Field surveys and sampling analysis have the advantage of maximizing the accuracy of the findings. However, each site requires multiple samples to be collected for laboratory analysis and therefore requires a significant investment of workforce, money, and time [8]. In the interview method, information about the site is obtained by interviewing the site manager, owner, government personnel, and residents around the site to judge the site's contamination. The drawback of this method is its subjectivity, which may be related to the gender, age, profession, and standpoint of the interviewees, and has a high degree of uncertainty [9–11].

Data science methods, represented by machine learning, have attracted significant attention in recent years. The primary purpose of machine learning is to evaluate or predict objectives after training a model with data in specific conditions. The advantage of machine learning is that hidden associations in the data can be learned through automatic mathematical analysis [12]. Currently, many machine learning algorithms are applied to solve groundwater-related problems. Six of these algorithms have been reported to be widely applied, including Logistic regression (LR), decision tree (DT), gradient boosting decision tree (GBDT), random forest (RF), multi-layer perceptron (MLP), and support vector machine (SVM). For example, in the Saladin Province of Iraq, LR was applied to assess groundwater nitrate contamination levels [13]. A DT model was developed to classify groundwater quality in the Ardebil aquifer, Northwest Iran [14]. Spatially distributed feature variables related to arsenic and manganese flows were selected for the GBDT model to obtain areas of high arsenic and manganese in the northern USA [15]. RF, LR, and MLP algorithms were used to model fluoride in groundwater in Datong Basin, China [16]. Four models, including MLP and SVM, were developed to estimate groundwater TDS in the East Azerbaijan Province [17]. Recent researches on identifying groundwater contamination using machine learning mainly focused on the identification of contamination source or intensity in a specific contaminated site. For example, self-organizing map algorithm was applied to identify the source of sulfate in groundwater of an abandoned mine [18]; the Bayesian and machine learning method was used to invert the contamination source release intensity and duration in a hypothetical case study [19]; the deep learning algorithms based on Long-Short Term Memory networks were utilized to identify the contamination source, initial release period, and release intensity in a hypothetical case study with irregular boundaries [20]. However, to our knowledge, few studies have been reported on using machine learning methods to identify potential groundwater contamination sites, especially hybrid machine learning models. Moreover, only a few studies have shown concern for explanatory analyses of the results.

Due to leaks from storage tanks or pipelines, groundwater contamination at gas stations is attracting significant attention worldwide [21]. Relevant reports indicated there might be about 242,000 leaking gas station storage tanks in the EU with contamination from leaks [22]. Four hundred ninety-five thousand leaking gas station storage tanks have been reported in the United States until 2010 [23]. There are 27,000 gas stations in Brazil with possible soil and groundwater contamination from oil spills [24]. There are over 120,000 gas stations in China, located in commercial and residential areas, urban traffic arteries, and highways; some are even in water sources and nature reserves. Total petroleum hydrocarbon (TPH) is one of the primary contaminants in groundwater from gas stations [22,25]. Aromatic hydrocarbons in TPH are carcinogenic, teratogenic, and mutagenic [26]. The China Geological Survey conducted a survey of some gas stations in Tianjin province. The results showed that the detection rate of TPH in groundwater samples was 85%, and the exceedance samples accounted for 40% of the total groundwater samples. Therefore, TPH was set as the target contaminant in this study, and Chinese gas station sites were set as the study area.

This study aimed to develop machine learning models based on multi-source data to identify the current status of groundwater TPH contamination under gas stations in China. LR, DT, GBDT, RF, MLP, and SVM were determined as the machine learning

algorithms used in this study. Forty feature variables from the field survey and database were considered as input variables for machine learning to train and test the models. The optimization effect of Bayesian optimization (BO) was also examined, and the importance of feature variables was discussed using the Shapley additive explanations (SHAP) method.

2. Materials and Methods

2.1. Methodology

As shown in Figure 1, there are three steps in the framework for developing models for groundwater contamination site identification. Firstly, data collection was conducted simultaneously from the field survey and database. Subsequently, six different machine learning models were built based on a training dataset. Three approaches (original dataset, min-max scaler, standard scaler) were used for data pre-processing. The model with the optimal result was selected according to model evaluation and was compared with the Bayesian optimization (BO) hybrid model to determine a better one. Finally, the feature variables were analyzed using the SHAP method to obtain the key variables. These key variables were combined with the optimal model obtained in step 2 to obtain the most satisfying model to identify groundwater TPH contamination sites.

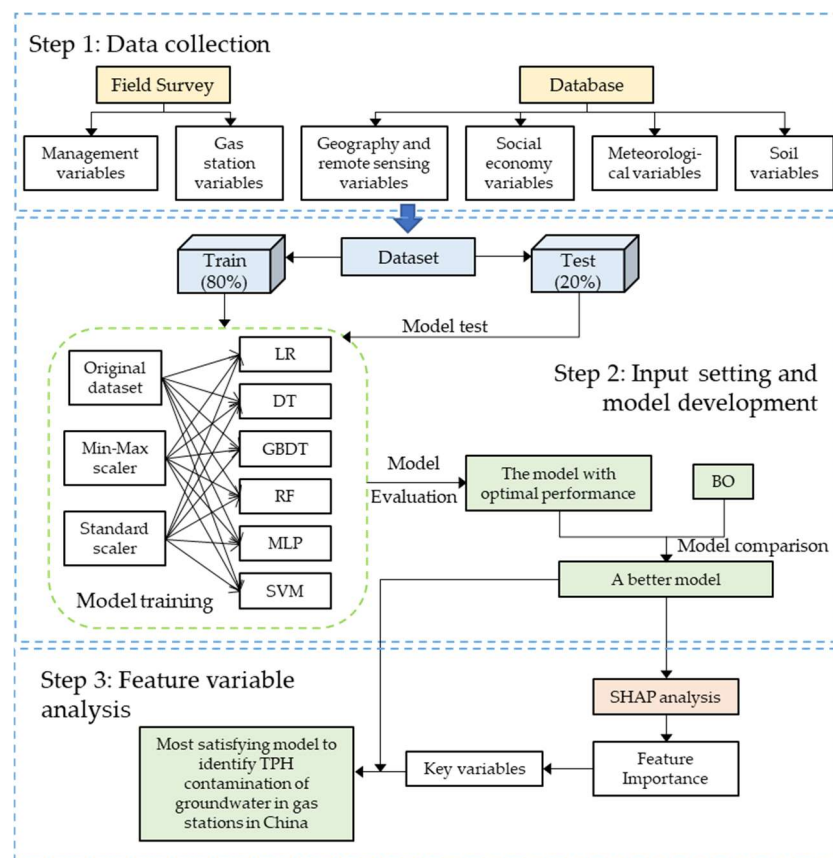


Figure 1. Methodological framework in this study.

2.2. Data

Field surveys and groundwater sampling were conducted at 103 gas stations in 24 regions of China following the requirements of the technical guidelines (HJ 25.1-2014) issued by the Ministry of Ecology and Environment of China. The sampling was carried out from 2017 to 2019. Sample collection, storage, transportation, and testing were determined according to guides issued by the Chinese authorities [27–29]. The groundwater TPH contamination of 103 gas stations was finally obtained as the target variable for machine learning in this

study. Eighty-three gas stations belonged to the “uncontaminated” category, while the other twenty belonged to the “contaminated” category.

The factors affecting TPH contamination of groundwater at gas stations were considered, and the available information was aggregated into the feature variables dataset for this study. The selection of feature variables is based firstly on authenticity, accessibility, completeness, and diversity of the data. The feature variables include six categories: management variables, gas station variables, geography and remote sensing variables, social economy variables, meteorological variables, and soil variables, with a total of 40 variables. All variables may reflect some association with groundwater contamination. Key investigation list (KIL), a management variable, reflected a greater risk of contamination around the gas station. The construction time, a gas station variable, was associated with groundwater contamination at the gas station. The distance to the nearest lake (dist_lake) and distance to the nearest river (dist_river) in the geographic and remote sensing variables might influence the status of groundwater recharge and runoff. Among the social economy variables, the night light index (NLI), population, and GDP correspond to the intensity of human activity, which might be a source of anthropogenic pollution of groundwater. Meteorological variables may influence groundwater recharge and discharge; for example, precipitation is usually positively correlated with vertical recharge and annual sunshine affects groundwater in many aspects such as precipitation and evaporation. The soil related variables are also included, considering the closest contact between soil medium and groundwater. To sum up, these variables come from different sources and reflect the role of the natural environment and human activities and are therefore included. Table 1 shows detailed information on these feature variables, including category, name, description, and type.

Table 1. Description of feature variables in this study.

Category	Variable	Description	Type
Management variables	KIL	Key investigation list. Whether the station is on the key investigation list or not	categorical
	OC	Operating condition. Whether the station is open for business	categorical
	Owner	The owner of the gas station	categorical
	LA	Leakage accident. Whether the station has ever had oil leakage accidents	categorical
	GWPA	Groundwater protection area. Whether the station is located in a groundwater protection area	categorical
Gas station variables	Construction Time	The length of time since the station was constructed	discrete
	No.Tanks	Total number of tanks at the gas station	discrete
	No.SingleTanks	Total number of single-layer tanks at the gas station	discrete
	Impermeable ponds	Whether the gas station has built impermeable ponds	categorical
Geography and remote sensing variables	Pipeline	Type of pipeline at the station	categorical
	Dist_lake	The distance from the nearest lake	continuous
	Dist_river	The distance from the nearest river	continuous
	Elevation	The elevation where the gas station is located	continuous
	NPP	Net primary productivity at the location of the gas station	continuous
	LAI	Leaf area index. The leaf area index of the location of the gas station	continuous
	Landuse	Land use types around gas stations	categorical
Social economy variables	NDVI	Normalized vegetation index of the gas station location	continuous
	NLI	Night light index. The night light index of the gas station location	continuous
	population	The population of the town where the gas station is located	continuous
	GDP	Total GDP of the cell grid where the gas station is located	continuous

Table 1. Cont.

Category	Variable	Description	Type
Meteorological variables	Permafrost type	The type of frozen soil	categorical
	EVP	Annual mean evaporation	continuous
	GST	Annual mean ground surface temperature	continuous
	PRE	Annual precipitation	continuous
	PRS	Annual mean pressure	continuous
	RHU	Annual mean relative humidity	continuous
	SSD	Annual sunshine	continuous
	TEM	Annual mean temperature	continuous
	WIN	Annual mean wind speed	continuous
Soil variables	Soil erosion	Types and properties of external forces of soil erosion	categorical
	clay_top	The proportion of clay in the topsoil (0–30 cm)	continuous
	sand_top	The proportion of sand in the topsoil	continuous
	silt_top	The proportion of silt in the topsoil	continuous
	soil moisture	The moisture of the topsoil	continuous
	clay_sub	The content of clay in the subsoil (30–100 cm)	continuous
	sand_sub	The content of sand in the subsoil	continuous
	CEC_S	Soil cation exchange capacity in 100–200 cm	continuous
	TK_S	Total potassium in soil from 15 to 30 cm	continuous
	gravel_S	Gravel content in soil from 30 to 60 cm	continuous
	pH_S	Soil pH in the range of 30–60 cm	continuous

2.3. Machine Learning Approach

2.3.1. Models

Here, we briefly introduce the basic principles of the six algorithms (LR, DT, GBDT, RF, MLP, SVM), the detailed principles of which can be found in the attached reference. LR is one of the most classical statistical machine learning models. LR can make categorical predictions from several selected categorical, discrete, or continuous variables and is a suitable technique for predicting binary outcomes [30,31]. Based on a logistic function, LR maps the linear regression results to the interval [0, 1] [32]. DT is a machine learning algorithm based on a tree structure that can reveal complex relationships that are difficult to identify with linear statistical models [33]. The tree includes the root, internal, and leaf nodes. During training, the role of the nodes is to test and assign the feature variable for each input until it reaches the leaf nodes. Each leaf node represents a class of instances [34]; for example, this study represents two classes of TPH out of threshold or not out of threshold. GBDT is an ensemble learning algorithm based on DT consisting of multiple trees. Therefore, it combines crucial advantages of the DT algorithm [35]. The basic idea of GBDT is boosting mechanism. A new tree is trained with the original feature variables in each iteration. This new tree will fit the residuals left by the previous tree, thus continuously reducing the model error [36]. Therefore, the number of trees ($n_{\text{estimators}}$) is one of the critical hyperparameters of the algorithm. RF is another ensemble learning algorithm based on DT that has been widely modeled in recent years [37]. Unlike GBDT, the idea of RF is bagging. In RF, each tree is trained using a different part of the same training set, meaning each tree learns different samples. A vote of all trees generates the final result of RF. This sampling mechanism allows RF to circumvent the effects of noise in the data [38]. MLP, also known as artificial neural networks, has a long history in groundwater modeling [39]. The MLP model consists of an input, hidden, and output layer. Neurons characterized by weights and biases interconnect these layers. The input layer is to input data, and the output layer is the target of the model prediction. An activation function is embedded in the hidden layer to transform the deviation-weighted summation of the layer to the next layer until it reaches the output layer [40]. Therefore, selecting an appropriate activation function plays an important role in the training of the MLP. In addition, the number of layers and neurons in each hidden layer constitute the hidden layer size, and experiments are needed to obtain the optimal hyperparameter settings. Finally, SVMs are a class of

generalized linear classifiers that perform binary data classification in a supervised learning manner. In the binary classification problem investigated in this study, SVMs attempt to locate a hyperplane in the n-dimensional input space that can separate the data points into two distinct classes. The boundaries of this hyperplane (called decision boundaries) also have to be as large as possible to make reliable classification predictions [41].

The computer configuration used to perform the experiments was CPU Core i7-6820HQ, 48 GB of RAM, and Windows 10 as the operating system. We used the Scikit-learn toolkit in Python to perform machine learning modeling (<https://scikit-learn.org/stable/index.html> (accessed on 3 January 2023)). The GridSearchCV module in Scikit-learn was called to determine the best hyperparameters for different algorithms and optimize the model's performance. We first identified the critical hyperparameters of each algorithm and the search scope of GridSearchCV through a literature review in conjunction with the problem of this study. GridSearchCV performs an exhaustive search for a specified machine learning algorithm within a hyperparameter search range with an exhaustive search. Afterward, GridSearchCV uses the adjusted hyperparameters to train the learner to find the most accurate parameter in the validation set from all parameters. Here, the validation set is generated by cross-validation with $k = 10$. The descriptions and ranges of the critical hyperparameters for the different algorithms are shown in Table 2.

Table 2. Hyperparameter settings [42–46].

Algorithm	Hyperparameter	Description	Range
LR	C	Regularization parameter. A smaller C means that the model may have better generalization, but it is also more likely to underfit	[0.0001, 0.001, 0.01, 0.1, 1, 10, 100, 1000, 10,000]
DT	criterion	Selects the criteria by which attributes will be selected for separation	[gini, entropy, log_loss]
	min_samples_leaf	The minimum number of samples required at a leaf node	[1, 2, 3, 4, 5, 6, 7, 8, 9, 10]
	max_depth	The maximum depth of the tree	[3, 4, 5, 6]
	min_samples_split	The minimum number of samples required to split internal nodes	[0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.2]
GBDT	criterion	Selects the criteria by which attributes will be selected for separation	[friedman_mse, squared_error]
	n_estimators	The number of boosting stages to perform. It is represented in GBDT as the number of decision trees	[10, 25, 50, 60, 70, 80, 90, 100, 110, 120, 130, 140, 150, 160, 170, 180, 190, 200, 500, 1000, 1500, 2000]
	DT hyperparameters	min_samples_leaf, max_depth, and min_samples_split	Same as DT
RF	n_estimators	The number of trees in the random forest	Same as GBDT
	DT hyperparameters	Criterion, min_samples_leaf, max_depth, and min_samples_split	Same as DT
MLP	hidden_layer_sizes	The number of hidden layers in the neural network and the number of neurons in each hidden layer	['layer1': 1 to 20 (n), 'layer2': 0 to n]
	activation	Activation function for the hidden layer	[identity, logistic, tanh, relu]
	max_iter	The maximum number of iterations of the solver	[100, 280, 460, 640, 820, 1000]
	learning_rate_init	The initial learning rate used. It controls the step size when the weight is updated	[0.001, 0.0108, 0.0206, 0.0304, 0.0402, 0.05]
SVM	momentum	Momentum for gradient descent update	[0.5, 0.58, 0.66, 0.74, 0.82, 0.9]
	C	Regularization parameter	[0.001, 0.01, 0.1, 1, 10, 100, 3300, 1000]
	kernel	Specifies the kernel type used in the algorithm	[linear, rbf, poly, sigmoid]

Although GridSearchCV is guaranteed to obtain the most satisfying hyperparameters within a specified range of parameters, this advantage also has the disadvantage of being

time-consuming. Therefore, after executing GridSearchCV and recording the results, the best-performing machine learning algorithm was selected. BO was introduced to tune and compare the machine learning algorithms with GridSearchCV results, including model performance and time consumption. BO is a powerful sequential optimization tool widely applied in machine learning modeling for its ability to obtain the best hyperparameter values quickly [47]. BO allows for mastery of prior knowledge, i.e., incorporating historical information in the iterative process and avoiding redundant and unnecessary evaluation of the objective function $c(x)$ [48,49]. In this study, the execution of BO is implemented by the `bayes_opt` toolkit.

2.3.2. Input Setting and Feature Importance Analysis

The dataset was randomly divided into a training set and a test set by applying the leave-out method at 80–20%. The model's training is performed on the training set, and the test set checks the model's generalization ability. We first used all 40 feature variables as input to machine learning to examine the performance of the algorithms. Three different continuous variable pre-processing methods were considered to examine the effect of pre-processing on model performance. The three continuous variable pre-processing methods are original dataset (OD), Min-Max Scaler (MMS, Equation (1)), and standard scaler (SS, Equation (2)):

$$X_{new} = \frac{X - X_{min}}{X_{max} - X_{min}} \quad (1)$$

where X_{new} represents the scaled value, X is the original value to be scaled. X_{max} and X_{min} represent the maximum and minimum values in the feature variables, respectively.

$$X_{new} = \frac{X - \mu}{\sigma} \quad (2)$$

where μ and σ are the mean and standard deviation of the feature variables.

SHAP is a game-theoretic method that can be applied to interpret the output of machine learning models [50]. SHAP interprets the output of a model as the sum of the actual values attributed to each input feature variable and is an additive feature attribution method [51]. The detail of the computational principles of SHAP can be found in the attached reference [52]. In this work, SHAP was applied to explain the attribution of the model based on a well-established machine learning model for groundwater TPH-contaminated site identification at gas stations.

2.3.3. Model Evaluation

F1 score and area under curve (AUC) were chosen to evaluate the performance of models. Gas stations with TPH-contaminated groundwater are marked as 1 and uncontaminated as 0. If the predicted result and actual value are both 1, the station is classified as a True Positive (TP); while, if the predicted result and actual value are both 0, the station is classified as a True Negative (TN). Conversely, if the predicted result is 1 but the actual result is 0, the station is classified as a False Positive (FP), and if the predicted result is 0 but the actual result is 1, the station is classified as a False Negative (FN). Precision refers to the proportion of results with a predicted value of 1 corresponding to an actual value of 1, i.e., $TP/(TP + FP)$. In contrast, recall refers to the proportion of samples with an actual value of 1 with a predicted outcome of 1, i.e., $TP/(TP + FN)$. F1 score is taken to be in the interval [0,1], the larger, the better. According to Equation (3), the F1 score can be calculated as follows:

$$\text{F1 score} = 2 \times \frac{\text{precision} \times \text{recall}}{\text{precision} + \text{recall}} \quad (3)$$

AUC is calculated based on the receiver operating characteristic curve (ROC), which consists of recall and 1-specificity for different classification probability thresholds. AUC is the area bounded by the ROC curve and the x -axis, $x = 1$. AUC is within the range [0.5, 1], with larger values indicating better model performance.

In addition, this study evaluates the training speed according to the time consumption, aiming to obtain the most efficient model.

3. Results and Discussion

3.1. Performance of Machine Learning Models

The model performances are given in Figure 2. In detail, Figure 2a displays that the F1 score performed well for the LR, GBDT, and SVM models on the training set, all achieving an F1 score of 1. GBDT achieved an F1 score of 1 on the test set, while the highest scores for LR and SVM were only 0.67 and 0.57, respectively (Figure 2c). Figure 2b shows that LR, GBDT, and SVM achieved better results than the DT, RF, MLP, and hierarchical cluster analysis (HCA) models regarding the AUC scores for training. The best AUC results on the test set were GBDT ($AUC = 1$) > LR ($AUC = 0.75$) > SVM ($AUC = 0.72$) (Figure 2d). The results illustrated that GBDT has a certain degree of generalization performance. GBDT accurately identified groundwater TPH contamination in 82 gas stations in the training set and validated the algorithm's capability in 21 gas stations in the test set. This can be attributed to the iterative residual correcting strategy of GBDT, which allowed GBDT to achieve better results than DT and RF [53]. LR performed poorly at dealing with unbalanced datasets, so the results were not satisfying [54]. In addition, SVM and MLP require high data density, which otherwise tends to underfit [55,56]. Moreover, the poor performance of the traditional HCA model compared to machine learning models may be attributed to the specificities of feature variables. These feature variables were not the classical hydrochemical variables related to groundwater TPH, which is a significant difference from other studies where groundwater hydrochemical characteristics were used as input variables in the HCA model [57]. The input variables of the ML model are more flexible, which means all potentially relevant variables can be set as input variables, while the HCA method tends to use groundwater chemistry indicators as input variables. Moreover, the ML model has a stronger ability to find nonlinear relationships among variables than the HCA model. Overall, the GBDT model based on GridSearchCV achieved the highest F1 score and AUC in both the training and test sets. The prediction performance of GBDT was satisfying, while the other models were not.

Models trained with the data processed by SS tend to achieve better performance. In the LR, SVM, and MLP models, the best training results were achieved pre-processed by SS, with F1 scores and AUCs higher than those of OD and MMS. However, in the testing phase, the SS-built models did not necessarily achieve optimal results. The MMS-based LR model performed better in the testing phase, while the SVM was better using OD, indicating that overfitting occurred when using SS for training LR and SVM [58]. Furthermore, for the tree models, including DT, GBDT, and RF, the F1 scores and AUC results obtained from the three pre-processing methods were equal. It may be attributed to the operational mechanism of DT: each input feature variable is treated separately, and therefore each variable does not involve inconsistencies in the magnitudes of different feature variables [59]. Figure 3 shows the confusion matrix of the best-performing models on the test set. GBDT performed well on the 21 samples in the test set, correctly identifying all 4 TPH-contaminated sites ($TP = 4$, $FP = 0$, $FN = 0$). In comparison, other models could only identify one or two. This demonstrates the generalization ability of the GBDT model in identifying TPH contamination in groundwater from gas stations. Therefore, the GBDT model, pre-processed by OD, was chosen for further analysis.

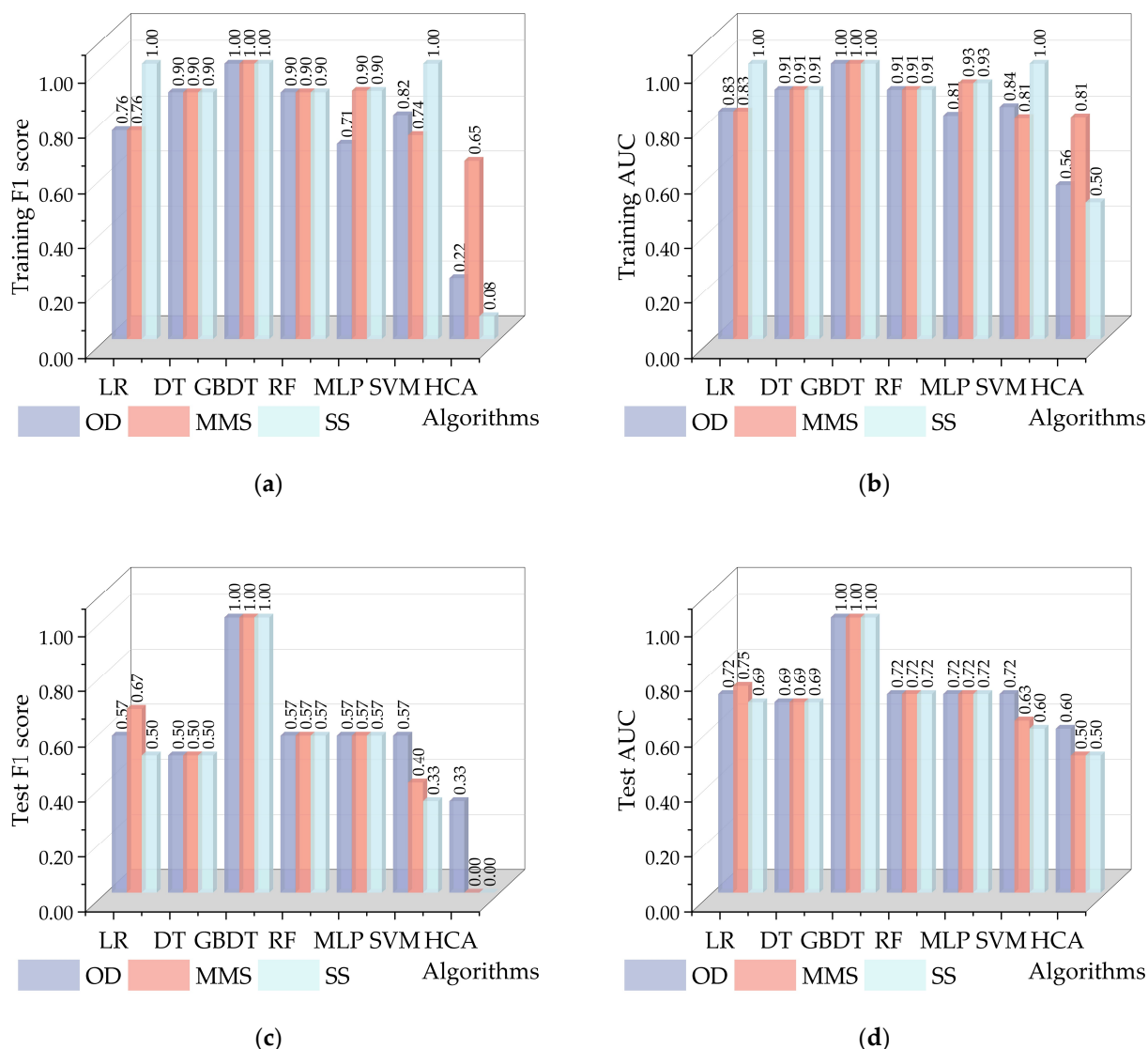


Figure 2. Performance evaluation of all machine learning models. (a) F1 score and (b) AUC of the model on the training set; (c) F1 score and (d) AUC of the model on the test set.

Table 3 shows the information obtained for the GBDT model. Based on the boosting mechanism, GBDT classifies data by overlaying base learners, in which the residuals from the training process are continuously reduced [36]. In this study, the hyperparameter associated with boosting is `n_estimators`, which eventually took a value of 25, implying that 25 boosting stages were performed during the implementation. Among the hyperparameters related to the tree model, the optimal criterion was `friedman_mse`. Therefore, `friedman_mse`, a modified mean squared error function, was chosen to measure the impurities of the nodes [60]. `Min_samples_split` determines the minimum number of samples required for each internal node to be divided. The optimal value of `min_samples_split` was 0.1, which means that 10% of the training samples were required to split an internal node. `Max_depth` means that a single DT is limited in further subdividing the nodes, with an optimal value of three. The optimal value of `min_samples_leaf` was three, indicating the minimum number of samples in each tree leaf node. `Min_samples_leaf` is complementary to `min_samples_split`.

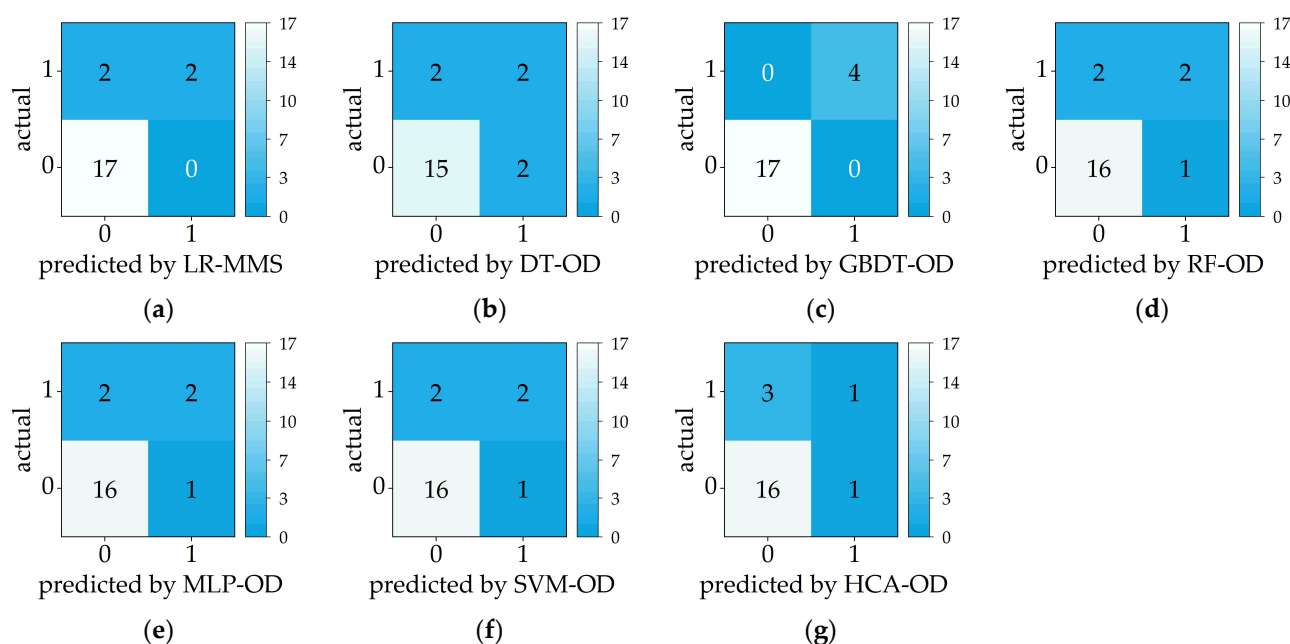


Figure 3. The confusion matrix of the best-performing models on the test set. In each subgraph, from top left to bottom right are FN, TP, TN, FP. (a) LR-MMS; (b) DT-OD; (c) GBDT-OD; (d) RF-OD; (e) MLP-OD; (f) SVM-OD; (g) HCA-OD.

Table 3. Information on the GBDT model for groundwater TPH contamination identification.

Algorithm	Pre-Processing	Hyperparameter	Parameter Optimum
GBDT	OD	criterion	friedman_mse
		n_estimators	25
		min_samples_leaf	3
		max_depth	3
		min_samples_split	0.1

3.2. Model Optimization

Excessive training time is unacceptable due to the wide range of different site types, pollutants, and the fact that the training time of the model increases with the sample size. Table 4 presents the time the model's training consumes based on the GridSearchCV method. MLP consumed the most time, followed by RF and GBDT. Overall, this was attributed to the exhaustive search mechanism of GridSearchCV. However, the single attempt time of MLP was only 1.21–1.28 s. This resulted from the fact that MLP had set the largest number of parameter groups (198,720). RF had the longest single attempt time (3.64 s), so its total time was much longer than GBDT. The GBDT model with the optimal predictive performance had the problem of inefficiency. The single attempt time was about 1.5 s, and the total was more than 5 h. The MLP algorithm set the largest number of parameter groups, since 230 different hidden layer sizes were designed. In addition, both RF and GBDT had a number of parameter groups of the order of 10^4 . As a result, MLP, GBDT, and RF were time-consuming. For algorithms such as LR, GBDT, RF, and MLP, MMS or SS pre-processing of the input shortened the time to train the model. Nevertheless, the total training time for GBDT was still unacceptable.

Table 4. Time consumption summary of training models based on the GridSearchCV method.

Algorithm	Pre-Processing	Number of Parameter Groups	Total Time (s)	Single Attempt Time (s)
LR	OD	9	53.10	5.90
	MMS	9	13.10	1.46
	SS	9	7.75	0.86
DT	OD	840	60.57	0.07
	MMS	840	60.07	0.07
	SS	840	70.70	0.08
GBDT	OD	12,320	19,125	1.55
	MMS	12,320	18,665	1.52
	SS	12,320	18,961	1.54
RF	OD	18,480	67,283	3.64
	MMS	18,480	66,574	3.60
	SS	18,480	65,326	3.53
MLP	OD	198,720	253,500	1.28
	MMS	198,720	248,400	1.25
	SS	198,720	240,780	1.21
SVM	OD	32	2.24	0.07
	MMS	32	3.95	0.12
	SS	32	3.02	0.09

Since GBDT can achieve optimal identification performance, the hyperparameters search process of GBDT was considered to be optimized to improve the efficiency of model training. Here, we proposed to optimize the model parameters by using BO. The settings of BO were as follows:

1. Black box function: the cross-validation score of the GBDT algorithm, and the metrics are F1 score and AUC.
2. Random search step: 5.
3. The number of iterations: 100.
4. Range of hyperparameters: see Table 5. The input of hyperparameters to BO can only be float instead of string and integer. Therefore, the criterion of GBDT is set to friedman_mse, which is also the default value of the model.

Table 5. Hyperparameter range for BO-GBDT.

Hyperparameter	Range
n_estimators	(1, 2000)
min_samples_leaf	(1, 10)
max_depth	(3, 6)
min_samples_split	(0.1, 1)

Table 6 displays the results of the hyperparameters optimized by BO. Compared to GridSearchCV, the optimal hyperparameters searched by BO differed from that of GridSearchCV. However, BO achieved the best performance on both the training and test sets, indicating that the hyperparameters optimized by BO can also exploit the best learning ability of GBDT [61]. Compared to GBDT, the training efficiency of BO-GBDT was much higher, taking only 1/37 of the total time of GBDT. In terms of the single attempt time, BO-GBDT took 5.13 s while GBDT spent 1.5 s for a single model, reflecting the value of BO-GBDT in improving overall efficiency. Therefore, the BO-GBDT model can improve the efficiency of identifying groundwater TPH contamination sites at gas stations without reducing the identification accuracy.

Table 6. The BO results in the GBDT model.

Hyperparameter	Hyperparameter Optimum	Performance	Training Time
n_estimators	978	Training: F1 score = 1,	513 s
min_samples_leaf	1	AUC = 1	
max_depth	3	Test: F1 score = 1,	
min_samples_split	0.9745	AUC = 1	

3.3. Feature Variables Analysis

The top 20 ranked feature variables were presented in the variable importance plot (Figure 4a), including WIN (wind), population, EVP (evaporation), TK_S (total potassium in the soil), PRE (precipitation), LA (leakage accidents), sand_sub (the content of sand in the subsoil), GDP, sand_top, pH_S, silt_top, SSD, Dist_river, construction time, clay_top, NDVI, LAI, soil erosion, NO.SingleTanks, RHU. WIN was regarded as the essential variable for identifying groundwater TPH contamination under gas stations in China (Figure 4a). The WIN distribution in the SHAP summary results further confirmed the high importance of wind speed in identifying groundwater TPH contamination at gas stations (Figure 4b). The results implied that wind speed lower than 1.8 m/s resulted in a higher probability of groundwater TPH contamination (Figure 5a). Part of the reason for this result may be the acceleration of the gasoline vaporization rate by increased surface wind speeds, which results in less TPH leaking into the groundwater [62]. Moreover, the natural attenuation of TPH could be enhanced by an active water cycle under strong wind [63]. The population showed high importance in identifying groundwater TPH contamination (Figure 4a). The SHAP value gradually increased with population, and meanwhile the positive effect on the probability of groundwater TPH contamination increased as well (Figure 4b). The SHAP value was positive when the population was more than 2780 p/km² (Figure 5b), indicating that groundwater TPH contamination is more likely to occur at gas stations in densely populated areas, which may be related to the high volume of transportation and storage due to the high demand for gasoline [64]. The association between population and groundwater TPH contamination reflected the enormous impact of human activities on groundwater around gas stations, including TPH transportation, storage, and accidental spills. EVP, as another meteorological variable, was also important to the BO-GBDT model. The SHAP values were negative with lower EVP values (Figure 4b). Specifically, when the EVP was below 1391 mm/a, the groundwater at the gas station was not likely to be contaminated with TPH (Figure 5c). In contrast, the probability of groundwater TPH contamination increased remarkably when EVP was higher than 1391 mm/a, which could be ascribed to the increase in TPH concentration caused by water evaporation [65,66]. Moreover, high EVP may result from high permeability, which may accelerate the transport of TPH from soil to groundwater.

TK_S between 0 and 1969 mg/kg has a negative SHAP value, while it has a positive SHAP value when TK_S is greater than 1969 mg/kg (Figure 5d). This indicates that the high TK_S is somewhat related to the TPH pollution of groundwater in the local gas station. In addition, PRE was identified as another important variable associated with groundwater TPH contamination. The results of the SHAP summary showed that the decrease in PRE increased the probability of groundwater TPH contamination (Figure 4b). The SHAP values start to turn negative when the PRE exceeds 560 (Figure 5e). This result may be related to the high natural attenuation of TPH at high value of PRE [67]. Meanwhile, PRE tends to be negatively correlated with TK_S due to the strong leaching effect caused by rainfall [68], which explains the positive correlation between TK_S and SHAP value. LA was also identified as a significant variable. Groundwater TPH contamination occurred at all gas stations where tank leaks have been detected (Figure 4b), indicating that once tank leaks occur, groundwater is likely to be contaminated. The results alert the environmental authorities to monitor the gas storage tanks more closely to avoid the risk of LA. The SHAP value increased with a decrease in sand_sub, indicating that TPH contamination of

groundwater at gas stations is more likely to occur with low sub-soil sand content. The SHAP value was negative when sand_sub was between 0 and 12.33% (Figure 5f). This may be related to the TPH trapping effect of the sand in the subsoil [69]. For features ranked after sand_sub, their SHAPs are concentrated around 0, indicating that they play a minor role in the well-established model (Figure 4).

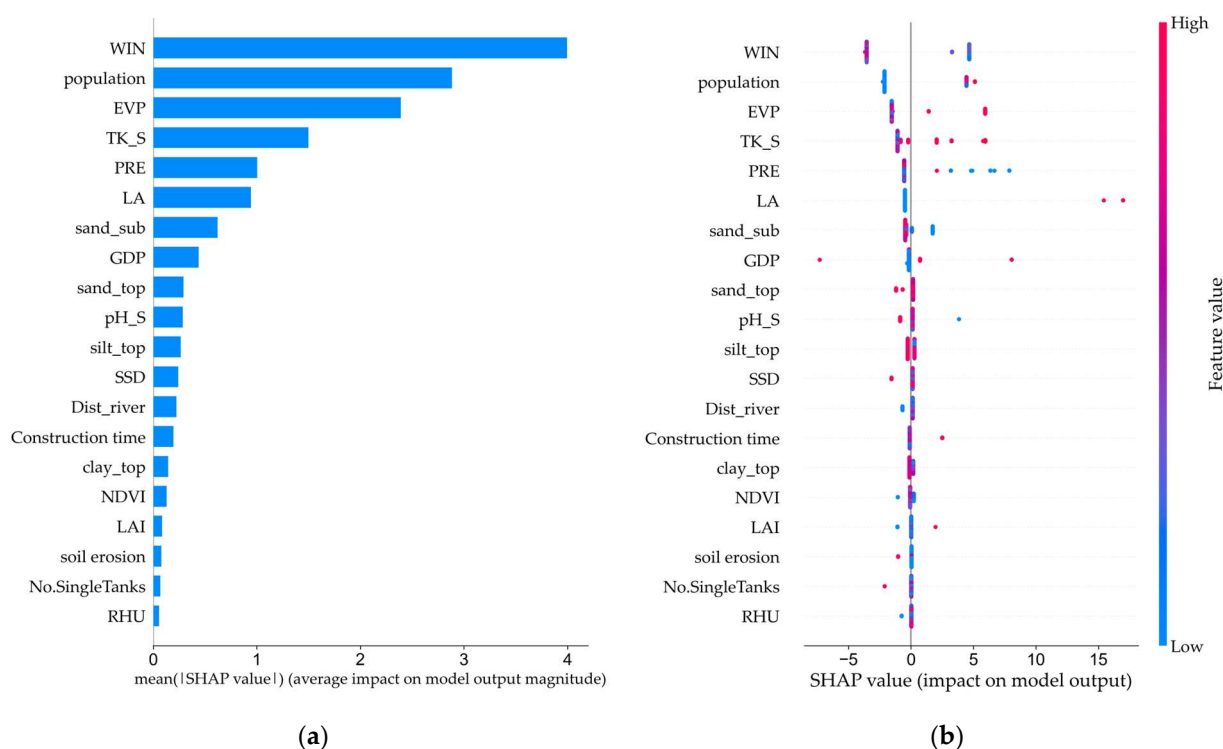


Figure 4. Results of SHAP analysis of the BO-GBDT model. (a) Bar diagram showing the feature variable importance.; (b) SHAP summary plot of feature variables.

Furthermore, the above seven crucial feature variables were set as inputs of BO-GBDT to investigate their identification performance of groundwater TPH pollution. The settings of the input variables are shown in Table 7.

Table 7. Input combinations for each BO-GBDT model.

Number of Variables	Input Variables
1	WIN
2	WIN, population
3	WIN, population, EVP
4	WIN, population, EVP, TK_S
5	WIN, population, EVP, TK_S, PRE
6	WIN, population, EVP, TK_S, PRE, LA
7	WIN, population, EVP, TK_S, PRE, LA, sand_sub

The performance metrics of the BO-GBDT model obtained by training with the seven input settings are shown in Figure 6. Overall, the F1 score and AUC of the BO-GBDT model showed an increasing trend when the number of input variables was increased, showing the boosting effect of increasing variables on the model performance. It is worth noting that the model already performed well when using only WIN as an input variable, with F1 scores and AUCs of 0.87 and 0.90 on the training set and 0.75 and 0.85 on the test set, respectively, reflecting the vital role of WIN in the model's prediction. This is in agreement with the findings presented in Figure 3a. At five variables, the predictive performance slightly decreases compared to four, suggesting that with few feature variables, adding features

may potentially introduce overfitting, as previously reported [70]. The model's performance saw a notable improvement when using six input variables (training F1 score = 1, test F1 score = 0.89, training AUC = 1, test AUC = 0.97). When the number of input features was seven, the results were consistent with those when the number of features was six. It was found that when using six or seven input variables, all the prediction errors involved cases where no contamination was predicted as contamination ($n = 1$). In that case, although the model's results are not perfect, they are relatively conservative. Therefore, a reliable model for identifying groundwater TPH contamination can be established using only six variables (WIN, population, EVP, TK_S, PRE, LA).

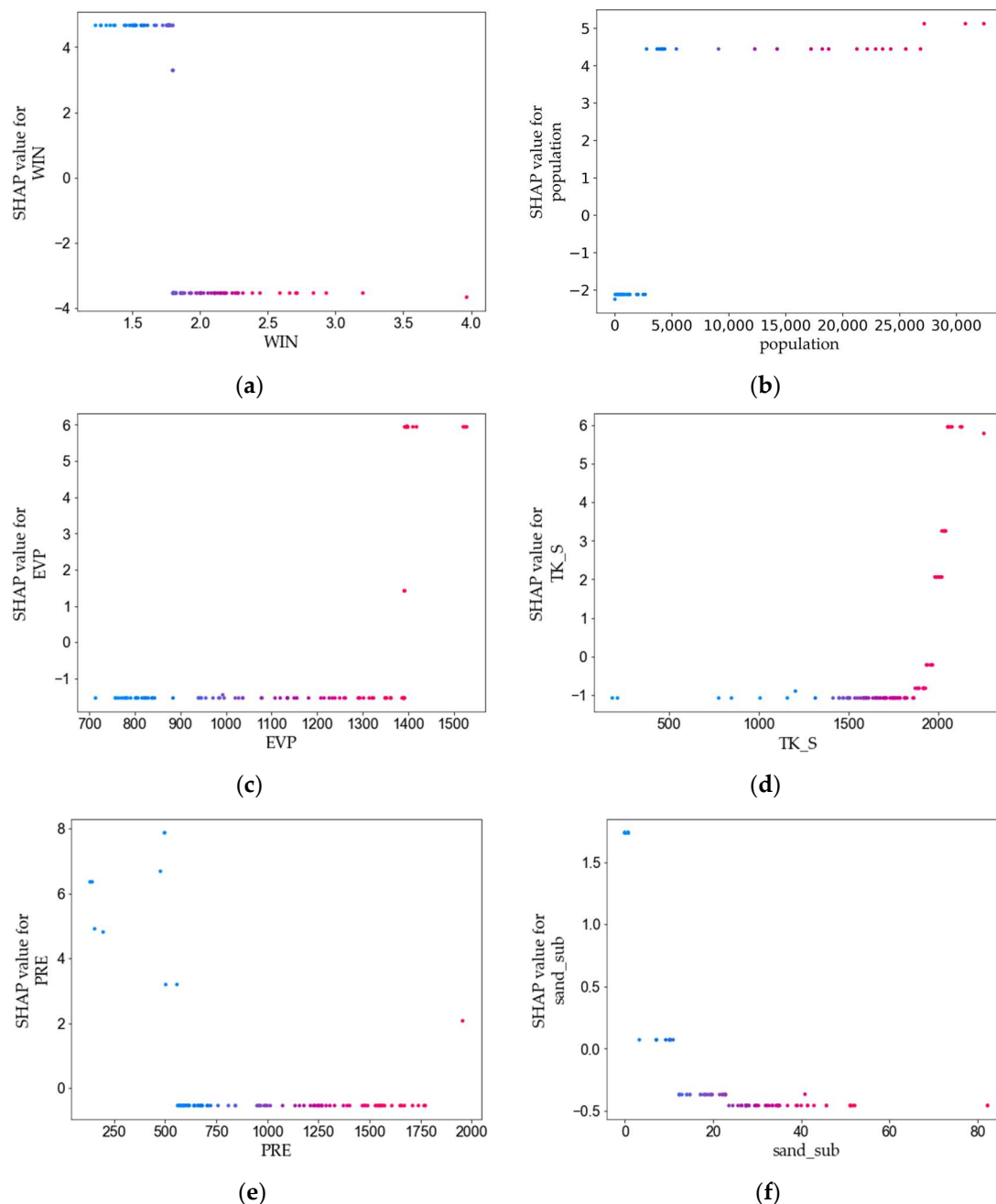


Figure 5. Scatter plots of feature variables and SHAP values. (a) WIN; (b) population; (c) EVP; (d) TK_S; (e) PRE; (f) sand_sub.

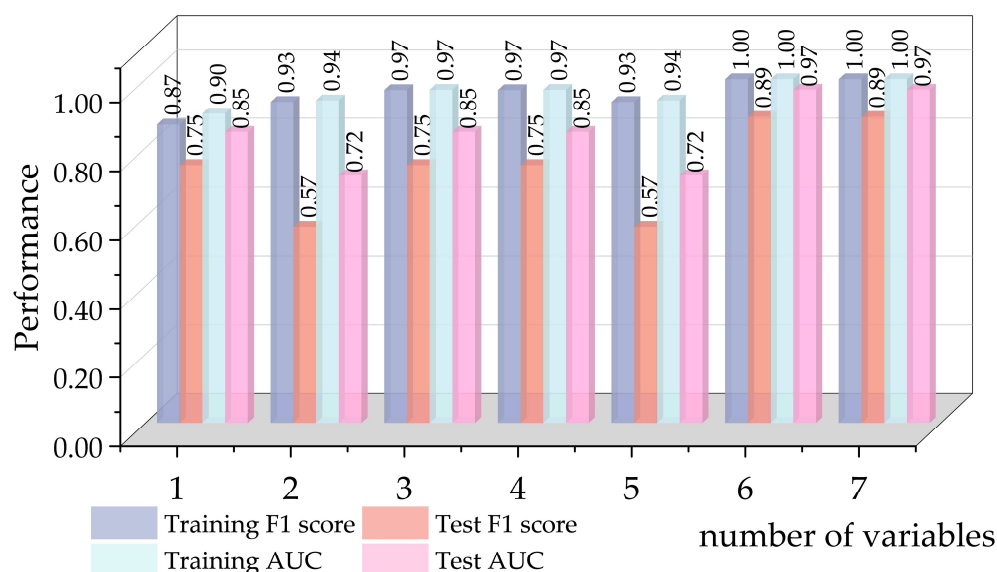


Figure 6. Performance of BO-GBDT model established by different input settings on the training and test sets.

The variables analysis results reveal that TPH contamination in groundwater at gas stations in China is a combination of natural and anthropogenic factors. China's eco-environmental authorities are undertaking a series of pollution prevention control of gas stations, as required by policy documents such as the National Groundwater Pollution Prevention and Control Plan (2011–2020) and the 14th Five-Year Plan for Soil, Groundwater and Rural Ecological Protection. The human activities at gas stations including the transportation and storage of gas need to be further regulated. We recommend that managers pay more attention to the supervision of gas storage tanks in densely populated areas in order to detect potential leaks on time. This study also provides a scientific basis for the site selection of new gas stations. From the point of view of avoiding groundwater contamination by TPH, areas with low wind speed, low evaporation, low population density, and high precipitation are considered suitable sites for new gas stations.

4. Conclusions

This study applied a machine learning approach to develop models to identify groundwater contamination sites. An evaluation of model prediction performance and efficiency was carried out. A case study was conducted using gas station sites in China with groundwater TPH as the target contaminant. The model developed by the GBDT algorithm had the most satisfying prediction performance (F1 score = 1, AUC = 1). Compared to GridsearchCV, the BO-GBDT model could significantly improve the modeling efficiency without degrading the prediction performance, with a time from 19,125 s to 513 s. Therefore, BO-GBDT was the most reliable prediction model in this study. The SHAP results displayed that the critical feature variables in the BO-GBDT model included wind, population, evaporation, total potassium in the soil, precipitation, and leakage accident.

It can be seen that the machine learning approach, particularly being optimized with BO, shows potential for identifying groundwater contamination sites. The limitations of the method mainly come from two aspects: (1) the machine learning model has inherent uncertainty itself, which may affect the generalization ability; (2) the relatively small sample size introduces uncertainty into the results of this study. In future studies, researchers should incorporate more gas station contamination samples for further model improvement and validation. Moreover, detailed experiments should be conducted to further verify the relationships between the variables and TPH contamination identified in this study. In addition, some other machine learning models, such as deep learning-based models, also deserve to be studied for groundwater contamination site identification. The idea of using a

machine learning approach to identify groundwater contamination sites is highly suggested to be employed in more countries/regions and more types of sites and contaminants.

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