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Article

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Non-destructive identification of gabbroic building stone using portable X-ray fluorescence (pXRF) analysis

Shoji Nishimoto[1] ✉, **Mutsuko Inui**[2], **Tsutomu Nakazawa**[3], **Amana Hiraga**[4] & **Hiroyuki Yamashita**[5]

Stone materials in architecture are increasingly recognized as cultural and geological heritage, but preservation concerns often limit traditional sampling-based analyses. This study evaluates the utility of portable X-ray fluorescence (pXRF) as a non-destructive method to identify three gabbroic stone materials used in the early 20th-century Japanese modern architecture by comparing them with stone blocks preserved by a historical stone company as reference samples. The results demonstrate that the stones can be successfully distinguished based on elemental ratios such as Rb/Sr and K_2O/CaO. In-situ analysis at a well-known modern building in Japan further confirms the practicality of this approach. The stone materials preserved from historical distribution proved effective for comparative geochemical analysis, particularly when the precise quarry location is unknown. The preservation and scientific study of these rare reference samples are vital not only for accurate provenance analysis but also for safeguarding the geological diversity embedded in architectural heritage.

In the fields of geoheritage and architectural conservation, stone materials used in historical and modern buildings are increasingly recognized as significant cultural and geological assets[1–3]. Identifying these stones not only contributes to the preservation of cultural heritage but also enhances our understanding of historical quarrying, distribution, and construction practices[4,5]. Thus, documenting building stones from a geological perspective is becoming increasingly important. Yet, sampling limitations often hinder the petrographic and mineralogical analyses of building stones. Consequently, many studies primarily rely on documentary records or visual observations, which can be inconclusive or inaccurate. Hence, a scientific method for stone identification is required; however, owing to preservation concerns, only non-destructive, in-situ analyses are permitted.

Portable X-ray fluorescence (pXRF) is a representative non-destructive analysis method for the scientific identification of stone materials. Its reliability has been demonstrated in archaeological research on obsidian species[6–9] and ceramics[10,11]. However, its utility for other types of stone has not yet been fully explored, as demonstrated by the limited number of studies[12,13]. Although there are some examples of provenance studies of building stone materials in sandstone[14–16], there are very few examples of its application to the identification of building stones used for modern architecture, where a wider variety of crystalline rocks, such as gabbro or granite, are employed. No previous studies have reported the identification of gabbroic stone materials based on elemental ratios using pXRF. To enhance the historical value of stone cultural heritage, it is necessary to expand the range of stone materials applicable to pXRF-based provenance surveys. In these studies using pXRF analysis[12–17], elemental ratios such as Rb/Sr are employed as geochemical indicators, as they are considered to be relatively robust against sample heterogeneity, because elemental concentrations (i.e., chemical compositions) obtained by pXRF analysis are known to be susceptible to factors such as weathering, surface contamination, and grain size[17–19].

In modern architecture, a greater variety of stone materials came into use due to advances in quarrying and transportation [20], making it necessary to expand the range of stones for which provenance analysis is possible. However, these building stones often have flattened surfaces that are relatively uncontaminated and unweathered, which makes them better suited for pXRF analysis than archaeological artifacts. The application of the pXRF method to such suitable materials makes it easier to build a comprehensive database of diverse stone types, which can, in turn, be fed back into the identification of archaeological stone artifacts.

During the period of modernization in early 20th-century Japan, many buildings in a Western architectural style were constructed, and the growing demand for building stones led to the development of new quarries[21,22]. Many of these structures, however, were lost either during World War II or during the subsequent war reconstruction period. Despite their historical significance and continued use in restoration, the stone materials used in these buildings have rarely been subjected to scientific identification using non-destructive methods such as pXRF. Consequently, some of the

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[1]Aichi University, Nagoya, Aichi, Japan. [2]Kokushikan University, Setagaya, Tokyo, Japan. [3]Geological Survey of Japan, AIST, Tsukuba, Ibaraki, Japan. [4]Institute of Science Tokyo, Meguro, Tokyo, Japan. [5]Kanagawa Prefectural Museum of Natural History, Odawara, Kanagawa, Japan. ✉e-mail: shojin@aichi-u.ac.jp

remaining historical buildings are now being restored without clear information on the originally used stone materials.

Fortunately, some of the stones used in modern architecture were preserved by the Yabashi Marble Co., Ltd., a stone company founded in 1901, making them available as reference samples. Using these stone samples, the applicability of pXRF to identify gabbroic stone materials used in modern architecture was assessed. Initially, our focus was on gabbroic stone materials, often referred to as 'black granite', as only a few types were widely used in early 20th-century Japanese architecture. The results confirmed that the method can successfully distinguish between different types. This study marks the first step in evaluating the suitability of pXRF analysis for identifying the various crystalline rocks commonly used for cladding in modern Japanese architecture.

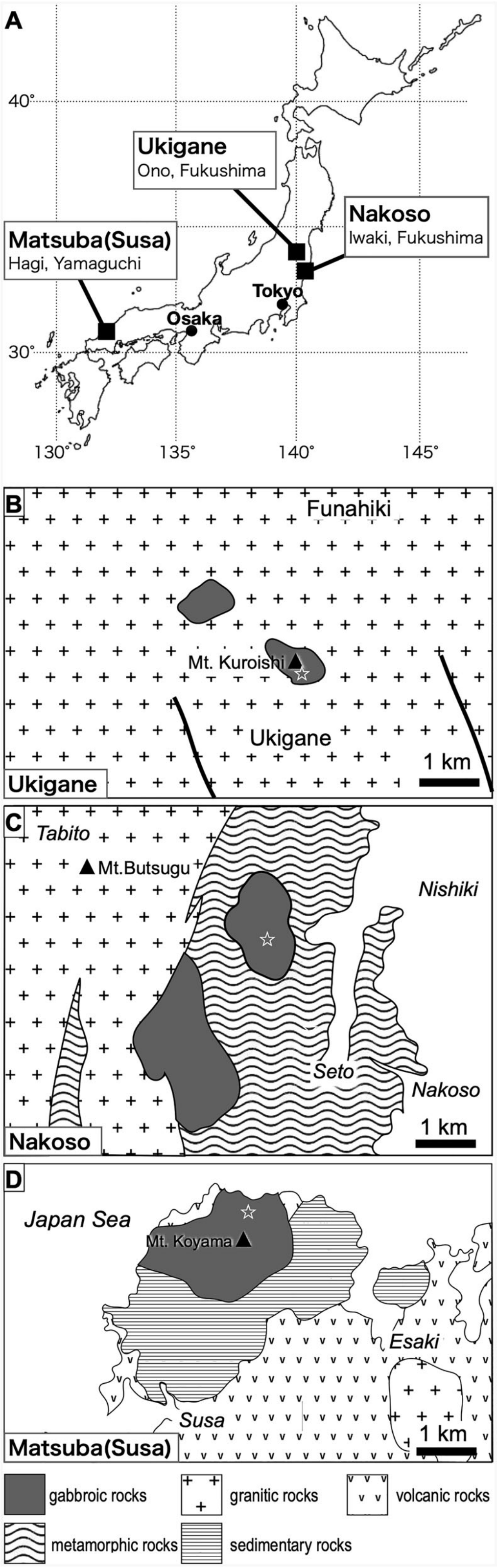


Fig. 1 | Geological locations of the historical gabbroic stones used in modern Japanese architecture. **A** Location map of Japan. Simplified geological maps of the quarry areas for three historical gabbroic stones: **B** Ukigane, **C** Nakoso, and **D** Matsuba (Susa). Each quarry site is indicated by a star. Adapted from Geological Survey of Japan, AIST (2023)[37].

Methods

Reference materials

Three gabbroic stone materials, Ukigane, Nakoso, and Matsuba (Susa), which were historically used in modern buildings during Japan's early 20th-century modernization period, were recognized and selected from stockpiled stone blocks kept in the storage yard of Yabashi Marble Co., Ltd. in Ogaki, Gifu Prefecture, which is the oldest building stone company in Japan. Figure 1 shows the approximate or inferred quarry locations of the three stone materials with simplified geological maps of their surrounding areas. Figure 2 shows the polished surfaces and photomicrographs of each stone material.

Ukigane is a building stone still quarried on Mt. Kuroishi-yama in Ono town, Fukusima Prefecture. It is geologically a small body of hornblende gabbro within the Cretaceous granitic rock widely distributed in the southern Abukuma Mountains[23]. Macroscopically, it is usually a massive, dark-colored rock (Fig. 2A). The lithology ranges from fine-grained to pegmatitic, even in the quarry outcrops, and the Schlieren structure is observed. Microscopically, the gabbro displays an equigranular texture composed of hornblende and plagioclase, with grain sizes of ~1–3 mm (Fig. 2a).

Nakoso is a building stone probably quarried around the Nakoso area of Iwaki city, Fukushima Prefecture, although the exact source is unclear. It is considered to originate from a small body of hornblende gabbro, which is part of the Tabito igneous complex[24] distributed in the northern Abukuma Mountains. Macroscopically, it is a massive, dark-colored rock (Fig. 2B) that occasionally exhibits schlieren structures, which are streaky or banded features formed by the alignment or concentration of plagioclase and mafic minerals during magmatic flow. Microscopically, it displays a subophitic texture in which larger hornblende crystals enclose euhedral, elongated plagioclase grains <1 mm in length (Fig. 2b).

Matsuba, or Susa, is a building stone quarried in the Susa area of Hagi City, Yamaguchi Prefecture. Geologically, it is a pyroxene gabbro that intrudes into sedimentary rocks[25]. Macroscopically, it is a massive, dark-colored rock (Fig. 2C) but appears lighter in color than Nakoso and Ukigane. Microscopically, it exhibits an equigranular texture composed mainly of plagioclase with grain sizes of ~1–3 mm and minor amounts of smaller pyroxene, hornblende, and biotite (Fig. 2c).

These selected stones, used as reference samples, were cut into postcard-sized plates and polished by the company. They were analyzed using a pXRF analyzer (Vanta™ by Evident) equipped with a Rh-anode X-ray tube at Kokushikan University in GeoChem mode utilizing the fundamental parameters (FP) method [26,27]. The instrument cycled through three beam settings: Beam 1 (40 kV, Al 2 mm filter) for mid-weight elements such as Rb, Sr, Fe, Mn and Ti; Beam 2 (10 kV, no filter) for light elements including Si, Al, Mg, K, Ca, and P; and Beam 3 (50 kV, Cu 350 μm filter) for heavy elements such as Ba and selected rare earth elements. Each measurement was conducted using a 9 mm analysis spot that lasted 50 seconds (30 s for Beam 1, 10 s each for Beams 2 and 3). Spectral inspection confirmed the presence of clear peaks even for trace elements such as Rb and Sr (Fig. 3). Because plutonic rocks such as gabbro have coarse mineral

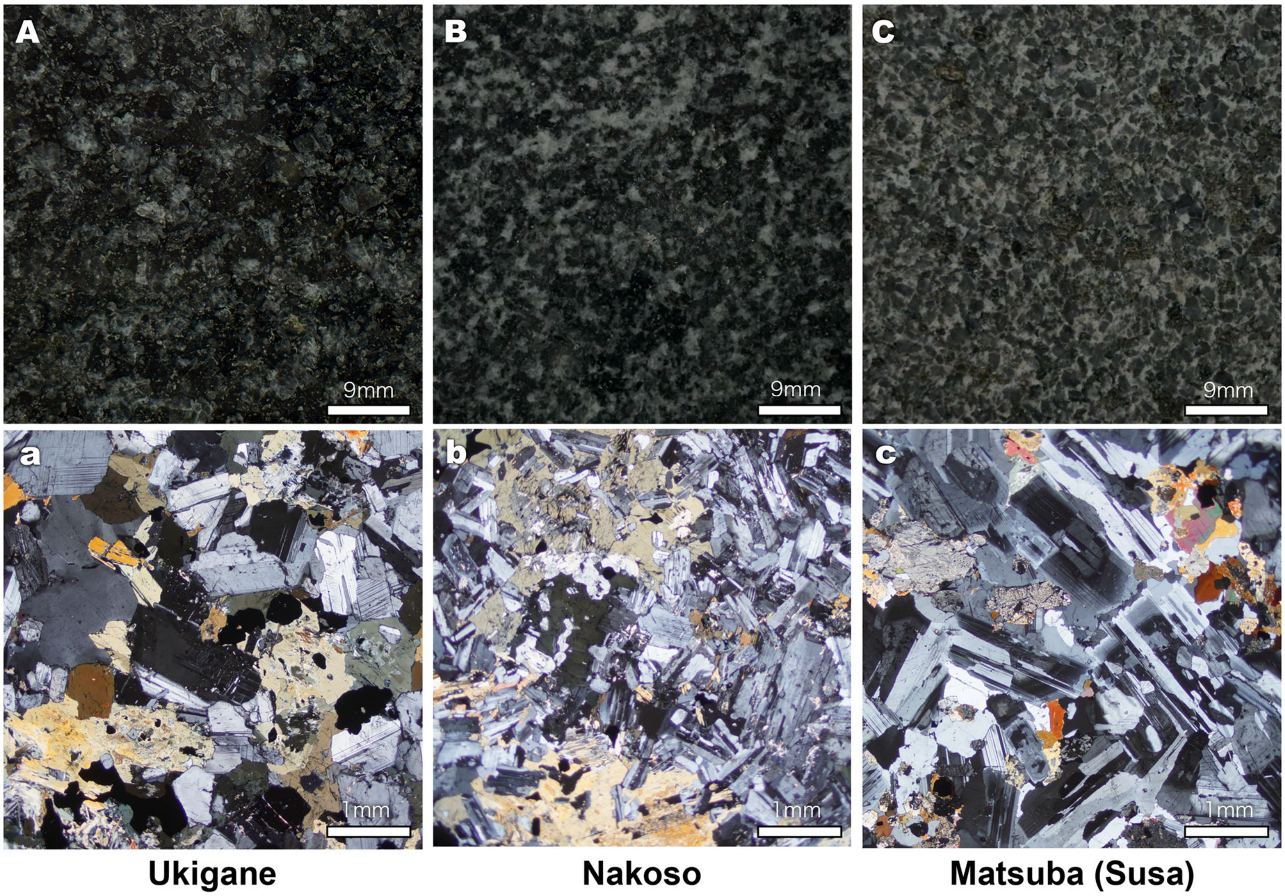
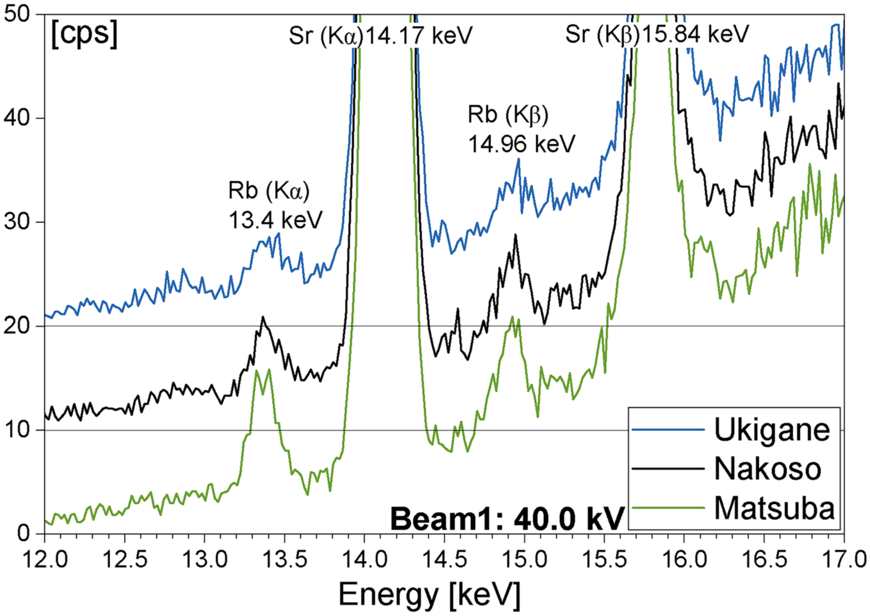


Fig. 2 | Representative appearance and textures of the three gabbroic stones used in early 20th-century Japanese architecture. Polished surfaces of gabbroic building stones: **A** Ukigane, **B** Nakoso, and **C** Matsuba (Susa). Crossed-polarized photomicrographs of the same stones: **a** Ukigane, **b** Nakoso, and **c** Matsuba. Scale bars: 9 mm for **A–C**, 1 mm for a–c.

Fig. 3 | X-ray fluorescence spectra in the energy range of 12.0–17.0 keV, covering the peaks of Rb and Sr, obtained with Beam 1 (40 kV). The spectra are vertically offset to enhance visual clarity.



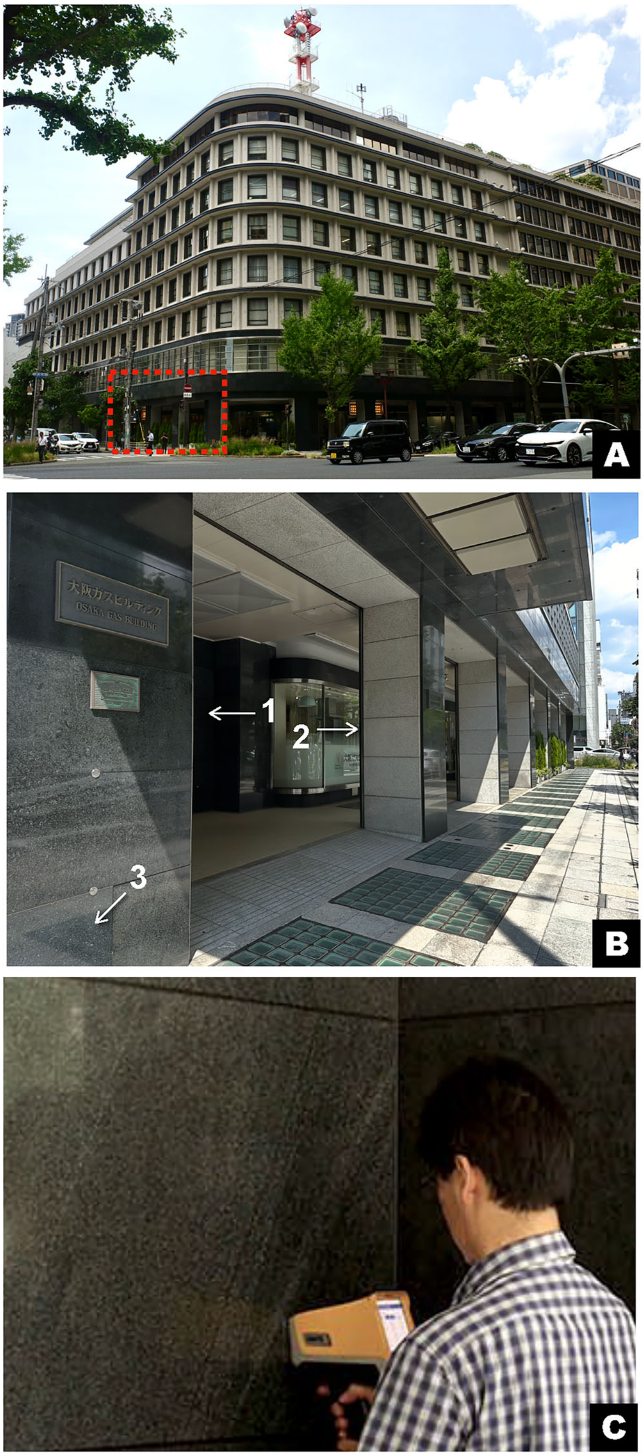


Fig. 4 | In-situ analysis points on gabbroic stone ("black granite") used as exterior wall cladding of the Osaka Gas Building. **A** Overall view of the building. The red dotted line indicates the area shown in (**B**). **B** Close-up view of the south façade, with numbered points indicating the measurement locations: (1) an area with a visible schlieren structure, (2) an area with a uniform massive structure, and (3) an area that appears to have been repaired. **C** In situ pXRF measurement on the cladding stone, where a schlieren structure is visible.

grains, the measured composition varies depending on the mineral phases within the irradiation area. Previous studies on granite have suggested that 5–20 measurements per sample are sufficient to obtain representative data[28,29]. Accordingly, twenty measurements per sample were conducted at randomly selected points on the polished surface.

For the analysis of bulk chemical compositions, the reference samples were first coarsely ground in a tungsten carbide mortar to a particle size of ~2–0.5 mm. They were then ultrasonically cleaned with distilled water, dried overnight at 95 °C, and further pulverized in an agate mortar to obtain a fine powder. Bulk chemical analysis was performed using a wavelength-dispersive X-ray fluorescence spectrometer (WD-XRF; Rigaku ZSX Primus II) with glass bead preparation at the Kanagawa Prefectural Museum of Natural History. The analytical conditions were a tube current of 60 mA and a maximum voltage of 50 kV. Each powdered sample was mixed with a lithium tetraborate flux at a ratio of 1:5, melted at 1200 °C, and cast into a glass disk prior to analysis.

In situ analysis

To validate this approach, an in-situ analysis was conducted at the Osaka Gas Building (Fig. 4A), located in central Osaka and originally designed by the Japanese architect Takeo Yasui (1884–1955). The construction began in April 1930 and was completed in March 1933. The building was later extended to the northern side in 1966 with a design by Shoichi Sano (1921–2014)[30]. Flat gabbroic stone cladding panels were used on the southern exterior walls of the original structure (Fig. 4B). Documentary records[31,32] included an internal document from Yabashi Marble Co., Ltd., stating that Nakoso was used for the building. Three types of cladding panels were selected for analysis: (1) one showing a visible schlieren structure (Fig. 4B1, C), (2) one with a uniform massive structure (Fig. 4B2), and (3) one likely to have been repaired (Fig. 4B3). All panels were analyzed under the same conditions as the reference samples. Each was measured twenty times at randomly selected positions.

Results and discussion

Reference samples

Table 1 summarizes the analytical results for the gabbroic stones used as reference stone plates (Ukigane, Nakoso, and Matsuba). A comparison of concentrations of Rb, Sr, Ca, and K between the average of 20 pXRF measurements and bulk compositions by WD-XRF measurements shows a good correlation (Fig. 5), although there were minor discrepancies. Ca concentrations were generally consistent between the two methods. K values were slightly lower in the pXRF data, and Rb and Sr also showed slightly higher concentrations compared to WD-XRF. The good agreement in Ca may reflect the fact that it is a major constituent of plagioclase, which dominates in gabbroic rocks, and is therefore less affected by matrix effects. In contrast, K is a major component of K-feldspar, which is only a minor phase in these rocks, and because K is a light element, its fluorescent X-rays are relatively weak and easily absorbed by air, possibly causing underestimation in pXRF measurements. The slight overestimation of Rb and Sr by pXRF is likely due to the detector's limited energy resolution and the lack of correction for hydrous minerals in the built-in quantification algorithms. Notably, although Rb is present only in trace amounts in gabbroic rocks, it was successfully detected by pXRF in all three stone types. This result demonstrates that Rb/Sr remains a meaningful parameter for distinguishing gabbroic stone materials. Since they are present in only trace amounts, their X-ray fluorescence signals are relatively weak, and slight spectral interferences or inaccuracies in matrix correction can have a relatively large effect on the final value. In addition, the calibration and spectral fitting algorithms built into the instruments may not be optimized for the matrix of the mafic rocks, which could lead to systematic deviations. Indeed, similar trends have been observed in sediment measurements when comparing pXRF and WD-XRF data [33].

To account for these differences, the pXRF values for Rb, Sr, K, and Ca were adjusted using correlation factors derived from least-squares regression lines, assuming that the WD-XRF values accurately represent the bulk compositions of the reference samples. Although a linear regression based

Table 1 | Average of twenty pXRF measurements at different positions and the bulk compositions by WD-XRD of the reference samples of the gabbroic stone materials (Ukigane, Nakoso, Mtasuba)

	Average and standard deviation (SD) of 20 pXRF measurements						Bulk composition by WD-XRD		
(wt%)	Ukigane	*(SD)*	Nakoso	*(SD)*	Matsuba	*(SD)*	Ukigane	Nakoso	Matsuba
SiO_2	41.82	*0.91*	48.18	*1.17*	55.04	*2.38*	43.36	49.04	54.30
TiO_2	1.82	*0.47*	1.82	*0.44*	0.70	*0.43*	1.74	1.40	0.71
Al_2O_3	22.07	*2.15*	19.96	*1.39*	19.72	*1.20*	20.16	18.70	20.72
FeO[a]	9.39	*0.90*	8.77	*1.26*	5.66	*1.55*	11.86	10.51	7.72
MnO	0.13	*0.03*	0.19	*0.04*	0.10	*0.03*	0.17	0.24	0.14
MgO	4.56	*1.57*	3.44	*0.84*	0.78	*0.97*	6.55	5.82	2.84
CaO	12.73	*1.04*	9.16	*0.59*	9.67	*0.77*	12.83	9.62	10.70
Na_2O	*not available*		*not available*		*not available*		1.20	2.12	2.39
K_2O	0.15	*0.07*	0.28	*0.07*	0.61	*0.14*	0.22	0.41	0.71
P_2O_5	0.04	*0.01*	0.02	*0.01*	0.03	*0.02*	0.02	0.19	0.09
total	92.71		91.83		92.30		98.12	98.03	100.31
(ppm)									
Ba	100	*30*	169	*27*	266	*30*	74	120	159
Cr	115	*33*	85	*35*	198	*76*	52	25	31
Cu	35	*15*	39	*23*	78	*10*	60	10	80
Ni	12.3	*11*	3.2	*8*	3.4	*7*	8.5	6.8	6.6
Rb	6.0	*3*	10.5	*2*	18.9	*4*	5.5	7.2	14.2
Sr	503	*59*	577	*47*	408	*22*	336	425	344
V	597	*172*	260	*83*	332	*233*	543	253	263
Y	13	*5*	11	*6*	12	*2*	13	11	15
Zn	72	*20*	72	*16*	49	*10*	79	106	72
Zr	13	*4*	32	*21*	26	*13*	28	39	52

Italicized values represent standard deviations (SD).

[a]Total Fe as FeO.

on only three data points is not statistically robust, it was employed to examine the observed discrepancies. The Rb/Sr–K_2O/CaO diagrams, which plot both the unadjusted and adjusted values for each stone, are presented in Fig. 6. Both diagrams demonstrate that the plotted areas for each stone do not overlap, allowing for a clear distinction based on their distributions. When elemental ratios are used in this manner, adjustment using bulk compositions by WD-XRF may not be necessary for practical purposes because the differences are unlikely to be significant. Therefore, in the identification of gabbroic stone materials, the use of compositional or elemental ratios obtained from pXRF analysis is practically sufficient, even without adjustment, provided that the same analytical equipment is used consistently.

In situ analysis

Figure 7 shows the results of the in-situ analysis at the Osaka Gus Building. Only the plots of the repaired parts overlapped with the reference area of Ukigane, while those of the other parts of the exterior wall overlapped with the reference area of Nakoso. These results demonstrate that pXRF analysis is applicable for identifying building stones, at least in the case of gabbroic rocks. Simultaneously, they indicate that the stone plates cut from the inventory of the stone company can serve effectively as reference samples for pXRF-based provenance analysis. Given that the lithology of Ukigane varies even within the exposed quarry and the precise source of Nakoso is unknown, the stone blocks preserved by the stone company and used as reference samples are considered to be representative of the materials that were commercially available at the time.

The clear identification achieved in this study is likely due to the flat and polished surfaces of the building stones, which enabled measurements under nearly identical conditions to the reference samples. This is particularly valuable in cases where quarry locations are uncertain. However, for such unadjusted identification to be successful, it is essential that the in-situ measurements be conducted using the same pXRF device as that used for analyzing the reference samples. This is because pXRF results strongly depend on the calibration strategy adopted by the manufacturer[34]. These findings underscore the importance of preserving legacy stone specimens in museums or public institutions. These legacy stone specimens preserved by stone companies served effectively as proxies for original quarry products. Such efforts are expected to contribute to the development of reliable geochemical databases for stone identification and, in turn, facilitate the advancement of non-destructive identification techniques for archaeological artifacts and historic buildings.

The fact that Ukigane was used exclusively in the repaired sections suggests that it was likely employed as a substitute for Nakoso during restoration work. This demonstrates the potential of pXRF analysis to support the identification of undocumented building stones and to provide valuable insights into the sourcing and distribution of materials, thereby contributing to the reconstruction of architectural history.

Petrological interpretation

Although Rb is generally present only in trace amounts in mafic rocks such as gabbro, its successful detection and the functionality of the Rb/Sr ratio as a discriminating indicator in this study suggest that this ratio is broadly applicable for stone identification beyond felsic compositions. Due to similar ionic radii, Rb can readily substitute for K in the K-feldspar crystal structure, whereas Sr can similarly substitute for Ca in plagioclase. As a result, rocks with more K-feldspar show higher Rb/Sr ratios[35]. Therefore, an

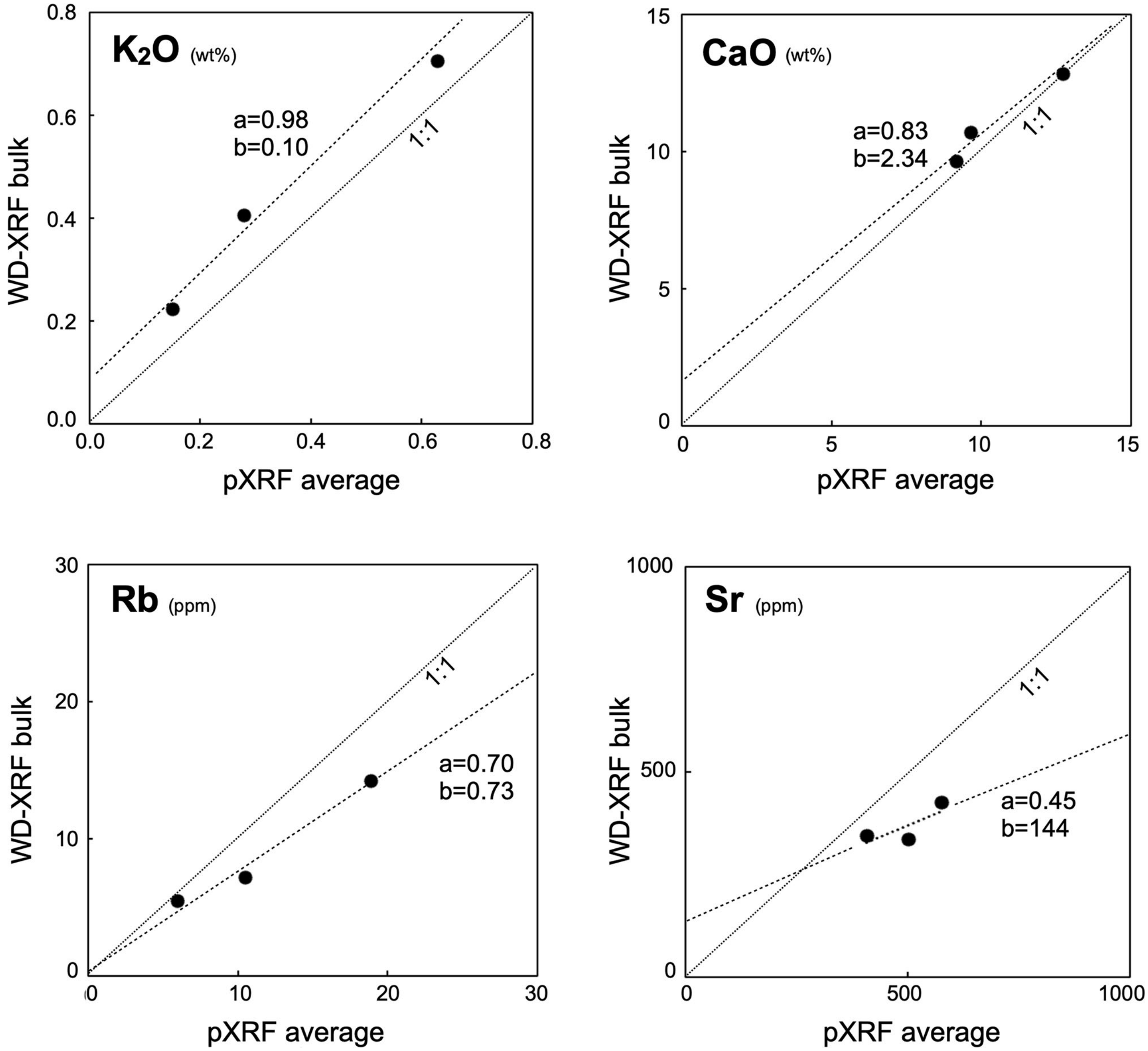


Fig. 5 | Comparison of K_2O, CaO, Rb, and Sr concentrations between the average of twenty pXRF measurements and the WD-XRF bulk measurements on the reference samples. Dotted lines represent least-squares regression lines. The slope (a) and intercept (b) of each regression line are indicated in the figure.

Rb/Sr–K_2O/CaO diagram illustrates the crystallization differentiation trends and likely reflects K-feldspar content in the gabbroic (mafic) rocks. Since elements, such as Rb and K, are more concentrated in minor K-feldspar than in the major minerals of gabbro, their detection with pXRF analysis suggests that minor minerals may be more useful for identification than coarse-grained major minerals, whose influence depends on the areal proportion of the mineral within the X-ray irradiated area. This is consistent with the effectiveness of Rb/Sr and Ti/Fe as geochemical indicators in the identification of felsic sandstones used for architectural materials[14]. Ti/Fe likely reflects the abundance of minor mafic minerals such as biotite, magnetite, and ilmenite in felsic rocks. In archaeology, non-destructive identification of stone tools using pXRF is widely practiced, especially for felsic materials such as obsidian, and Fe, Ti, and Mn are often used as indicators, along with Rb and Sr[36]. These examples further suggest that the Rb/Sr ratio is broadly applicable for identifying stone materials, whereas the K_2O/CaO ratio may be particularly suited to mafic rocks.

Although Nakoso and Ukigane may be distinguishable when observed side by side, it is difficult to visually identify them when separated in architectural settings. Therefore, the ability to distinguish these stones through scientific analyses is highly significant. However, because “black granites” have been imported overseas since the end of World War II, it is necessary to compile comparative data to differentiate them from domestic stones.

Such petrological interpretation not only aids in the scientific identification of gabbroic stone materials but also offers insights into their geological provenance and regional diversity. The variations in geochemical signatures among the stones reflect differences in their magmatic histories and, by extension, the geological characteristics of their source areas. This underscores the value of petrological studies in highlighting the regional identity of building stones. These results suggest that geochemical investigations for stone identification can also provide petrological insights into the regional variability of gabbroic rocks. This further highlights the importance of preserving historically distributed stone specimens, not only for accurate provenance analysis, as previously discussed, but also for documenting the geological diversity of culturally significant stone materials.

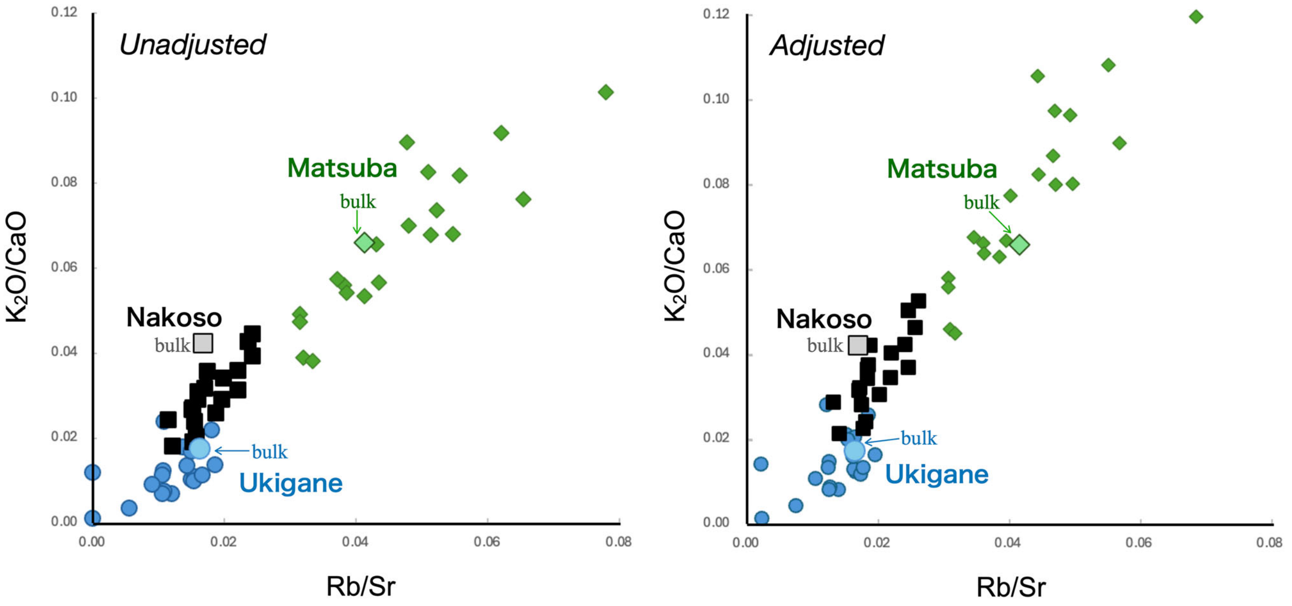


Fig. 6 | Rb/Sr–K_2O/CaO diagrams showing distinct geochemical fields for the three gabbroic stones used in early 20th-century Japanese architecture. The plots of the three gabbroic stones, Ukigane, Nakoso, and Matsuba, are clearly separated based on pXRF values plotted before (left) and after (right) adjustment using WD-XRF bulk compositions, demonstrating distinct geochemical fields for each. The result indicates that the adjustment has minimal effect on stone discrimination.

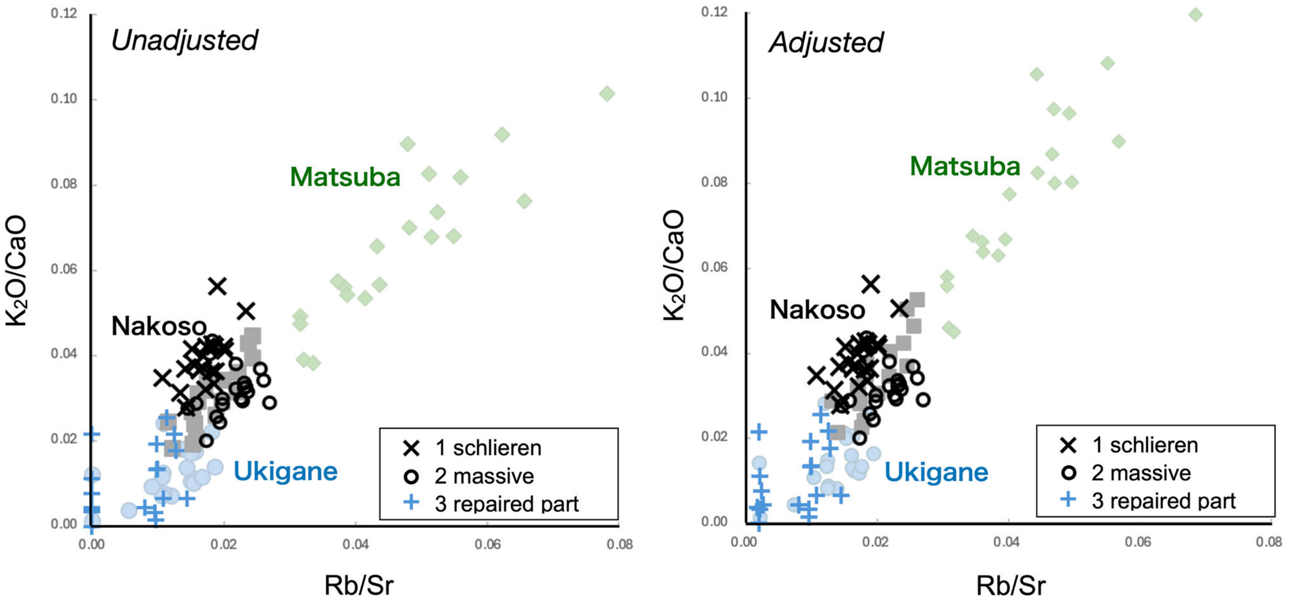


Fig. 7 | Rb/Sr–K_2O/CaO diagrams for the gabbroic stones used as exterior wall cladding of the Osaka Gas Building. The shaded reference fields (identical to those in Fig. 6) are shown in the background, with in-situ pXRF data plotted as unadjusted (left) and adjusted (right) based on bulk compositions determined by WD-XRF. Sampling points for the in-situ pXRF measurements are indicated as: (1) area with visible schlieren structure, (2) uniform massive area, and (3) repaired part.

Data availability

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Author contributions

S.N. designed the study, analyzed data, wrote the manuscript, organized the references, and reviewed the literature. M.I. analyzed reference samples and reviewed the literature. S.N., M.I., T.N., and A.H. performed the in situ analysis. H.Y. performed the bulk analysis. All authors have discussed the results, read and agreed to the published version of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

Correspondence and requests for materials should be addressed to Shoji Nishimoto.