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Historic commodity of sulfur prevailed during the early to middle 19th century in Japan: A stable isotopic analysis for tracing the provenance

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ABSTRACT

Demands for native volcanic sulfur as one of the ingredients of gunpowder reached a maximum during the early to middle 19th century, when abrupt changes in the political regime occurred in Japan. The historic commodity of sulfur prevailing during this time was analyzed for stable isotopic ratios (expressed as $\delta^{34}S$) to examine the provenance. The sampled sulfur involves the Siebold collection (acquisition by P. F. von Siebold in Nagasaki, northern Kyushu and kept in Naturalis, Leiden, The Netherlands) and Egawa library (Nirayama, central Japan) for which exact location of the origin is ambiguous or absent. The sulfur isotopic data were evaluated by comparison with the revised database for the spatial distribution of $\delta^{34}S$ values of operative sulfur mines throughout the Japanese archipelagos. With a few exceptions, the commodity sulfur was transported through short-distance marketing systems within close proximity to Nagasaki and Nirayama.

Contents

1. Introduction

Volcanism provides useful natural resources for common human life. Among the known volatiles, native volcanic sulfur, i.e., elemental sulfur, was a major commodity for international trading in eastern Asia during historic times (Mizota and Yamanaka, 2014). In the Japanese archipelago, wide-spread volcanism provided operative deposits of native sulfur which served as one of the ingredients of gunpowder (mechanical mixture consisting of saltpetre (KNO_3), native sulfur and charcoal carbon in an approximate proportion of 70:15:15). Demands for the native sulfur reached a maximum during the early to middle 19th century when political regimes changed

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from a feudal system characteristic of the Shogunate to the Meiji restoration. Among the gunpowder ingredients at the time, world-wide marketing route of saltpetre had studied (Mizota et al., 2015). Trading routes of importation and transportation of domestic productions were proven by distinctive $\delta^{15}N$ values of nitrates. Nevertheless, economically operative sulfur resources are localized within the Japanese archipelago, being concentrated in northern and southern islands. Normally, sulfur resources are not located in urban regions where the demand for sulfur is elevated. Trading routes from source to destination for fulfilling the demand may exist during the early to middle 19th century, while very limited socio-historical studies have been conducted (Itagaki, 2002; Nakanishi, 2006) based on the historic documents. Alternative way toward the use of historic objectives is required.

$^{34}S/^{32}S$ ratios, commonly designated as $\delta^{34}S$ values of native sulfur (as shown by per mil deviation relative to meteorite Cañon Diablo troilite, abbreviated as V-CDT), provide direct evidence for the origin (Ueda et al., 1979a,b; Mizota and Yamanaka, 2014). By comparing the values in the database (Mizota and Yamanaka, 2014) to the unknown samples, plausible sulfur mines can be identified. The commodity sulfur specimens that were manufactured in the 1970's and marketed as Sicilian are shown by the isotopic analysis to consist of non-Sicilian salt-dome sulfur crystallized over a Sicilian matrix (Peterson et al., 2003). Nevertheless, validity of such a stable isotopic approach has not been tested for the historic native commodity sulfur prevailed during the times. In the present study, we extended a previous study (Mizota and Yamanaka, 2014), focusing on the samples from early to middle 19th century, i.e., Siebold collection from Naturalis (Natural History Museum, located at Leiden, The Netherlands) and Egawa library (located at Nirayama, central Japan). These two institutions have kept suitable samples for which secondary deterioration of the intrinsic isotopic signatures is limited.

2. Materials and methods

2.1. Description of samples

Commodity sulfur of native volcanic origin which prevailed during the early to middle 19th century in Japan was collected from two institutions. The approximate production age of the sulfur was estimated by relevant historic documents (Siebold, 1897; Nakada, 1998).

2.1.1. Siebold collection

Eight hundred twenty-eight specimens of the Japanese rocks and minerals collected during the early of the 19th century are stored in the National Museum of Natural History (Naturalis), Leiden, The Netherlands (Tagai, 2003; Tagai and Mikouchi, 2008). The photo images and brief lithological description are available online from The University Museum, The University of Tokyo (URL; http://umdb.um.u-tokyo.ac.jp/DKoubutu/FSiebold/recordlist.php). German physician, Philip Franz von Siebold collected these historic specimens as a Dutch surgeon of Dutch East India Army. He arrived at Dejima Trading Post (Nagasaki, south-western Japan) on June 1823 and left there on December 1829. Being put under the house arrest in those seven years stay, he made enormous rock and mineral collections and received ample financial support by Dutch government through commercial and/or private bases (Kurihara, 1997a,b; 1998). Most activity of Siebold as a scientist during his stay at Dejima has been documented in Japanese local journals or monographs with limited distribution. The geological specimens involve twelve native volcanic sulfur (Table 1) which are highly unique in terms of the commodity sulfur prevailed during the time. Except for two sulfur samples (Registered Nos. 329203 and 329211) which represent yellowish sublimates on the hydrothermally altered rocks from Unzen volcano (Nagasaki, Kyushu), almost all samples are sulfur blocks or powder (No. 411000). Due to its brittle nature (Mohs' scale of hardness: 2) together with long-distance transportation involving domestic (source sulfur mines to Dejima, Nagasaki) and international maritime routes (Japan to The Netherlands), distinctive edges of the sulfur block were lost, resulting in the modified smooth surfaces. Some of the sulfur specimen are associated with labels written in Germany and/or classic Japanese which are insufficient for the identification of the relevant sulfur mines (Tagai, 2003; Ohba and Tagai, 2010), and others were not labeled or labels were lost during stock. Tagai and Mikouchi (2008) interpreted the plausible province of the sulfur specimens based on the labels. Nevertheless, direct or firm evidence to verify the validity for the origin of the specimen sulfur awaits further analysis.

2.1.2. Egawa library

Native sulfur (Fig. 1A) and firing bomb (Fig. 1B) from Egawa library (Nirayama, Izunokuni City, Shizuoka Prefecture) (Fig. 2) which prevailed during the comparable times with Siebold collection were examined.

Miocene to Pleistocene terrestrial volcanism gave rise to deposits native sulfur in Izu Peninsula where the territory of Hidetatsu Egawa

Table 1
Description and stable isotopic analysis for volcanic native sulfur from the Siebold collection.

RGN[a]	Locality[b,c]	Numbers of replicated analysis	Sulfur content (%)	$\delta^{34}S$ (‰) average (SD)	Source mines as evidenced by the present isotopic analysis	Range of plausible trading
Given location						
411002	Northeastern Japan	7	93 to 103	−0.8 (0.1)	No plausible mine	Inconclusive
329204	Aso-Jigoku[d]: Kumamoto	5	95 to100	+1.7 (1.5)	Aso-Jigoku	Identical with label
422001	Saga and Nagasaki	4	96 to 99	+7.9 (0.9)	Iwo-Torishima	Long-distance
329203	Unzen: Nagasaki	2	97 and 99	+3.3	Unzen	Identical with label
329211	Unzen: Nagasaki	1	108	−7.3	No plausible mine	Inconclusive
329206	Kirishima-1: Miyazaki	3	100 to 109	+2.1 (0.7)	Satsuma-Iwojima	Within Kyushu
329207	Kirishima-2: Miyazaki	7	97 to 115	−0.3 (0.6)	Kuju-Iwoyama	Within Kyushu
329205	Sakurajima: Kagoshima	2	103 and 107	+6.4	Iwo-Torishima	Long-distance
Commodity or unknown location						
329208	*Ara-iou*[e]	8	73 to 99	0.0 (1.3)	Kuju-Iwoyama	Within Kyushu
329210	*Takanome*[f]	3	98 to 110	0.0 (0.7)	Kuju-Iwoyama	Within Kyushu
329209	Without label	6	101 to 108	+0.4 (0.6)	Tsukahara-Nabeyama	Within Kyushu
411000	Sulfur powder	1	16	−0.6	No plausible mine	Inconclusive

[a] Registered number of Naturalis collection.
[b] Referred from database of the University of Tokyo.
[c] Followed by the attached labels in the specimen box.
[d] Followed by the description given by P. F. von Siebold (1897).
[e] Means Japanese “crude commodity”.
[f] Means Japanese “highly refined sulfur commodity”.