Composition analysis and identification of ancient glass objects based on Random Forest

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Abstract. Glass is a valuable physical evidence of our early trade exchanges, and ancient glass is susceptible to weathering by the burial environment, resulting in changes in its composition ratio. In this study, the surface weathering of glass artifacts was analyzed in relation to their glass type, decoration and color; the statistical patterns of the chemical composition content of artifact samples with and without weathering were analyzed in relation to the glass type and a Random forest (RF) model was developed to predict the chemical composition content of the artifacts before weathering based on the weathering point detection data.

Keywords: Glass; Machine Learning; Random Forest; K-means; Pearson Correlation Coefficient.

1. Introduction

As an important channel of cultural exchange between China and the West in ancient times, glass was introduced to China through the Silk Road in the early days [1], and after that China gradually realized the localized production of glass. In ancient Chinese glass forging, the main component is quartz sand (SiO2) [2]. When forging glass, fluxes need to be added, and the chemical composition of the final forged glass varies greatly depending on the fluxes added. Adding grass ash (K2CO3) as a flux, what we get is high potassium glass with high potassium content, adding lead ore (PbO, BaO), what we get is lead barium glass. Under the influence of the internal and external environment for a long time, some glasses have weathered. Some weathering is more obvious, but some weathering is not directly identifiable with the naked eye. In this study, the surface weathering of glass artifacts was analyzed in relation to their glass type, decoration and color; the statistical patterns of the chemical composition content of artifact samples with and without weathering were analyzed in relation to the glass type and a Random forest (RF) model was developed to predict the chemical composition content of the artifacts before weathering based on the weathering point detection data.

In this study, we analyze the relationship between surface weathering and type, decoration, and color of glass artifacts, analyze the statistical pattern of chemical composition of artifacts with and without weathering, and predict the content of chemical composition of weathered artifacts (areas) before weathering [3]. Since type, decoration, and color are parameters described in words, these parameters cannot be directly brought into the calculations, and before that we need to standardize them into numerical parameters that can be brought into the calculations. Since these data do not conform to a normal distribution, the lower statistical utility but wider applicability of the spearman and kendall coefficients are considered for the correlation analysis. Or we can analyze them by algorithms such as random forest in machine learning [4]. When analyzing the statistical patterns of chemical compositions, we can perform descriptive statistics on them and discuss their extreme values, means, standard deviations, variances, skewness, and kurtosis. When predicting the chemical composition before weathering, we can use the algorithm in machine learning to bring in the existing data before and after weathering for training, and then bring in the weathered chemical composition for solution.[5]

2. Model building and solving

2.1. Data pre-processing

(1) Deletion of missing data points

From the known data, it can be seen that there are artifacts with the following missing color data (Table 1).

Table 1. Color data missing artifact table

Artifact Number	Ornamentation	Type	Color	Surface weathering
19	A	Lead Barium	---	Weathering
40	C	Lead Barium	---	Weathering
48	A	Lead Barium	---	Weathering
58	C	Lead Barium	---	Weathering

It can be seen that only the characteristic values of surface color of artifacts with type lead-barium have missing values.

In the next analysis, we eliminate these artifacts with missing values of color because the color index is a quantitative variable that has no correlation with artifact number, decoration, or type and cannot be predicted using conventional prediction models.

(2) Data outlier removal

From the Annexes and the conditions in the question, it is clear that the null value means that the chemical element component is not detected, so we import the null value in the table and replace the null value with 0.

Due to the qualification in the question, the chemical proportion data of each cultural object is characterized by "compositionality", i.e., the sum of its composition may not be equal to 100% due to testing methods and other reasons. In this question, the sum of the proportion of components between 85% and 105% of the data is considered valid. The proportion of chemical composition of each cultural object sampling point and need to meet the following conditions.

$$85\% <= \sum_{j=1}^{14} ratio_{i,j} <= 105\% \tag{1}$$

where i represents the artifact numbered i, j represents the j-th chemical composition, and $ratio(i,j)$ represents the ratio of the jth chemical composition of the i-th artifact.

We use the sum function in excel on the data for each artifact sampling points for the accumulation of chemical composition percentage, can be obtained to meet the qualifying conditions of artifacts as shown in Table 2.

Table 2. Table of the proportion of artifacts meeting the qualifying conditions

Artifact Number	SiO_2	Na_2O	K_2O	…	Sum of proportion
01	69.33	0	0	…	97.61
02	36.28	0	0	…	99.89
03 Part 1	87.05	0	0	…	100
03 Part 2	61.71	0	0	…	98.88
04	65.88	0	0	…	96.06
…	…	…	…	…	…

Artifacts that do not meet the qualifying conditions are numbered 15 and numbered 17. Among them, artifact number 15: $\sum_{i=1}^{14} ratio_{i,_{15}} = 79.47$,artifact number 17: $\sum_{i=1}^{14} ratio_{i,_{17}} = 71.89$, are not satisfied with the chemical element ratio and limited in the question, so we remove the outliers that do not satisfy the conditions of the question.

As can be seen from Figure 1, the number of unweathered high-potassium glass is the same as the number of unweathered lead-barium glass, but the number of weathered lead-barium glass is about five times higher than the number of high-potassium glass, which shows that the proportion of weathered lead-barium glass is much higher than that of high-potassium, and lead-barium glass is more easily weathered than high-potassium glass, thus giving archaeologists the inspiration that lead-barium glass should be preserved as soon as possible after excavation. Combined with the literature, it can be seen that the lead in lead-barium glass can react directly with CO_2 and H_2O in the environment to produce Pb_2O_3, and Pb_2O_3 will gradually collect on the surface of the glass, which is also the weathering process of the glass.

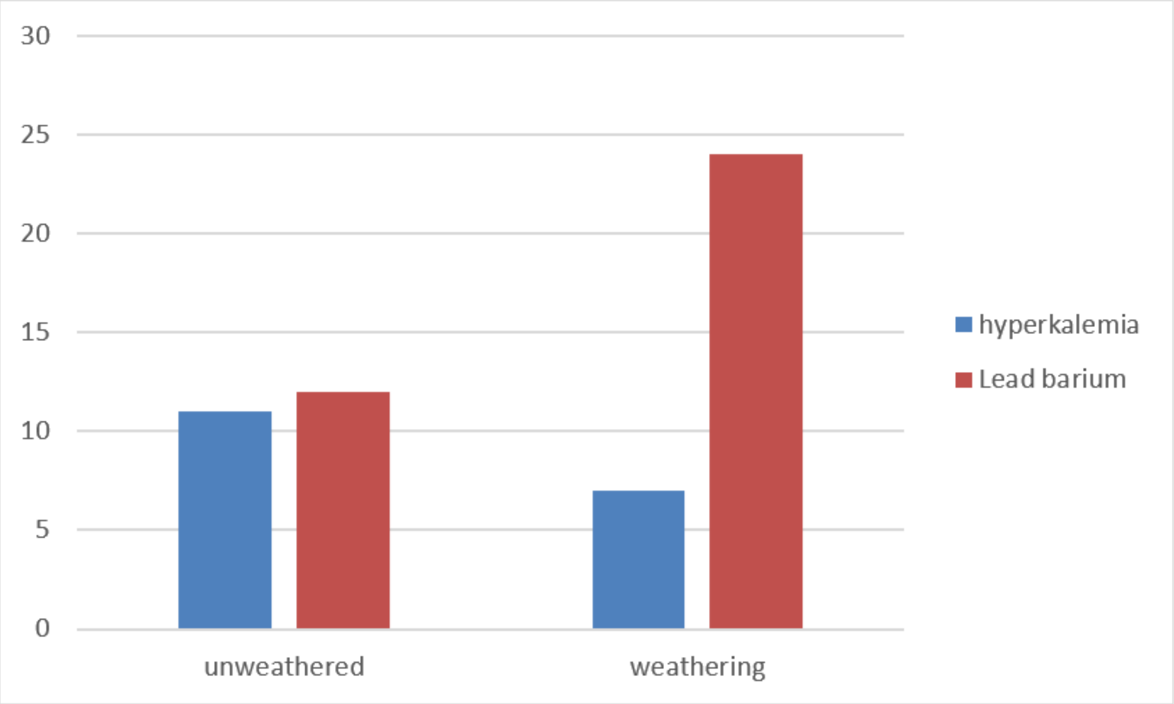

Figure 1. Statistics of glass types and weathering

As can be seen from Figure 2, the weathered glass has the highest amount of light blue glass weathered, the second amount of blue-green glass weathered, which is much higher than the number of other colors of glass weathered, dark green glass weathered as the third, the number of other colors of glass weathered is less, while dark blue and green glass is almost absent. Among the unweathered glass, the highest number of light blue glass weathered, the second number of blue-green glass weathered, much higher than the number of other colors of glass, dark green glass weathered remained third, the number of purple, dark blue and light green glass was about the same, green glass was about 1/2 times of purple, dark blue and light green glass, and there was basically no black glass. It can be seen that the light blue glass has the highest number of excavations, the blue-green glass has the second highest number, and the other glasses have fewer excavations. It is reasonable to speculate that glass artifacts of other colors may be less stable due to many factors, such as light, and may not be easily preserved or have a chemical mechanism for conversion to light blue or blue-green artifacts.

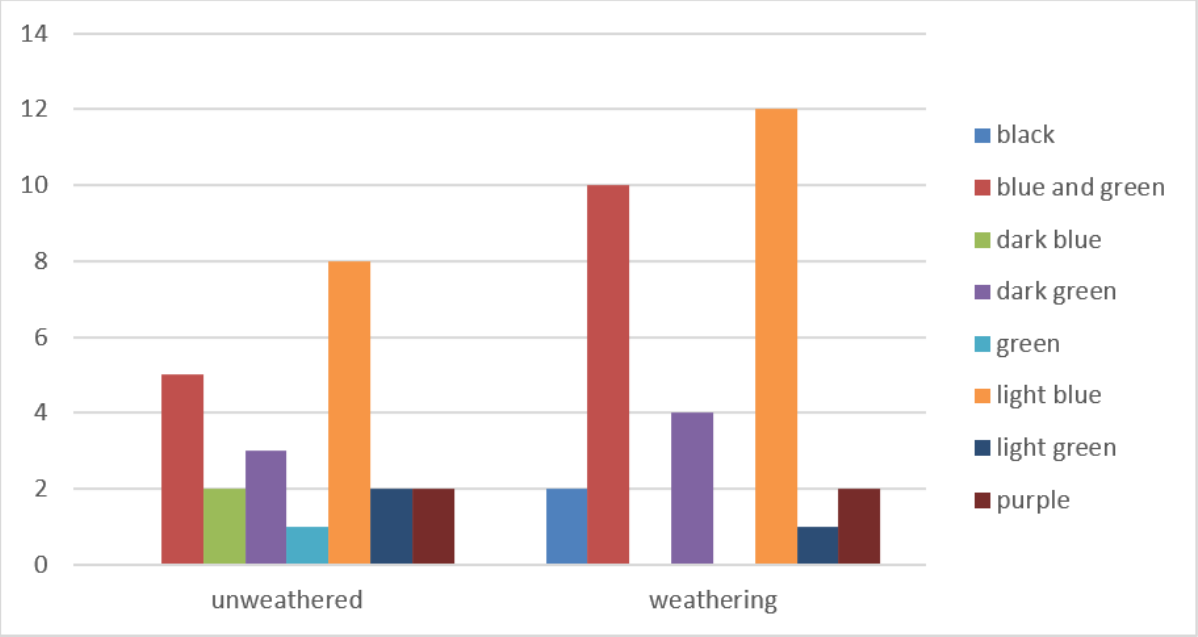

Figure 2. Statistics of glass color and weathering

As can be seen from Figure 3, the number of glass weathered with ornamentation C is the largest, the number of glass with ornamentation A is the second, much larger than the number of glass with ornamentation B. The glass with ornamentation C is about seven times larger than the glass with ornamentation B, and the glass with ornamentation A is about four to five times larger than the glass with ornamentation B. Among the unweathered glass, the number of glass with ornamentation C is the largest, the number of glass with ornamentation A is the second, and there is almost no glass with ornamentation B There is almost no glass with B decoration. It can be seen that, among the excavated glass artifacts, the number of glass with the C motif is the largest, the number of glass with the A motif is the second, and there is basically no glass with the B motif. This shows that the glass decorated with C and A is easier to preserve, and the glass decorated with B is less easy to preserve.[6]

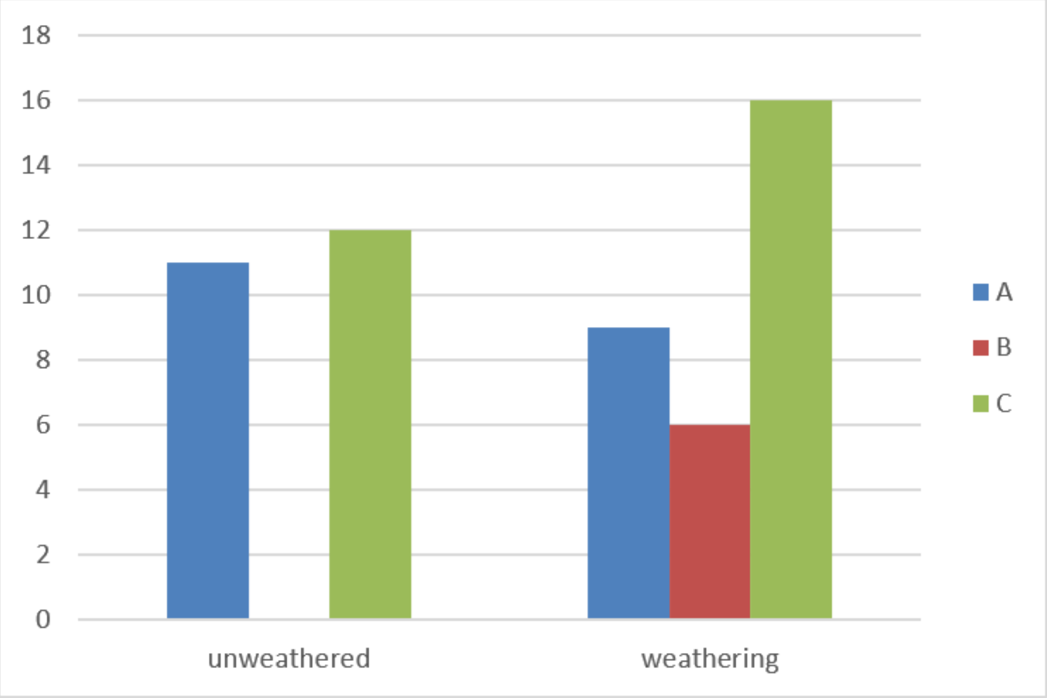

Figure 3. Statistics of glass decoration and weathering

2.2. Background and introduction of the random forest model

Brieman proposed the random forest algorithm in 2001[7]. The random forest algorithm is an important tool for nonlinear modeling because it has a good tolerance to outliers for non-equilibrium samples, is not prone to overfitting in the processing of data, and has high prediction accuracy.

We introduce the random forest algorithm into this problem, and use the random forest to assign weights to the three indicators of the artifacts in the annex $Sheet1$: ornamentation, type, and color, and we set the above three indicators as W,T,C.

Breiman and Cutler borrowed the algorithm of random decision forest to combine classification trees into a random forest, this method is equivalent to the method of randomly selecting sample data, generating classification trees, and then classifying and aggregating the results, which becomes the random forest algorithm (RF) [8]. Compared with neural networks, support vector machines (SVMs), decision trees (DTs), etc., RF does not increase exponentially in the amount of operations and operational complexity, but has significantly higher prediction accuracy and is more robust in dealing with modeling situations with missing data and non-equilibrium.

Random forest regression is used to build a decision tree regression model by bootstrap sampling method to obtain different sample sets. We assume that the training set is the result of random sampling from a random vector Y, X with independent distribution, and the predicted value of the random forest regression is the average of the regression results of k times of the decision tree.

$$\mathrm{H(x)} = \frac{1}{k}\sum_{i=1}^{k} h_i(x) \tag{2}$$

where $\sum_{i=1}^{k} h_i(x)$ represents the mean of the regression results of the representative k decision trees, and H(x) represents the result of the regression.

In this problem we use the random forest classification method, in the original sample set by bootstrap sampling method to get k different sample sets, build a decision tree model, get k classification results, and vote on the samples to decide their final classification, we assume that $h_i(x)$ is the final prediction, I is the indicator function, H(x) is the classification result of a single decision tree, and Y is the output variable.

$$\mathrm{H(x)} = \arg\max_{\mathrm{Y}} \sum_{i=1}^{k} I(h_i(x) = Y) \tag{3}$$

The larger the margin function is, the more reliable the model of classification prediction will be.

$$\mathrm{mg(X,Y)} = av_k I(h_k(x) = Y) - max\ \ av_k I(h_k(x) = j) \quad (j \neq k) \tag{4}$$

where $h_k(x)$ represents the classification model sampled from the original dataset.

This gives us the generalization error of our established decision tree classification model.

$$PE^* = P_{X,Y}(mg(X,Y) < 0) \tag{5}$$

Eventually, the generalization errors of all decision trees converge to :

$$\lim_{n\to\infty} PE = P_{XY}(P_\theta(k(X,\theta) = Y) - maxP_\theta(k(X,\theta) = j) < 0) \tag{6}$$

where n is the number of decision trees in the random forest classification model.

In the first sub-question, we are required to analyze the relationship between the surface weathering of glass artifacts and their glass type, ornamentation and color.

First, we use the random forest classification algorithm to analyze the magnitude of the correlation between glass type, decoration, color, and weathering of the surface of the artifacts.

Because of the randomness in the process of the random forest model, the results of each run may be slightly different, and we run it several times, and we determine the 8 stable output values in which there is no large deviation, and we can get the results as shown in Table 3.

Table 3. Table of initial indicator weights

Times	$Weight(T)*$	$Weight(W)*$	$Weight(C)*$
1	0.4157	0.5780	0.0063
2	0.4189	0.5758	0.0053
3	0.4146	0.5766	0.0088
4	0.4139	0.5742	0.0119
5	0.4220	0.5746	0.0034
6	0.4167	0.5783	0.0050
7	0.4235	0.5742	0.0023
8	0.4164	0.5735	0.0101

We determine the 8 stable output values without large deviations, let the indicator glass type, decoration, color of the weight value in each output there is a random perturbation, then the weight expression is

$$Weight(i)^{*} = weight(i) + \delta_i \tag{7}$$

where $i = \{T, W, C\}$.

where $Weight() *$ is the true output value of the weights in 8 times, $weight$ is the value of the weights we finalize, and δ_i is the perturbation value in 8 times, where $i = \{T, W, C\}$ we assume that δ_i satisfies the normal distribution, then

$$\delta_i = N_{i,j}(\mu_i, \sigma_i^{\,2}) \tag{8}$$

The results are shown in Table 4.

Table 4. Table of weights

Indicators	$weight(T)$	$weight(W)$	$weight(C)$
weights	0.4157	0.5780	0.0063

2.3. Test of Spearman correlation coefficient on the results

The Spearman rank coefficient is defined as the Pearson correlation coefficient between rank variables and is a nonparametric indicator used to measure the correlation coefficient between two variables.[9]

Since this data does not satisfy the normal distribution, nor the discontinuous data of linear relationship, we tested the reasonableness of the random forest prediction by Spearman's correlation coefficient in spss [10]. As shown in Table 5, the correlation coefficient between glass type and weathering is greater than that between glass decoration and weathering, and greater than that between glass color and weathering. The results are the same as those of the random forest classification assignment values.

Table 5. spearman correlation coefficient table

Feature	Ornamentation	Glass Type	Color
Surface weathering	0.0307	0.344**	0.0142

**at the 0.01 level (two-tailed) with significant correlation.

2.4. Cardinality test

The cardinality test determines the size of the cardinality value by the degree of deviation between the actual observed value of the statistical sample and the theoretical inferred value. If the chi-square value is larger, the greater the degree of deviation between the two; conversely, the smaller the deviation between the two; if the two values are exactly equal, the chi-square value is 0, indicating that the theoretical value is exactly the same.

From Table 6, it can be seen that the p-value of glass decoration with weathering is 0.084, the p-value of glass type with weathering is 0.009, and the p-value of glass color with weathering is 0.507. Therefore, it can be concluded that the correlation between glass type and weathering is greater than the correlation between glass decoration and weathering, and the correlation between glass decoration and weathering is greater than the correlation between glass color and weathering.

Table 6. Cardinality test table

Title	Name	Surface weathering		Total	X^2	Calibration X^2	*P*
		No weathering	Weathering				
Ornament	A	14	14	28	4.957	4.957	**0.084**
	B	0	6	6			
	C	13	22	35			
Total		27	42	69			
Type	High Potassium	14	6	20	6.880	5.452	**0.009****
	Lead Barium	13	36	49			
Total		27	42	69			
Color	Blue-Green	8	9	17	6.287	6.287	**0.507**
	Pale Blue	8	16	24			
	Purple	2	4	6			
	Dark Green	3	4	7			
	Deep Blue	3	0	3			
	Black	0	8	8			
	Light green	2	1	3			
	Green	1	0	1			
Total		27	42	69			
* $p<0.05$ ** $p<0.01$							

2.5. High potassium, lead and barium descriptive statistics analysis

The glass was divided into four types: lead-barium differentiated, lead-barium undifferentiated, high potassium weathered and high potassium unweathered, and descriptive statistical analysis was performed on the data of these four types in turn(Table 7). As can be seen from Table 7, there are only B ornamentation and blue-green color in the weathering type of high potassium glass, and the total content range is in a relatively stable interval; there are A and C ornamentation in the unweathered type of high potassium glass, and their colors are blue-green, light blue and dark blue; there are A and C ornamentation in the weathered type of lead-barium, and only B ornamentation in the unweathered type of lead-barium.

Table 7. High potassium and lead barium glass description statistics

Glass Type	Whether weathering	Ornament	Color	Total content range
High Potassium	Weathering	B	Blue-Green	99.81%-100%
	Unweathered	A、C	Blue green, light blue, dark blue	97.25%-100%
Lead Barium	Weathering	A、C	Blue-green, black, light blue, light green, dark green, purple	90.17%-99.89%
	Unweathered	B	Dark green, light green, purple, green, light blue, dark blue	88.41%-99.98%

From the comparison of the chemical composition of high potassium before and after weathering, it can be concluded (Table 8) that the silica content of high potassium glass products will increase substantially, from 67.9872% to 93.9633%, accounting for a large proportion of the total chemical composition of high potassium glass; the content of sodium oxide, potassium oxide, and calcium oxide will decrease significantly; lead oxide and barium oxide have no detectable value, indicating that with the weathering of high potassium will produce some loss of oxides.

From the literature [2], it is clear that this glass belongs to the soda-lime system, and with the absence of the main elements such as K and Na, the corresponding silicon content will increase due to (1) the Si-O bond can adsorb moisture in the air and thus form hydroxyl groups, which can further combine with water molecules to form hydrogen bonds, and the increase in moisture leads to easy weathering of the glass. (2) Alkaline ions will exchange ions with hydrogen ions or hydrated hydrogen ions in the air to form, which will directly erode the glass.

From the comparison of the chemical composition of lead-barium glass before and after weathering (Table 9), it can be concluded that the silica content and lead oxide content of lead-barium glass products are significantly reduced; sodium oxide has a certain degree of reduction, and no value of sodium oxide is detected in the severe weathering situation; potassium oxide has a small increase from unweathered to weathered, and the value remains relatively stable from weathering to severe weathering; calcium oxide increases with the degree of weathering The content of magnesium oxide increased to some extent from unweathered to weathered, and decreased to some extent from weathered to severely weathered, indicating the loss of magnesium element from weathered to severely weathered. The contents of barium oxide and phosphorus pentoxide keep increasing with the degree of weathering, indicating that the elements of barium and phosphorus increase with the process of weathering.

Combined with the literature, it can be seen that the lead in lead-barium glass can react directly with and in the environment to produce, which will gradually gather on the surface of the glass, which is also the weathering process of the glass, and this also explains the decrease of the elemental lead content.

Table 8. Description statistics of high potassium glass

Element	Whether weathering	N	Min	Max	Average	Standard deviation	Variance	Bias	Kurtosis
Silicon dioxide (SiO2)	Weathering	6	92.35	96.77	93.9633	1.73362	3.005	0.854	-0.388
	Unweathered	12	59.01	87.05	67.9842	8.75510	76.652	1.158	0.536
Sodium oxide (Na2O)	Weathering	0	0	0	0	0	0	0	0
	Unweathered	3	2.10	3.38	2.7800	0.64374	0.414	-0.551	0
Potassium oxide (K2O)	Weathering	4	0.59	1.01	0.8150	0.18735	0.035	-0.328	-2.239
	Unweathered	11	5.19	14.52	10.1791	2.72171	7.408	-0.288	-0.325
Calcium oxide (CaO)	Weathering	6	0.21	1.66	0.8700	0.48777	0.238	0.504	0.988
	Unweathered	10	2.01	8.70	6.3990	2.02628	4.106	-1.075	1.265
Magnesium oxide (MgO)	Weathering	2	0.54	0.64	0.5900	0.07071	0.005	0	0
	Unweathered	10	0.52	1.98	1.2950	0.49820	0.248	-0.295	-1.250
Alumina (Al2O3)	Weathering	6	0.81	3.50	1.9300	0.96449	0.930	0.779	0.181
	Unweathered	12	3.05	11.15	6.6200	2.49151	6.208	0.482	-0.492
Iron oxide (Fe2O3)	Weathering	6	0.17	0.35	0.2650	0.06950	0.005	-0.300	-1.418
	Unweathered	10	0.42	6.04	2.3180	1.54924	2.400	1.446	3.846
Copper oxide (CuO)	Weathering	6	0.55	3.24	1.5617	0.93482	0.874	1.218	2.231
	Unweathered	11	0.47	5.09	2.6755	1.54109	2.375	0.099	-1.000

Lead oxide (PbO)	Weathering	0	0	0	0	0	0	0	0
	Unweathered	7	0.11	1.62	0.7057	0.62761	0.394	0.622	-1.782
Barium oxide (BaO)	Weathering	0	0	0	0	0	0	0	0
	Unweathered	5	0.00	2.86	1.4360	1.07183	1.149	-0.008	0.043
Phosphorus pentoxide (P2O5)	Weathering	5	0.15	0.61	0.3360	0.17771	0.032	0.902	0.881
	Unweathered	11	0.16	4.50	1.5300	1.43083	2.047	1.679	1.615
Strontium oxide (SrO)	Weathering	0	0	0	0	0	0	0	0
	Unweathered	6	0.04	0.12	0.0833	0.03141	0.001	-0.228	-1.760
Tin oxide (SnO2)	Weathering	0	0	0	0	0	0	0	0
	Unweathered	1	2.36	2.36	2.3600	0	0	0	0
Sulfur dioxide (SO2)	Weathering	0	0	0	0	0	0	0	0
	Unweathered	3	0.36	0.47	0.4067	0.05686	0.003	1.206	0

Table 9. Description statistics of lead barium glass

Element	Whether weathering	N	Min.	Max.	Average	Standard deviation	Variance	Bias	Kurtosis
Silicon dioxide (SiO2)	Weathering	21	4.61	39.57	24.4733	8.46033	71.577	-0.241	0.242
	Unweathered	23	31.94	75.51	54.6596	11.82859	139.916	-0.371	-0.538
Sodium oxide (Na2O)	Weathering	3	1.22	2.22	1.6067	0.53715	0.289	1.561	0
	Unweathered	10	0.92	7.92	3.8700	2.08408	4.343	0.698	0.044
Potassium oxide (K2O)	Weathering	6	0.14	1.05	0.4017	0.33367	0.111	1.967	4.101
	Unweathered	15	0.11	1.41	0.3353	0.33041	0.109	2.859	8.788
Calcium oxide (CaO)	Weathering	20	0.37	6.40	2.7980	1.77577	3.153	0.434	-0.899
	Unweathered	20	0.38	4.49	1.5185	1.26272	1.594	1.339	0.774
Magnesium oxide (MgO)	Weathering	12	0.47	2.73	1.1650	0.59012	0.348	1.695	4.336
	Unweathered	15	0.51	1.67	0.9820	0.33225	0.110	0.449	-0.066
Alumina (Al2O3)	Weathering	21	0.50	5.73	2.6119	1.52499	2.326	0.742	-0.296
	Unweathered	23	1.42	14.34	4.4561	3.26245	10.644	1.988	4.233
Iron oxide (Fe2O3)	Weathering	12	0.23	2.74	0.9825	0.81235	0.660	1.029	0.195
	Unweathered	11	0.17	4.59	1.5400	1.25395	1.572	1.566	2.848
Copper oxide (CuO)	Weathering	21	0.19	10.57	2.3300	3.03692	9.223	2.124	3.715
	Unweathered	21	0.11	8.46	1.5681	2.01095	4.044	2.383	6.387
Lead oxide (PbO)	Weathering	21	25.39	61.03	44.1976	10.10527	102.117	-0.148	-0.528
	Unweathered	23	9.30	39.22	22.0848	8.21515	67.489	0.620	-0.456
Barium oxide (BaO)	Weathering	18	3.26	32.25	13.5850	8.91047	79.396	1.337	0.696
	Unweathered	23	2.03	26.23	9.0017	5.82528	33.934	1.797	3.758
Phosphorus pentoxide (P2O5)	Weathering	19	0.07	14.13	5.8147	4.23203	17.910	0.388	-0.730
	Unweathered	17	0.08	6.34	1.4194	2.03287	4.133	1.745	1.839
Strontium oxide (SrO)	Weathering	18	0.19	1.12	0.4944	0.23045	0.053	1.275	2.248
	Unweathered	17	0.12	0.91	0.3629	0.21210	0.045	1.884	3.150
Tin oxide (SnO2)	Weathering	1	0.47	0.47	0.4700	0	0	0	0
	Unweathered	3	0.23	0.44	0.3567	0.11150	0.012	-1.485	0

Sulfur dioxide (SO2)	Weathering	3	1.96	15.03	6.5233	7.37351	54.369	1.718	0
	Unweathered	1	3.66	3.66	3.6600	0	0	0	0

2.6. Weight-prediction model building and solving

Problem 1.3 requires us to predict the pre-weathering chemical composition content of a weathered artifact based on the test data of fourteen indicators at the weathering point of the artifact after weathering and after severe weathering. We proposed two modeling ideas, one is based on the conventional machine learning algorithm to predict the chemical composition content of the artifact before weathering, and the other is based on the algorithm derived from the weight magnitude of the three indicators. Based on the weights of the indicators, we propose the weight-prediction model.

3. Conclusions

The accuracy of the chemical proportion of undifferentiated glass predicted by the prediction model based on the random forest algorithm in this study is close to 100%, which indicates the high reasonableness of the selection of our prediction model, and likewise the high accuracy of the data we brought in for training and learning; the classification model based on kmeans-hierarchical cluster analysis, there is no difference in the cluster analysis results obtained before and after the dimensionality reduction by principal component analysis, which indicates that our model The accuracy and rationality of the model are high. The results of the model are mostly presented in the form of graphs, which are highly readable and easy to understand. The model is solved by professional software such as excel, spss, stata, matlab, etc., and the reliability is high.

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