

Elemental Characteristics of Atmospheric Inhalable Particulate Matter at the Mogao Caves, Dunhuang: Postprint

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Abstract

The mass concentrations of 27 elements in atmospheric inhalable particulate matter inside and outside the cave chambers of the Dunhuang Mogao Grottoes were determined using inductively coupled plasma mass spectrometry. The results indicate that typical crustal elements Na, Mg, Al, K, Ca, and Fe accounted for over 96% of the total mass concentration of all measured elements. Comparative analysis of elemental mass concentrations in PM_{2.5} and PM_{10-2.5} inside and outside the caves revealed that elements including Li, Na, Mg, K, Ca, Co, Ni, Zn, Sr, Cs, Ba, and U were predominantly present in PM_{10-2.5}. During dust storm events, the mass concentrations of Co, Ga, As, Cs, and Ba in the external cave environment decreased, suggesting that the dust process exerts a dilution effect on these elements. Enrichment factor analysis demonstrated that the EF values for B, Na, Ca, Mn, Cu, Zn, As, Sr, Tl, Pb, Bi, and U in inhalable particulate matter both inside and outside the caves exceeded 10, indicating that pollutants from anthropogenic activities were the primary drivers of enrichment for these elements. Mineral pigments rich in Ca, Mn, Cu, Zn, As, and Pb were extensively utilized in the creation of cave murals, and also contributed to the enrichment of trace elements in regional atmospheric inhalable particulate matter. A significant linear correlation was observed between tourist numbers and Bi concentrations inside and outside the caves, demonstrating that variations in environmental Bi concentrations are influenced by tourist activities. This study represents the first systematic monitoring and analysis of elemental contents in atmospheric inhalable particulate matter at the Mogao Grottoes, and the findings provide a scientific foundation for the management and control of inhalable particulate matter pollution affecting grotto cultural heritage and their surrounding environments.

Full Text

Chemical Elemental Characteristics of Atmospheric Inhalable Particulates in Dunhuang Mogao Grottoes

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Abstract

This study measured the mass concentrations of 27 elements in atmospheric inhalable particulates inside and outside the caves of Dunhuang Mogao Grottoes using inductively coupled plasma mass spectrometry. The results revealed that typical crustal elements accounted for over 96% of the total measured elemental mass concentration. During dust weather events, the mass concentrations of elements such as Li, Be, and Zn increased by more than fourfold, while the concentrations of Co, Ga, As, Cs, and Ba decreased, indicating that dust processes have a dilution effect on these elements. Enrichment factor analysis showed that elements including B, Na, Ca, Mn, Cu, Zn, As, Sr, Tl, Pb, Bi, and U had enrichment factor values greater than 10, suggesting that pollutants from human activities play a major role in the enrichment of these elements. Mineral pigments rich in elements such as Pb and Zn have been extensively used in cave mural production, which may also contribute to the enrichment of trace elements in regional atmospheric inhalable particulates. Tourist numbers showed a significant linear correlation with Bi concentrations both inside and outside the caves, indicating that visitor activities influence environmental Bi concentrations. This research represents the first systematic monitoring and analysis of elemental content in atmospheric inhalable particulates at the Mogao Grottoes, providing a scientific basis for the management and control of inhalable particulate pollution affecting the grottoes and their preservation environment.

Keywords: Dunhuang Mogao Grottoes; inhalable particulates; chemical elements; enrichment factor

Introduction

As research on the environmental impacts of atmospheric particulate matter deepens, conservation experts worldwide have increasingly focused on the effects of atmospheric particles on cultural heritage preservation environments. Due to limitations in preservation methods and conditions, most cultural relics and historic sites are exposed to ambient air, making them vulnerable to various natural disasters and necessitating scientific and preventive conservation measures. Atmospheric suspended particulates significantly influence aerosol chemical composition, air quality, and climate change, thereby threatening the long-term preservation of cultural artifacts. As early as the mid-19th century, the British National Gallery began investigating the corrosive effects of indoor air on cultural objects. Researchers have collected airborne particulate samples at the Alhambra monument, analyzing their composition using micro-Raman spectroscopy and electron probe X-ray microanalysis (EPXMA), which revealed that certain chemical components pose risks to heritage materials. Domestic studies have demonstrated that atmospheric SO_2 is one of the causes of erosion and micro-crack formation on painted cultural relics' lacquer surfaces. Li Hua and colleagues conducted in-depth research on indoor microclimate conditions, aerosol mass concentrations, and chemical composition characteristics at the Qin Terracotta Warriors Museum and Han Yangling Museum, evaluating the long-term impacts of museum tourism planning and environmental improvement policies on heritage preservation atmospheres by examining their evolution over 15 years. Cao Junji et al. monitored the atmospheric environment in an undisturbed tomb chamber of Zhang Anshi' s cemetery from the Western Han Dynasty, providing scientific evidence for subsequent archaeological excavations.

Dunhuang Mogao Grottoes, a world-renowned cultural heritage site, is located on the edge of the Kumtag Desert in an extremely arid region with perennially low rainfall. Spring and summer seasons frequently experience dust weather events, with strong winds and sand accelerating the erosion and contamination of cliff bodies and cave murals and sculptures. In recent years, our research group has monitored and analyzed seasonal variations in atmospheric particulates $\text{PM}_{2.5}$ and PM_{10} , dynamic distributions of soluble salt ions, and microbial community characteristics in particulate matter inside and outside the Mogao Grottoes caves. However, in-depth analysis of elemental composition in atmospheric particulates and their mechanisms of mural deterioration has not yet been conducted. Previous studies have shown that smaller particulate matter more readily adsorbs harmful substances from the air, and once deposited on artifact surfaces, accelerates deterioration processes. Particulates enriched with transition metal elements can catalyze and accelerate chemical reactions between pollutant gases and artifact surface materials. Meanwhile, the influence of dust weather on the types and contents of elements in airborne particulates remains unclear, making research on the chemical elemental composition and variation patterns of inhalable particulates in the Mogao Grottoes environment particularly necessary.

This study monitors inhalable particulates inside and outside the Mogao Grottoes caves, measuring the content of 27 elements to investigate variations in elemental composition under the influence of seasonal, climatic, and human activity factors, as well as the impacts of dust weather and tourists on elemental concentrations. The aim is to provide scientific atmospheric environmental data for the protection of Mogao Grottoes artifacts and their preservation environment.

1. Materials and Methods

1.1 Sampling Site Setup

Sampling was conducted at Cave 16 inside the Mogao Grottoes and at a meteorological station outside the caves. The sampling height was 1.5 m above ground level at both locations. The sampling site locations are shown in [Figure 1: see original paper].

1.2 Sample Collection

Atmospheric particulate matter was collected using a Dichotomous Partisol-Plus Model 2025 sampler (Thermo Scientific, USA). This instrument simultaneously collects $PM_{2.5}$ and $PM_{10-2.5}$ samples. The flow rate was set at $15\text{ L} \cdot \text{min}^{-1}$ for PM_{10} and $1.67\text{ L} \cdot \text{min}^{-1}$ for $PM_{2.5}$. Samples were collected on polytetrafluoroethylene (PTFE) membranes (Whatman, USA) with a diameter of 47 mm.

Sampling was conducted from mid-March to late December. According to ambient air quality conditions, sampling duration was set to 24 h in spring (March-May) and summer (June-August), and 168 h in autumn (September-November) and winter (December-February). Dunhuang frequently experiences dust weather in spring and summer, so separate collections of atmospheric particulates inside and outside the caves were conducted during sandstorm and blowing sand events, with collection periods set from the beginning to the end of each dust event.

Before sampling, membranes were balanced in a desiccator for 24 h and weighed using an electronic balance (auy220, SHIMADZU, Japan). Weighing was repeated until the mass difference between three consecutive measurements was within $\pm 0.4\text{ mg}$, with the average value recorded. After sampling, membranes were again balanced in a desiccator for 24 h and weighed following the same procedure. Weighed membranes were stored individually in clean membrane boxes, sealed, and refrigerated until analysis.

1.3 Sample Elemental Analysis

To investigate seasonal variations of elements in inhalable particulates at the Mogao Grottoes, half of each PTFE membrane was cut and placed in a con-

tainer with 15 mL of 5% HNO_3 . The container was ultrasonically extracted for 20 minutes at 30°C, repeated three times. After ultrasonication, the container was removed, shaken thoroughly, and allowed to cool to room temperature. The solution was then filtered, and the mass concentrations of 27 elements in the particulates were determined using inductively coupled plasma mass spectrometry (ICP-MS, X-7, Thermo Elemental, USA).

2. Results and Discussion

2.1 Elemental Composition of Inhalable Particulates at Mogao Grottoes

The mass concentrations of 27 elements in $\text{PM}_{2.5}$ and $\text{PM}_{10-2.5}$ collected inside and outside the caves are shown in . The elements with highest concentrations both inside and outside the caves were Na, Mg, Al, K, Ca, and Fe—typical crustal elements. These six elements accounted for 96.4% of total elemental mass concentration in outside samples and 96.6% in inside samples, with minimal difference between locations. Calcium exhibited the highest concentration at approximately 60%, with values of $3,397.78 \text{ ng} \cdot \text{m}^{-3}$ outside and $2,985.03 \text{ ng} \cdot \text{m}^{-3}$ inside. This predominance of crustal elements aligns with findings from the Alhambra World Heritage site. The Mogao Grottoes are located about 25 km from Dunhuang city, surrounded by Gobi desert without industrial pollution sources, indicating these crustal elements originate primarily from natural sources.

Dust weather significantly impacted elemental mass concentrations. During dust events from March to May, mass concentrations of elements such as Li, Be, Zn, and others increased by more than four times compared to non-dust conditions, while concentrations of Co, Ga, As, Cs, and Ba decreased. During sandstorms, atmospheric particulates from the Kumtag Desert are transported northeastward, increasing particulate mass concentrations at the Mogao Grottoes. The strong air currents associated with dust weather have a dilution effect on these elements, reducing their mass concentrations in inhalable particulates. The concentration changes of elements such as B, Sc, Ga, As, Cs, Tl, Pb, and Bi were not significant and remained essentially unaffected by dust processes.

2.2 Seasonal Variation Trends of PM_{10} Elemental Concentrations

Comparisons of $\text{PM}_{2.5}$ and $\text{PM}_{10-2.5}$ concentrations between the external environment and Cave 16 are shown in [Figure 2: see original paper]. Seasonal variations in atmospheric particulate elemental concentrations followed similar trends both inside and outside the caves, with greater fluctuations in spring and summer due to extreme weather events, while autumn and winter showed more stable patterns.

The concentrations of elements such as B, Na, Mg, Al, K, Ca, Ti, Mn, Fe, and

Sr exhibited higher values in spring and lower values in winter. In contrast, elements including Sc, V, Cr, Co, Ni, Cu, Zn, As, Mo, Cd, Sn, Sb, Ba, Tl, Pb, Bi, and U showed higher concentrations in winter and lower concentrations in spring. Notably, the winter concentration of element Bi was approximately 15 times higher than in spring. The Mogao Grottoes tourist area experiences its off-season in winter with fewer visitors and vehicles, but surrounding rural areas within 15 km begin coal-fired heating, causing increases in coal-related elements such as As, Mo, Cd, Sn, Sb, and Bi. The highest concentrations of the three most abundant elements (Ca, Fe, Al) occurred in spring, with external concentrations of $7,302.91 \text{ ng} \cdot \text{m}^{-3}$, $2,355.17 \text{ ng} \cdot \text{m}^{-3}$, and $1,910.13 \text{ ng} \cdot \text{m}^{-3}$, respectively, and internal concentrations of $5,247.47 \text{ ng} \cdot \text{m}^{-3}$, $723.57 \text{ ng} \cdot \text{m}^{-3}$, and $1,274.45 \text{ ng} \cdot \text{m}^{-3}$, respectively. Elements such as V, Ni, Cu, Zn, As, Mo, Cd, Sn, Sb, and Bi showed highest concentrations in summer outside the caves but in spring inside the caves, while concentrations of Na, Mg, Al, K, Ca, Ti, Mn, Fe, and Sr showed minimal seasonal variation.

2.3 Distribution of Elements in Coarse ($\text{PM}_{10-2.5}$) and Fine ($\text{PM}_{2.5}$) Particulates

Inhalable particulates are divided into two size fractions: coarse particles ($\text{PM}_{10-2.5}$, 2.5–10 μm) and fine particles ($\text{PM}_{2.5}$, $<2.5 \mu\text{m}$). Analysis of elemental mass concentrations in fine and coarse particulates collected inside and outside the caves in 2015 revealed distinct distribution patterns [Figure 4: see original paper]. Elements such as Na, Mg, Al, K, Ca, Ti, Mn, Fe, Sr, and Ba primarily existed in coarse particulates ($\text{PM}_{10-2.5}$), though the proportions differed between internal and external environments. Elements including V, Ni, Cu, Zn, As, Mo, Cd, Sn, Sb, Tl, Pb, Bi, and U were mainly distributed in fine particulates ($\text{PM}_{2.5}$).

Comparative analysis showed that in external samples, the concentration of element Bi in coarse particulates ($0.0077 \text{ ng} \cdot \text{m}^{-3}$) was greater than in fine particulates ($0.0039 \text{ ng} \cdot \text{m}^{-3}$). Inside the caves, coarse particulate Bi concentration was $0.0045 \text{ ng} \cdot \text{m}^{-3}$ while fine particulate concentration was $0.0053 \text{ ng} \cdot \text{m}^{-3}$, showing an opposite distribution pattern compared to external samples. During dust weather, the percentage increase of these elements in coarse particulates inside the caves was significantly lower than outside, indicating that cave doors, windows, and long passageways impede the transport of $\text{PM}_{10-2.5}$. The mass concentration of $\text{PM}_{10-2.5}$ decreased during dust events, demonstrating a dilution effect on these elements. Conversely, the percentage of automotive exhaust-related elements such as Pb and Zn in coarse particulates generally increased during dust weather, likely due to transport from external areas or Dunhuang city by strong convective weather processes and winds.

Element Bi, a confirmed carcinogen, is a light metal that easily disperses into the air, posing significant health risks. The primary atmospheric sources of Bi are coal and petroleum development and utilization. Some pigments used in Mogao Grottoes murals contain trace amounts of Bi, which may contribute to

Bi concentrations in atmospheric particulates. The percentage difference of Bi between coarse and fine particulates was minimal both inside and outside the caves, suggesting particle size has limited influence on Bi enrichment in airborne particulates.

2.4 Correlation Between Tourist Activities and Elemental Mass Concentrations

Correlation analysis was conducted between tourist numbers during the sampling period and elemental concentrations in coarse ($PM_{10-2.5}$) and fine ($PM_{2.5}$) particulates inside and outside the caves. Among the 27 elements, only Bi showed a significant positive correlation with visitor numbers [Figure 6: see original paper]. Comparison of elemental mass concentrations between non-dust and dust weather samples [Figure 5: see original paper] revealed that in the external environment, most elements showed increased percentages in coarse particulates during dust events, except for B, Sc, Ga, As, Cs, and Tl, which showed decreased percentages. Element Bi, which primarily exists in $PM_{2.5}$, showed significantly reduced mass concentration during dust weather, with increased content in coarse particulates, indicating that strong convective weather dilutes Bi, particularly in fine particulates.

These findings demonstrate that tourist activities contribute to Bi concentrations in the grottoes area. Bismuth is a non-toxic, non-carcinogenic element widely used in metallurgy, chemicals, electronics, aerospace, and medicine. Bismuth oxychloride is a pearlescent pigment widely used in cosmetics due to its non-toxic properties. The Bi in the Mogao Grottoes atmospheric environment likely originates from cosmetics used by tourists. During non-dust weather, Bi concentration was $0.0163 \text{ ng} \cdot \text{m}^{-3}$, increasing to $0.0453 \text{ ng} \cdot \text{m}^{-3}$ during dust events. Inside the caves, dust weather had less pronounced effects on elemental percentages, though Bi percentage in coarse particulates increased.

2.5 Enrichment Factor Analysis of Elements in Inhalable Particulates

Enrichment factor (EF) analysis is used to study elemental enrichment in atmospheric particulates and evaluate contributions from natural versus anthropogenic sources, first applied by Zoller et al. in studying Antarctic atmospheric particulate sources. EF is calculated as:

$$EF_X = \frac{(C_X/C_R)_{aerosol}}{(C_X/C_R)_{crust}}$$

where C_X represents the element being evaluated, C_R represents the reference element, and EF_X is the enrichment factor value. Reference elements are typically abundant crustal elements with minimal anthropogenic interference, stable chemical properties, and low volatility, commonly Fe, Al, or Si. This study selected Fe as the reference element.

EF analysis revealed that enrichment levels inside and outside the caves were generally consistent. Elements with EF values less than 10 (Sc, V, Cr, Co, Ni, Ga, Rb, Sr, Zr, Mo, Cd, Sn, Cs, Ba, and U in descending order) originated primarily from crustal sources, mainly from weathered soil or rock. Elements with EF values greater than 10 (B, Na, Ca, Mn, Cu, Zn, As, Tl, Pb, and Bi in ascending order) showed enrichment from anthropogenic pollutant emissions. Research indicates that Pb and Zn primarily originate from coal combustion and vehicle emissions; As mainly comes from fuel oil, waste incineration, and metal smelting; and Cu primarily originates from coal combustion.

Pb is a persistent toxic element and the only heavy metal with restricted concentration limits in China's ambient air quality standards. Excessive Pb intake can cause multi-organ system damage, particularly to the brain, kidneys, and blood system, warranting attention and pollution control measures. The EF values for B, Na, Mg, Al, K, Ca, Ti, Mn, Fe, and Sr were higher inside the caves than outside, indicating that the internal cave environment influences enrichment of these elements. Mineral pigments rich in Pb and Zn (such as $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, $2\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2$, and Pb_3O_4) were extensively used in mural production and may contribute to enrichment of trace elements in environmental aerosols. Tourist movement inside caves continuously resuspends floor particulates. The EF value for element Bi was greater than 10 outside but less than 10 inside, suggesting Bi enrichment is associated with human activities in the external environment.

3. Conclusions

This study represents the first systematic sampling and analysis of 27 elements in inhalable particulates inside and outside Cave 16 at the Dunhuang Mogao Grottoes. The main conclusions are:

- 1) Typical crustal elements (Na, Mg, Al, K, Ca, Fe) were the dominant components in inhalable particulates both inside and outside the caves, accounting for over 96% of total elemental mass concentration, with Ca being the most abundant at approximately 60%. Elements such as B, Na, Ca, Mn, Cu, Zn, As, Sr, Tl, Pb, Bi, and U showed higher concentrations in spring and lower concentrations in winter, while elements including Sc, V, Cr, Co, Ni, Cu, Zn, As, Mo, Cd, Sn, Sb, Ba, Tl, Pb, Bi, and U exhibited higher concentrations in winter and lower concentrations in spring. Notably, Bi winter concentrations were about 15 times higher than spring concentrations.
- 2) Elements such as Na, Mg, Al, K, Ca, Ti, Mn, Fe, Sr, and Ba primarily existed in coarse particulates ($\text{PM}_{10-2.5}$). During dust weather, these elements showed significantly lower percentage increases inside caves compared to outside, indicating that cave doors, long passageways, and win-

dows impede $PM_{10-2.5}$ transport. Dust weather reduced the mass concentration of these elements, demonstrating a dilution effect.

- 3) Bismuth concentrations showed a significant positive correlation with tourist numbers, with EF values greater than 10 and higher inside caves than outside. Large numbers of tourist visits contribute to Bi enrichment in the grottoes environment.
- 4) Enrichment factor values for elements such as Sc, V, Cr, Co, Ni, Ga, Rb, Sr, Zr, Mo, Cd, Sn, Cs, Ba, and U were less than 10, indicating these elements primarily originated from crustal sources. Elements including B, Na, Ca, Mn, Cu, Zn, As, Tl, Pb, and Bi had EF values greater than 10, indicating that pollutant emissions from human activities significantly influenced the enrichment of these elements.

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