

Study on the influence of weathering process on the chemical composition of ancient glass based on CLR

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Abstract. The aim of this study was to investigate the effects of weathering processes on the chemical composition of high-potassium and lead-barium glass. First, correlation and difference analyses were performed on the surface weathering of glass artifacts in relation to their glass type, ornaments, and color, all of which showed that glass type had a significant effect on surface weathering, and that lead-barium glass was more susceptible to weathering than high-potassium glass. Then, the complex and difficult compositional data of glass artifacts were transformed into simple and easy to handle CLR data by central logarithmic ratio transformation (CLR), and independent sample t-tests or Satterthwaite approximate t-tests were performed for each chemical composition content of high-potassium glass and lead-barium glass before and after weathering, respectively, to identify the influence of weathering process on the chemical composition content of the two glasses. Finally, the chemical composition content of the weathered glass artifacts before weathering was predicted from their chemical composition content after weathering by distribution matching method and transformed back to the compositional data by CLR inversion.

Keywords: Ancient glass, Compositional data, Centered Log-ratio, Compositional prediction.

1. Introduction

Glass played an important role in the cultural communication between East and West in ancient times. The study of the composition of glass before and after weathering is of great significance to archeologists. Past studies have achieved certain results in the conservation and study of ancient glass. Many scholars have focused on the manufacturing process of ancient glass and the dating of samples [1,2]. In addition, some scholars have explored the chemical composition of ancient glass [3]. These studies provide important references for us to deeply understand the history of ancient glass, however, there are relatively few systematic studies on the effects of the weathering process of ancient glass. Therefore, this study aims to provide new perspectives and methods for the conservation and cultural research of ancient glass by conducting an in-depth study on the weathering process of ancient glass and revealing the mechanism of the effect of weathering on its chemical composition.

2. Research on the effect of weathering processes on the chemical composition of ancient glass based on CLR.composition of ancient glass

2.1. Data preprocessing

2.1.1 Data cleaning and filling

The data are all from question C of the 2022 National College Students Mathematical Modeling Competition. After cleaning the invalid data, we believed that because the measurement accuracy of measuring instruments had a certain lower limit, the vacant value could be considered as a chemical component with very low content. At the same time, because the Centered Log-ratio (CLR) involved logarithmic operation, the 0 value and the vacant value were filled as 0.01 to ensure the data was meaningful.

2.1.2 Correction of compositional data and central-log-ratio transformation

Compositional data is a class of data with complex properties, for which the component $x = (x_1, x_2, \dots, x_D)$ satisfies equation (1).

$$S^D = \{ \mathbf{x} = (x_1, x_2, \dots, x_D) | x_i \geq 0, i = 1, 2, \dots, D; \sum_{i=1}^D x_i = 1 \} \quad (1)$$

It has been proved that the sample space of the compositional data is not the usual Euclidean space, but a simplex of the so-called Aitchison geometry[4-6]. Its most characteristic feature is compositionality, i.e., the cumulative sum of the proportions of the components should be 100%. The chemical composition data of glass is a typical compositional data, and the "closure effect" caused by the constant value of the sum will lead to pseudo-correlation of the chemical data of glass, which will bias the results of the correlation between the chemical elements of glass, and also prevent multivariate statistical methods from being performed directly in the simple Euclidean space[7].

In this paper, the idea of processing glass composition data is as follows: the modified composition data are processed by Centered Log-ratio (CLR) transformation, remove the fixed sum restriction, and then analyzed, and finally reduced to composition data by inverse transformation.

The cumulative sum of component ratios due to detection means and other reasons is not 100%. Since all the information of the compositional data is embedded in the ratios between components rather than their absolute values, the cumulative sum of component ratios is corrected to 100% by equation (2).

$$\mathbf{X} = \left(\frac{x_1}{\sum_{j=1}^D x_j}, \dots, \frac{x_D}{\sum_{j=1}^D x_j} \right) \quad (2)$$

The following transformations are made to the modified component vector $\mathbf{X}=(x_1, x_2, \dots, x_D)$, $x_i > 0$ [8]:

$$\mathbf{Y} = (y_1, y_2, \dots, y_D) = \left(\ln \frac{x_1}{g(\mathbf{X})}, \dots, \ln \frac{x_D}{g(\mathbf{X})} \right) \quad (3)$$

Where $g(\mathbf{X}) = (\prod_{i=1}^D x_i)^{1/D}$ is the geometric mean of \mathbf{X} .

The resulting \mathbf{Y} is the CLR data, which has the following characteristics:

1. The CLR data is a one-to-one mapping relationship with the original compositional data, and the processed data has the same number of dimensions as the original data, and the results are more interpretable.

2. It reveals the relative information implied by the absolute data, and the larger the value indicates its higher relative content.

3. Pseudo-correlations among the elements of the original data can be identified and eliminated.

4. Transformation of the raw data into data conforming to a normal distribution facilitates the application of statistical methods for analysis.

5. The new data have a mean value of 0. Negative values indicate below average content and positive values indicate above average content.

Equation (4) allows the calculation of the values of the compositional data corresponding to the CLR data.

$$x_i = \frac{e^{y_i}}{e^{y_1} + e^{y_2} + \dots + e^{y_D}}, i = 1, 2, \dots, D \quad (4)$$

To make the data analysis easier and more intuitive, we have shifted the CLR data (+5) so that all CLR data are positive. Since equation (4) involves exponential and division operations, the final inverse transformed compositional data are not affected by the shifting process.

2.2. Analysis of the relationship between the surface weathering of glass artifacts and their ornament, type and color

This paper analyzes the relationship between the surface weathering of glass artifacts and their ornament, type and color in terms of both correlation and difference.

2.2.1 Correlation Analysis

The observed data revealed that surface weathering, ornamentation, type and color were categorical variables, and the correlations were examined by cross-tabulation analysis through

SPSS24 software, using ϕ coefficient, Crammer's V and Pearson's contingency coefficient as quantitative analysis indicators [9-11]. The three indicators were interpreted as follows:

1. ϕ coefficient is usual y used to measure the correlation of four-cel table data formed by dichotomy, but ϕ coefficient is not suitable for measuring the correlation of multi-dimensio nal frequency table.

2. Pearson's Contingency Coefficient is a correction of the ϕ coefficient, which is more suitable for multidimensional contingency table data with the same number of rows and columns.

3. Cramer's V is a method of bivariate correlation analysis. When the number of rows and columns is different, Cramer's V can be used as a correction for the ϕ coefficient.

2.2.2 difference analysis

Aiming at the difference analysis between the three categorical variables of Ornament, type and color and surface weathering, according to the applicable premise of different test methods, this paper selects the appropriate method from Pearson Chi-square test , Yates calibrated Chi-square test and Fisher exact test [12-14]

The selection criteria of Pearson Chi-square test, Yates calibrated Chi-square test and Fisher exact test are as follows:

(1) For $2 * 2 (R = 2, C = 2)$ lists of columns, use Pearson's chi-square test when $n \geq 40$ and *the expected frequencies (E) are all ≥ 5* ; use Yates' corrected chi-square test when $n \geq 40$ and *1 of the grids appears to have $1 \leq E < 5$* ; and use Fisher's exact test for any of the grids when either $E < 1$ or $n < 40$ occurs.

(2) For the $R * C$ (*any one of R, C is greater than 2; and $R \geq 2$, and $C \geq 2$*) list, use Pearson's chi-square test when *all $E > 1$ and $1 \leq E < 5$ and the proportion of grids is less than 20%*; otherwise use Yates corrected chi-square test.

The conditions of application of the three test methods are shown in Figure 1.

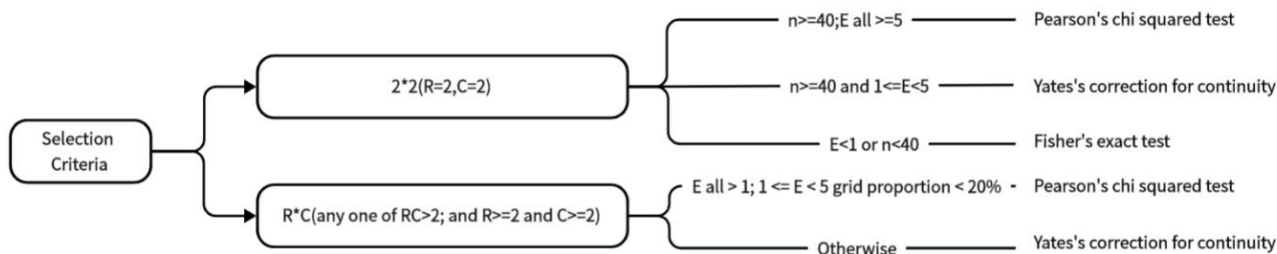


Figure 1. Applicable conditions for the three test methods

2.3. The change of chemical composition content before and after weathering based on t test

Since the glass type has a great influence on the glass composition, this paper divides the sample data into two glass types, high-potassium and lead-barium, and then identifies significant changes in the internal composition of the glass before and after weathering for each type of glass by using the independent samples t-test or the Satterthwaite's approximate t-test for each type of glass, respectively. Since the chemical compositions of the two types of glass before and after weathering obeyed normal distributions, it was only necessary to determine whether the variances before and after weathering were equal to determine which method should be chosen for the test.

Firstly, the variance homogeneity of each chemical component before and after weathering was determined by Levene test.

For chemical constituents with homogeneity of variance, the change in composition can be inferred directly using the t statistic.

$$t = \frac{\bar{X}_1 - \bar{X}_2}{\sqrt{\frac{(n_1 - 1)S_1^2 + (n_2 - 1)S_2^2}{n_1 + n_2 - 2} \left(\frac{1}{n_1} + \frac{1}{n_2} \right)}} \quad (5)$$

For chemical components of variance heterogeneity, inference is made by Satterthwaite approximate t test, using t 'statistics and correcting for degrees of freedom.

$$t' = \frac{\bar{X}_1 - \bar{X}_2}{\sqrt{\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}}} \quad (6)$$

$$v = \frac{\left(\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}\right)^2}{\frac{\left(\frac{S_1^2}{n_1}\right)^2}{(n_1-1)} + \frac{\left(\frac{S_2^2}{n_2}\right)^2}{(n_2-1)}} \quad (7)$$

2.4. Prediction of chemical composition content of glass samples before weathering based on distribution matching

Since there is no matched data of the same sample before and after weathering to establish a one-to-one link between preweathering and postweathering, and at the same time, the sample size is too small, traditional regression model or machine learning cannot be simply used, or it will lead to too large prediction bias. In this paper, we propose to use the idea of distribution matching to make predictions under the premise of satisfying only the normal distribution, the assumptions are more relaxed than the implied assumptions (normal distribution and homoscedasticity) used directly in regression or machine learning, and the results are intuitive and highly interpretable [15].

The method can be understood as the weathering process causes a change in the mean and variance of the normal population $X_j^{k_0}$ of the pre-weathering chemical compositions, and $X_j^{k_0}$ becomes the normal population $X_j^{k_1}$ of the post-weathering chemical compositions. The weathering process acts on each object stably and relatively equally, and thus its relative position on the same normal population remains unchanged, so the chemical composition $x_{ij}^{k_1}$ of a glass artifact after weathering can be inferred from its chemical composition $x_{ij}^{k_0}$ before weathering through the idea of distribution matching.

Let the normal population of each chemical composition of high-potassium unweathered glass be $X_j^{10} \sim N(\mu_{10}, \sigma_{10}^2)$, the normal population of each chemical composition of high-potassium weathered glass be $X_j^{11} \sim N(\mu_{11}, \sigma_{11}^2)$, the normal population of each chemical composition of lead-Barium unweathered glass be $X_j^{20} \sim N(\mu_{20}, \sigma_{20}^2)$, the normal population of each chemical composition of lead-Barium weathered glass be $X_j^{21} \sim N(\mu_{21}, \sigma_{21}^2)$. For the samples in the corresponding normal totals before and after weathering are:

$$x_{ij}^{k_0} = \frac{\sigma_{k_0}}{\sigma_{k_1}} (x_{ij}^{k_1} - \mu_{k_1}) + \mu_{k_0} \quad (8)$$

where x_{ij}^{kb} is the i-th object of X_j^{kb} .

3. Results

3.1. Data preprocessing results

Some of the panned CLR data are shown in Table 1.

Table 1. Partially Translated CLR Data

Heritage Sampling Sites	Types	Whether weathering	SiO2	Na2O	K2O	CaO	MgO	Al2O3	Fe2O3	CuO	PbO	BaO	P2O5	SrO	SnO2	SO2
02	Lead-Barium	Weathering	9.25	1.06	5.71	6.51	5.83	7.41	6.28	4.32	9.52	1.06	6.94	4.00	1.06	1.06
09	High-Potassium	Weathering	11.71	2.55	6.63	6.68	2.55	7.43	6.01	7.59	2.55	2.55	6.10	2.55	2.55	2.55
10	High-Potassium	Weathering	12.12	2.94	7.46	5.99	2.94	7.34	6.20	7.37	2.94	2.94	2.94	2.94	2.94	2.94
39	Lead-Barium	Weathering	9.65	1.78	1.78	6.49	1.78	5.69	1.78	6.25	10.49	8.36	6.53	5.89	1.78	1.78
56	Lead-Barium	Weathering	9.87	1.89	1.89	6.68	1.89	7.11	1.89	6.26	10.21	9.23	7.43	1.89	1.89	1.89

3.2. Results of an analysis of the relationship between the surface weathering of glass artifacts and their ornamentation, type and color

3.2.1 Results of correlation analysis

The correlation analysis results of the three variables and surface weathering properties are shown in Table 2.

Table 2. Correlation analysis of ornamentation, type and color with surface weathering properties

Analysis items	ϕ	Cramer's V	Pearson
Surface weathering * Ornaments (2*3)	-	0.326(0.059)	0.310(0.059)
Surface weathering * Types (2*2)	0.316(0.020*)	0.316(0.020*)	0.302(0.020*)
Surface weathering * Color (2*8)	-	0.341(0.581)	0.323(0.581)

Note: p values in parentheses; * means $p < 0.05$, ** means $p < 0.01$

According to the results of correlation analysis, at the significance level of 0.05, there is a significant and strong correlation between types and surface weathering, In contrast, no significant correlation exists between ornaments and surface weathering, nor between color and surface weathering.

3.2.2 Results of difference analysis

The test methods for the difference analysis between surface weathering and ornamentation, type and color were selected, and the selection results were shown in Table 3.

Table 3. Selection of chi-square test method

Analysis items	$E \geq 5$	$1 \leq E < 5$	$E < 1$	Cnt	n	Applicable test methods
Surface weathering * Ornaments (2*3)	4(66.67%)	2(33.33%)	0(0.00%)	6	54	Yates's correction for continuity
Surface weathering * Types (2*2)	4(100.00%)	0(0.00%)	0(0.00%)	4	54	Pearson's chi-square test
Surface weathering * Color (2*8)	4(25.00%)	8(50.00%)	4(25.00%)	16	54	Yates's correction for continuity

Note: Cnt refers to the total number of squares

As can be seen from the above Table 3, the applicable test method for surface weathering and ornaments is Yates corrected Chi-square test; The test method suitable for surface weathering and types is Pearson Chi-square test; Yates calibrated Chi-square test is applicable for surface weathering and color.

The results obtained from the difference analysis using the above tests respectively are shown in Table 4.

Table 4. difference analysis of the three variables with respect to surface weathering properties

First-Level Characteristics	Secondary Characteristics	Surface weathering		TOT AL	Inspection method	Statistical values
		YES	NO			
Ornaments	A	11	9	20	Yates's correction for continuity	5.747 (0.056)
	B	0	6	6		
	C	13	15	28		
	Total		30	54		
Types	Lead-Barium	12	24	36	Pearson's chi-square test	5.400 (0.020*)
	High-potassium	12	6	18		
	Total	24	30	54		
Colors	Light green	2	1	3	Yates's correction for continuity	6.287 (0.507)
	Light blue	8	12	20		
	dark green	3	4	7		
	Dark blue	2	0	2		
	Violet	2	2	4		
	Green	1	0	1		
	Blue-Green	6	9	15		
	Black	0	2	2		
	Total	24	30	54		

Note: p-values in parentheses; * indicates $p < 0.05$, ** indicates $p < 0.01$

The results of the difference analysis in Table 4 show that at the significance level of 0.05, there is a significant difference between type and surface weathering, while there is no significant difference between pattern and color and surface weathering.

The results of correlation analysis and difference analysis show that the glass type has a significant effect on the surface weathering, while the glass ornaments and color have almost no effect on the surface weathering. In addition, according to Table 4, the weathering rate of lead-barium glass is 66% and that of high-potassium glass is 33%, indicating that lead-barium glass was more susceptible to weathering than high-potassium glass.

3.3. The change rule of chemical composition content before and after weathering based on t test

Independent sample t test or Satterthwaite approximate t test were conducted on the data of the two types of glass before and after weathering, and the results were sorted out as shown in the following Table 5 and Table 6.

Table 5. Changes in chemical composition content during weathering of high-potassium glass

Significant changes		No significant change
Significant increase	Significant decrease	
SiO ₂ 、 (CuO)	(K ₂ O)	Na ₂ O、 CaO、 MgO、 Al ₂ O ₃ 、 Fe ₂ O ₃ 、 PbO、 BaO、 P ₂ O ₅ 、 SrO、 SnO ₂ 、 SO ₂

Note: Chemical components with significant changes at the significance level of 0.10 are in parentheses.

Table 6. Changes in chemical composition content during weathering of lead-barium glass

Significant changes		No significant change
Significant increase	Significant decrease	
CaO、 PbO、 P ₂ O ₅ 、 (SrO)SiO ₂ 、 Na ₂ O、 K ₂ O、 Al ₂ O ₃	MgO、 Fe ₂ O ₃ 、 CuO、 BaO、 SnO ₂ 、 SO ₂	

Note: Chemical components with significant changes at the significance level of 0.10 are in parentheses.

It can be seen from the above Table 5 and Table 6 that SiO₂, significantly increases before and after weathering of high-potassium glass. At the significance level of 0.10, the increase of CuO content and the decrease of K₂O content are also significant, while the content of other chemical components has no significant change. Before and after weathering of lead-barium glass, CaO, PbO, P₂O₅ and SrO significantly increase, while SiO₂, Na₂O, K₂O and Al₂O₃ significantly decrease. At the significance level of 0.10, the content of SrO also increases significantly, while the content of other chemical components does not change significantly.

3.4. Prediction results of chemical composition contents of glass samples before weathering based on distribution matching

The mean and variance of the CLR data to calculate the weathering and non-weathering chemical composition content of the two types of glass samples are shown in the Table 7.

Table 7. Means and standard deviations of chemical composition contents before and after weathering for the two types of glass samples

Types	Indicators	SiO ₂	Na ₂ O	K ₂ O	CaO	MgO	Al ₂ O ₃	Fe ₂ O ₃	CuO	PbO	BaO	P ₂ O ₅	SrO	SnO ₂	SO ₂
High-Potassium non-weathering	Mean value	10.09	2.68	7.59	6.60	5.26	7.70	5.61	6.20	3.51	2.98	5.56	2.30	1.73	2.20
	Standard deviation	0.63	2.58	2.05	2.36	1.72	0.37	1.70	1.42	2.00	2.27	1.61	1.04	1.93	1.70
High-Potassium weathering	Mean value	11.73	2.58	5.50	6.88	3.94	7.74	5.83	7.48	2.58	2.58	5.42	2.58	2.58	2.58
	Standard deviation	0.26	0.25	2.26	0.52	1.94	0.34	0.27	0.74	0.25	0.25	1.35	0.25	0.25	0.25
Lead-Barium non-weathering	Mean value	9.79	3.74	3.33	5.33	4.17	7.11	3.47	5.28	8.85	7.84	4.17	3.78	1.67	1.47
	Standard deviation	0.48	2.93	1.58	1.63	2.06	0.51	2.39	1.76	0.57	0.80	2.17	1.39	1.15	1.24
Lead-Barium weathering	Mean value	8.93	1.93	2.29	6.48	3.89	6.52	4.04	5.56	9.42	7.37	6.32	4.27	1.46	1.55
	Standard deviation	0.55	1.88	1.60	0.66	2.01	0.63	1.90	1.80	0.72	2.07	1.88	1.14	1.14	1.64

The CLR data form of the chemical content before weathering, predicted from equation (8), and its compositional content before weathering is shown in Table 8.

Table 8. CLR prediction data based on distribution matching

Heritage Sampling Sites	Types	Weathering prediction	SiO ₂	Na ₂ O	K ₂ O	CaO	MgO	Al ₂ O ₃	Fe ₂ O ₃	CuO	PbO	BaO	P ₂ O ₅	SrO	SnO ₂	SO ₂
02	Lead-Barium	Non-weathering	10.08	2.38	6.72	5.40	6.16	7.83	6.31	4.06	8.93	5.42	4.88	3.45	1.27	1.10
09	High-Potassium	Non-weathering	10.04	2.33	8.61	5.68	4.02	7.37	6.80	6.41	3.24	2.68	6.39	2.16	1.47	1.97
10	High-Potassium	Non-weathering	11.04	6.46	9.37	2.52	4.37	7.26	7.97	5.99	6.44	6.31	2.61	3.82	4.55	4.69
39	Lead-Barium	Non-weathering	10.43	3.51	2.83	5.33	2.01	6.43	0.62	5.95	9.69	8.22	4.41	5.76	2.00	1.64
56	Lead-Barium	Non-weathering	10.62	3.68	2.94	5.82	2.12	7.59	0.76	5.96	9.47	8.56	5.45	0.87	2.11	1.73

The CLR prediction data is transformed back to the compositional data representation by equation (4), as shown in Table 9.

Table 9. Predicted data of components obtained by inversion process

Heritage Samplin g Sites	Types	Weathering prediction	SiO ₂	Na ₂ O	K ₂ O	CaO	MgO	Al ₂ O ₃	Fe ₂ O ₃	CuO	PbO	BaO	P ₂ O ₅	SrO	SnO ₂	SO ₂
02	Lead- Barium	Non- weathering	78.89	0.14	3.57	0.02	1.06	5.72	0.90	0.07	7.63	1.94	0.02	0.03	0.01	0.00
09	High- Potassi- um	Non- weathering	70.51	0.03	16.86	0.90	0.17	4.86	2.78	1.87	0.08	0.04	1.83	0.03	0.01	0.02
10	High- Potassi- um	Non- weathering	77.02	0.79	14.46	0.02	0.10	1.76	3.59	0.49	0.77	0.68	0.02	0.06	0.12	0.13
39	Lead- Barium	Non- weathering	82.21	0.61	0.06	0.01	0.01	1.40	0.00	0.35	10.61	4.37	0.01	0.33	0.01	0.01
56	Lead- Barium	Non- weathering	83.75	0.69	0.06	0.04	0.01	3.11	0.00	0.31	7.71	4.28	0.02	0.00	0.01	0.01

4. Conclusion

Based on the chemical composition ratio data of classified glass relics, the effect of weathering process on the chemical composition content of high-potassium glass and lead-barium glass was studied in this study. Firstly, the correlation analysis and difference analysis of the relationship between the surface weathering of glass relics and the glass type, ornaments and color are carried out. The results all show that glass type had a significant effect on surface weathering, and that lead-barium glass was more susceptible to weathering than high-potassium glass. Then, the complex and difficult compositional data of the glass relics were transformed into simple and easy to handle CLR data by central log-ratio transformation (CLR). Independent sample T-test or Satterthwaite approximate T-test were conducted for the chemical component contents of the high-potassium glass and lead-barium glass before and after weathering, respectively. The chemical compositions of the two kinds of glass that changed significantly during weathering were identified. Finally, according to the chemical composition content of weathered glass relics, the chemical composition content before weathering is predicted by distribution matching method, and it is converted back to the composition data by CLR inverse transformation.

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