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To cite this version:

Akane Yamakawa, Kimiyo Nagano, Miyuki Ukachi, Kaoru Onishi, Katsuyuki Yamashita, et al.. Sr Isotopic Composition of NIES Certified Reference Material No. 28 Urban Aerosols. Frontiers in Environmental Chemistry, Frontiers Media, 2021, 2, pp.771759. 10.3389/fenvc.2021.771759. hal-03450508

HAL Id: hal-03450508
https://hal-univ-pau.archives-ouvertes.fr/hal-03450508
Submitted on 26 Nov 2021

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Sr Isotopic Composition of NIES Certified Reference Material No. 28 Urban Aerosols

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An interlaboratory study of the National Institute for Environmental Studies (NIES) certified reference material (CRM) No. 28 Urban Aerosols collected from the filters of a central ventilating system in a building in the Beijing city center from 1996 to 2005 was performed to obtain an information value of the Sr isotopic composition. The Sr isotopic composition was measured using multi-collector-inductively coupled plasma-mass spectrometry (MC-ICP-MS) to confirm the CRM’s within- and between-bottle homogeneity, and the results showed a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.710227 ± 0.000019 (2SD, n = 18). The Sr isotopic compositions were intercompared using thermal ionization mass spectrometry (TIMS), which showed good agreement with values obtained at NIES. Subsequently, a consistent $^{87}\text{Sr}/^{86}\text{Sr}$ ratio was observed between two dissolution (hotplate vs. high-pressure bomb) and Sr separation (Sr spec resin vs. cation exchange resin) methods. To validate and reproduce the accuracy of our analytical methods, the Sr isotopic compositions of secondary reference materials, JB-1b and JA-2, were also measured. Our results showed that NIES CRM No. 28 is appropriate for the quality control of Sr isotope measurements of particulate matter analyses for environmental and geochemical studies.

Keywords: Sr isotopes, atmospheric particles, MC-ICP-MS, TIMS, NIES CRM

INTRODUCTION

Atmospheric particulate matter (PM) is a complex mixture of particles with diverse chemical compositions and sizes. The chemical compositions vary depending on their source (natural vs. anthropogenic), environmental condition (e.g., temperature, humidity, and redox condition), and atmospheric processing (e.g., radiation, convection, and transport). PM emissions from urban and industrial areas are a critical environmental problem that affects the climate, human health, visibility, biogeochemical cycles, and atmospheric chemistry. Identifying the source(s) of emitted PM is critical for providing scientific strategies to improve air quality.

Recent studies have assessed the utility of strontium (Sr) isotopes $^{87}\text{Sr}/^{86}\text{Sr}$ to identify sources of atmospheric PM (e.g., Capo et al., 1998; Kanayama et al., 2002; Grousset and Biscaye, 2005; Lahl Geagea et al., 2008; Widory et al., 2010; Duarte et al., 2017). Sr has four natural isotopes: $^{84}\text{Sr}$, $^{86}\text{Sr}$,
$^{87}$Sr and $^{88}$Sr. $^{87}$Sr is a radiogenic isotope of $^{87}$Rb by $\beta$-decay (half-life = $4.88 \times 10^{10}$ years; De Laeter et al., 2003). Due to an initial difference in Rb/Sr and age-integrated effects, the $^{87}$Sr/$^{86}$Sr ratios of Earth surface materials vary widely (e.g., more radiogenic ancient crustal rock or less radiogenic carbonate as the parent materials of soil). Therefore, Sr isotope ratios measured at a receptor site provide clues to the source of the Sr or the mixing ratio of multiple sources. For example, a previous study on atmospheric PM in Beijing indicated that atmospheric Sr was mainly controlled by coal combustion and to a lesser extent by cement plants and/or smelters (Widory et al., 2010).

To characterize emission sources, accurate methods for determining Sr isotopic ratios in PM are required, and matrix matching between samples and standards is important for the quality control of the analysis. Currently, the NIES and other research institutes (e.g., National Institute of Standards and Technology and the European Commission Joint Research Center–Institute for Reference Materials and Measurements) provide commercially available standard aerosol reference materials; however, Sr isotopic compositions of PM reference materials have not been reported. To overcome this limitation, we aim to obtain an information value of the $^{87}$Sr/$^{86}$Sr ratio for NIES CRM No. 28 Urban Aerosols. The objectives of this study are: (i) to determine the Sr isotope distribution within and between the bottles of the CRM using multi-collector-inductively coupled plasma-mass spectrometry (MC-ICP-MS); (ii) to confirm the consistency of the interlaboratory CRM isotopic ratio using two types of instruments, MC-ICP-MS vs. thermal ionization mass spectrometry (TIMS); and (iii) to confirm the consistency of the Sr isotopic composition using different digestion methods (hotplate vs. high-pressure bomb) and Sr separation methods (Sr spec resin vs. cation exchange resin).

**MATERIALS AND METHODS**

**Reagents**

All acids used in this study were Ultrapure-100 (Kanto Chemical Co., Inc., Japan), and aqueous solutions were prepared using Milli-Q water (Japan Millipore Ltd., Japan) at NIES. At Okayama University, EL grade 70% HNO$_3$ (Kanto Chemical Co., Inc., Japan) was twice distilled without dilution using a Teflon still. EL grade 36% HCl (Kanto Chemical Co., Inc., Japan) was diluted to ∼6 M using Milli-Q water and then twice distilled using a Teflon still. TAMAPURE-AA-100 HClO$_4$ (Tama Chemicals Co., Ltd., Japan) was used without further purification.

**Samples**

**NIES CRM No. 28**

NIES CRM No. 28 Urban Aerosols, collected from the filters of a central ventilating system of a building located in Beijing city center, was produced to evaluate the analytical accuracy of determining the mass fraction of selected elements (18 certified and 14 reference values) (Mori et al., 2008). The certified value of Sr is 469 ± 16 mg/kg (certificate is available on https://www.nies.go.jp/labo/crm-e/hrfba300000ble6p-att/No. 28_E.pdf). The Hg isotopic composition was also determined as an information value for the CRM (Yamakawa et al., 2020).

**JA-2 and JB-1b**

Sr isotopic measurements of the secondary reference, JA-2 and JB-1b, were performed using the same methods to manage the analytical accuracy of our method. These geological samples were produced at the Geological Survey of Japan in the National Institute of Advanced Industrial Science and Technology. The $^{87}$Sr/$^{86}$Sr ratios of JA-2 and JB-1b are reported in Miyazaki and Shuto (1998) and Yuhara et al. (2000), respectively.

**NIST SRM 987**

NIST SRM 987 was prepared at each laboratory by the following methods. At NIES and Kumamoto University, strontium carbonate powder was dissolved in dilute HNO$_3$ to make a stock solution. The solution was then adjusted to 300 ng g$^{-1}$ in 2% HNO$_3$ for the isotope measurement. At Okayama University, a ∼1 µg ml$^{-1}$ stock solution of NIST SRM 987 was prepared by dissolving strontium carbonate powder in dilute HCl.

**Sample Preparation**

**Sample Decomposition**

Three bottles (bottle No. 044, 375, and 597) were randomly selected to assess the between- and within-bottle homogeneity of Sr isotope. The three subsamples were taken from each bottle and decomposed on a hotplate with a concentrated acid mixture of HNO$_3$/HClO$_4$/HF. Approximately 100 mg of powdered samples of the CRM, JA-2 and JB-1b were weighed and decomposed overnight in 5 ml of HNO$_3$ at 140°C. Then, 2 ml of 7 M HClO$_4$ was added and heated overnight at 200°C and dried to 1 ml. A mixture of 2 ml of 13 M HNO$_3$, 1 ml of 7 M HClO$_4$, and 1 ml 30 M HF was added and heated for 2 h. The decomposed samples were heated to dryness at 200°C by step heating. The resulting sample cake was redissolved in 1 ml of 3 M HNO$_3$, and the insoluble fraction was removed by centrifuging. This procedure was repeated several times, and the supernatant was subjected to the following chemical separation.

At Okayama University, the Sr isotope ratios were investigated using different acid decomposition and Sr separation methods. To obtain a representative sample, approximately 200 mg of powdered sample (bottle No. 35) was digested. The sample was dissolved in an HF/HNO$_3$ mixture using Teflon capsules sealed in stainless steel bombs for 72 h at 190°C. Once complete digestion was achieved, the sample was transferred to a 15 ml Teflon vial and 0.2 ml of HClO$_4$ was added before drying down at 120°C–200°C. The evaporated sample was further treated in 0.2 ml of HClO$_4$ to avoid fluoride precipitation. The sample was subsequently dissolved in HCl and ∼25% split was taken for Sr isotopic analysis.

**Chemical Separation**

Sr was separated from the other elements, particularly Rb, in the digest using Sr spec resin (Eichrom Technologies, US) at NIES. One milliliter of Sr spec resin was packed in a size S polypropylene column (Muromachi Chemical Inc., Japan). The resin was cleaned by passing 3 ml of 3 M HCl, 18 ml of 0.05 M...
The MC-ICP-MS was used in this study was a Nu Plasma II (Nu Instruments, UK) at NIES. Although Sr was isolated from the matrix components and Rb by column chemistry using Sr spec resin, the signal for $^{85}$Rb was simultaneously measured for the isobaric correction. As krypton (Kr) is present as a contaminant in the Ar plasma gas, the contribution of Kr had to be corrected. During sample decomposition and chemical separation, the recovery yield was $>95\%$. The total procedural blank for Sr was $<1\,\text{ng}$, which was negligible compared to the sample size used in this study. For the isotopic measurement, the final solutions were diluted to a Sr concentration of 300 $\mu\text{g}\,\text{g}^{-1}$ using 2 M HNO$_3$.

At Okayama University, Sr was separated by passing it through cation exchange resin in 2 M HCl (1 ml of AG50 × 12, 200–400 mesh, packed in a size S polypropylene column (Muromachi Chemical Inc.)). To achieve complete separation of Rb, the Sr separation was repeated twice. The total procedural blank was $\sim30\,\text{pg}$, which was insignificant relative to the amount of Sr extracted.

**Sr Isotope Ratio Determinations: Reproducibility and Accuracy**

**MC-ICP-MS**

The reproducibility of the Sr isotopic compositions of the NIST SRM 987 was monitored during the study period to validate the analytical stability of our operating conditions. The $^{87}$Sr/$^{86}$Sr ratios of the samples were adjusted using the recommended NIST SRM 987 value of 0.710248 (McArthur et al., 2001). The Sr isotopic composition of JA-2 was also measured at Okayama University, showing the $^{87}$Sr/$^{86}$Sr ratios of 0.706311 $\pm$ 0.000024 (25D, n = 9) (Table 2). These values are identical within an acceptable error to their literature counterparts (JA-2: 0.706331 $\pm$ 0.000013 (25D, n = 5) in Miyazaki and Shuto, 1998; JB-1b: 0.704095 $\pm$ 0.000012 (25D, n = 13) in Yuhara et al., 2000). The $^{87}$Sr/$^{86}$Sr isotopic values of JA-2 and JB-1b, same aliquots prepared at NIES, showed 0.706311 $\pm$ 0.000007 and 0.706325 $\pm$ 0.000007 for JA-2, and 0.704083 $\pm$ 0.000007 and 0.704084 $\pm$ 0.000006 for JB-1b at Kumamoto University (the analytical error is described in 2SE, and 150 ratios were taken in a single measurement) (Table 2). The Sr isotopic composition of JA-2 was also measured at Okayama University, showing the $^{87}$Sr/$^{86}$Sr ratios of 0.706295 $\pm$ 0.000024 (25D, n = 9) (Table 2). These results indicate that our technique was robust enough to measure $^{87}$Sr/$^{86}$Sr ratios.

**Table 1** Details of the operational parameters of the MC-ICP-MS (Nu Plasma II; Nu Instruments, UK) at NIES.

| Instrumentation | Nu Plasma II |
|-----------------|--------------|
| Monitored isotopes | $^{88}$Sr, $^{87}$Sr, $^{87}$Rb, $^{86}$Sr, $^{85}$Kr, $^{85}$Rb, $^{84}$Sr, $^{83}$Kr, $^{82}$Kr |
| RF power | 1300 W |
| Plasma gas | 13.0 L min$^{-1}$ |
| Auxiliary | 0.8 L min$^{-1}$ |
| Nebulization | 1.0 L min$^{-1}$ |
| Integration time | 8 sec |
| Number of cycles per block | 20 cycle/block |
| Number of blocks | 4 blocks |
| Sr concentrations of sample and standard | 300 ng g$^{-1}$ |
| Sensitivity of sample and standard ($^{87}$Sr) | $-10 \times 10^{-11}$ A |

HNO$_3$, and 3 ml of 3 M HNO$_3$. The sample dissolved in 1 ml of 3 M HNO$_3$ was loaded onto the column after conditioning the resin using 1 ml of 3 M HNO$_3$. The fraction containing Rb (and Ca, K, Mg, Ba, etc.), which began eluting immediately, was discarded by passing an additional 3 ml of 3 M HNO$_3$, 4 ml of 6 M HNO$_3$, and 1 ml of 3 M HNO$_3$. The Sr fraction was obtained by passing 5 ml of 0.05 M HNO$_3$ through a filter. During sample decomposition and chemical separation, the recovery yield was $>95\%$. The total procedural blank for Sr was $<1\,\text{ng}$, which was negligible compared to the sample size used in this study. For the isotopic measurement, the final solutions were diluted to a Sr concentration of 300 $\mu\text{g}\,\text{g}^{-1}$ using 2% HNO$_3$.

At Okayama University, Sr was separated by passing it through cation exchange resin in 2 M HCl (1 ml of AG50 × 12, 200–400 mesh, packed in a size S polypropylene column (Muromachi Chemical Inc.)). To achieve complete separation of Rb, the Sr separation was repeated twice. The total procedural blank was $\sim30\,\text{pg}$, which was insignificant relative to the amount of Sr extracted.

**RESULTS AND DISCUSSION**

**Sr Isotope Measurement Reproducibility and Accuracy**

The reproducibility of the Sr isotopic compositions of the NIST SRM 987 was monitored during the study period to validate the analytical stability of our operating conditions. The $^{87}$Sr/$^{86}$Sr ratios of the samples were adjusted using the recommended NIST SRM 987 value of 0.710248 (McArthur et al., 2001). The Sr isotopic composition of JA-2 was also measured at Okayama University, showing the $^{87}$Sr/$^{86}$Sr ratios of 0.706311 $\pm$ 0.000024 (25D, n = 9) (Table 2). These values are identical within an acceptable error to their literature counterparts (JA-2: 0.706331 $\pm$ 0.000013 (25D, n = 5) in Miyazaki and Shuto, 1998; JB-1b: 0.704095 $\pm$ 0.000012 (25D, n = 13) in Yuhara et al., 2000). The $^{87}$Sr/$^{86}$Sr isotopic values of JA-2 and JB-1b, same aliquots prepared at NIES, showed 0.706311 $\pm$ 0.000007 and 0.706325 $\pm$ 0.000007 for JA-2, and 0.704083 $\pm$ 0.000007 and 0.704084 $\pm$ 0.000006 for JB-1b at Kumamoto University (the analytical error is described in 2SE, and 150 ratios were taken in a single measurement) (Table 2). The Sr isotopic composition of JA-2 was also measured at Okayama University, showing the $^{87}$Sr/$^{86}$Sr ratios of 0.706295 $\pm$ 0.000024 (25D, n = 9) (Table 2). These results indicate that our technique was robust enough to measure $^{87}$Sr/$^{86}$Sr ratios.
TABLE 2 | Sr isotopic compositions of JA-2 and JB-1b.

| Sample | Instrumentation and Reference | n | Mean 87Sr/86Sr | 2SD
|--------|-------------------------------|---|----------------|---|
| JA-2   | MC-ICP-MS (NIES)              | 7 | 0.706315       | 0.000022 |
|        | TIMS (Kumamoto Univ.)         | 1 | 0.706311       | 0.000007  |
|        | TIMS (Kumamoto Univ.)         | 1 | 0.706325       | 0.000007  |
|        | TIMS (Okayama Univ.)          | 9 | 0.706295       | 0.000024  |
|        | Miyazaki and Shuto (1998)*    | 5 | 0.706331       | 0.000013  |
| JB-1b  | MC-ICP-MS (NIES)              | 8 | 0.704093       | 0.000023  |
|        | TIMS (Kumamoto Univ.)         | 1 | 0.704083       | 0.000007  |
|        | TIMS (Kumamoto Univ.)         | 1 | 0.704084       | 0.000006  |
|        | Yuhara et al. (2009)*         | 13| 0.704095       | 0.000012  |

*The analytical error is described in 2SE. 150 ratios (15 cycles x 10 blocks) were taken in a single measurement.

**Reported 87Sr/86Sr ratios of the NIST SRM 987 are 0.710251 ± 0.000004 (2σm,n = 51) in Miyazaki and Shuto (1998), and 0.710251 in Yuhara et al. (2000). The 87Sr/86Sr ratios of the samples are corrected for interlaboratory bias by adjusting the mean value of the NIST SRM 987 standard run with the samples to the value of 0.710248.

TABLE 3 | Sr isotopic ratios of NIES CRM No. 28 Urban Aerosols.

| Instrumentation | Pretreatment | Decomposition | Sr separation | Bottle No. | Number of subsampling | Number of measurements for each subsample | Mean 87Sr/86Sr | 2SD |
|-----------------|--------------|---------------|---------------|------------|-----------------------|------------------------------------------|----------------|-----|
| MC-ICP-MS (NIES) | Nu Plasma II | Hotplate with HNO3/ HClO4/HF mixture | Sr spec resin | 044        | 3                     | 2                                        | 0.710221       | 0.000014 |
|                 |              |               |               | 375        | 3                     | 2                                        | 0.710233       | 0.000024 |
|                 |              |               |               | 597        | 3                     | 2                                        | 0.710228       | 0.000015 |
|                 | TIMS (Kumamoto Univ.) | Hotplate with HNO3/ HClO4/HF mixture | Sr spec resin | 044*       | 4                     |                                           | 0.710229       | 0.000013 |
|                 |              |               |               | 375*       | 4                     |                                           | 0.710226       | 0.000012 |
|                 |              |               |               | 597*       | 4                     |                                           | 0.710223       | 0.000009 |
| TIMS (Okayama Univ.) | Finnigan | High-pressure bomb with HNO3/ HF/HClO4 | Cation exchange resin, AG50 × 12, 200–400 mesh | 035        | 12                    |                                           | 0.710226       | 0.000019 |

*Sample aliquots, decomposed and pretreated at NIES, were analyzed at Kumamoto University. Average internal precisions were ± 0.000013, ± 0.000007, and ± 0.000009 (2SE) at NIES, Kumamoto University, and Okayama University, respectively.

Homogeneity of Sr Isotopic Compositions for NIES CRM No. 28

Table 3 shows the Sr isotopic ratio of NIES CRM No. 28. Within- and between-bottle were evaluated using MC-ICP-MS at NIES, yielding 87Sr/86Sr isotopic ratios of 0.710227 ± 0.000019 (2SD, n = 18). The uncertainty of the Sr isotopic values is an expanded uncertainty determined using a coverage factor k = 2, which corresponded to a confidence interval of ~95%. To investigate the homogeneity of the isotopic results in CRM (Table 4), the 87Sr/86Sr isotopic ratios were tested using a one-way analysis of variance (ANOVA). The between-bottle variation was not statistically significant (p > 0.05 and Fcalculated value < Fcritical value) as evaluated by one-way ANOVA. Therefore, when applied to the Sr isotopic ratios presented in this study, the CRM was homogeneous.

Interlaboratory Studies

For the MC-ICP-MS vs TIMS comparison, the Sr isotopic ratios of the three CRM samples (bottle No. 044, 375, and 597), which had been digested and processed for Sr separation at NIES, were measured by TIMS at the Kumamoto University to confirm the consistency of the 87Sr/86Sr isotopic ratios due to different analytical instruments: NIES was 0.710227 ± 0.000019 (2SD, n = 18) and Kumamoto University was 0.710229 ± 0.000011 (2SD, n = 12) (Figure 1; Table 3). For sample decomposition and Sr separation, different methods were performed to evaluate bias during sample digestion and Sr separation at NIES and Okayama University. During the decomposition of the HNO3/HClO4/HF mixture using a hotplate at NIES, undissolved residues were observed, whereas a high-pressure bomb with HNO3/HF/HClO4 at Okayama University achieved complete digestion. The 87Sr/86Sr isotopic ratio obtained at Okayama University was 0.710226 ± 0.000019 (2SD, n = 12) (Figure 1; Table 3). Despite the incomplete dissolution at NIES, the 87Sr/86Sr
An isotopic ratio was consistent with the latter. Thus, the NIES CRM No. 28 Urban Aerosols was homogeneous enough for the Sr isotopic measurement of 100–200 mg subsamples using the methods described in Sample Preparation. This CRM will be of great value for the analytical quality assurance of environmental monitoring studies of PM.

**Potential CRM Emission Sources**

The present study investigates the use of $^{87}$Sr/$^{86}$Sr isotope systematics to help determine the origin of atmospheric aerosols. By characterizing the isotopes of ambient PM, potential sources of pollution near sampling sites can be identified, and their contribution to the contents of PM can be estimated. As a test, we compared the $^{87}$Sr/$^{86}$Sr ratio of NIES CRM No. 28 with those determined by a previous source determination study. Widory et al. (2010) analyzed 63 samples of ambient PM$_{2.5}$ and 23 samples of ambient total suspended particle (TSP) collected at various locations around Beijing from September 2005 to September 2006. The $^{87}$Sr/$^{86}$Sr ratios of PM$_{2.5}$ and TSP ranged from 0.7085 to 0.7108, with the TSP having a larger variation. They also reported the $^{87}$Sr/$^{86}$Sr ratios of potential emission sources near Beijing; coal combustion yielded aerosols with the lowest $^{87}$Sr/$^{86}$Sr (0.708970–0.709492), smelter-derived particles produced the greatest radiogenic value (0.712064), and cement factories created particles with intermediate $^{87}$Sr/$^{86}$Sr (0.709963–0.710528). They concluded that Sr in atmospheric PM in Beijing was mainly controlled by coal combustion and to a lesser extent by cement plants and/or smelters. Although sampling periods of the CRM, PM$_{2.5}$ and TSP overlapped only for 1 year, and the $^{87}$Sr/$^{86}$Sr ratio of CRM was a 10-years integral, the CRM $^{87}$Sr/$^{86}$Sr ratio plotted within the range of the reported ratios for PM$_{2.5}$ and TSP and near those of cement factories and coal combustion (Figure 2). The $^{87}$Sr/$^{86}$Sr ratio of potential end-members, soil (0.711784–0.714797) and smelters, were significantly lower than that of the CRM.

To gain more insight into the sources of the CRM, enrichment factors (EFs) were calculated related to the Earth’s upper continental crust (Taylor and McLennan, 1995) with Fe as the reference element (3.50%). The mass fractions of the CRM metallic elements were reported by Mori et al. (2008). EFs >10 of the selected element (regarding anthropogenic sources), were higher in the order of Sb (120.5), As (72.1), Cd (68.5), Pb (24.2), Mo (22.7), and Zn (19.3) (Table 5). Some previous studies have reported that the major source of Sb and Cu in urban atmospheres was brake abrasion particles (Hjortenkrans et al., 2007; Iijima et al., 2007). However, CRM Cu EFs were not as high as Sb (EF$_{Cu}$ = 5.0). High Sb EFs were also found in aerosols collected in Beijing from 2001 to 2006, and coal combustion was suggested as a possible extra source of Sb (Okuda et al., 2008). In addition, coal combustion has been considered a major source of As, the second-highest CRM EF in aerosols (Kowalczyk et al., 1978; Wang et al., 1999).

As a preliminary result, another fingerprint tracer, Pb isotopic composition, was obtained for the CRM at the Institut des Sciences Analytiques et de Physico-chimie pour l’Environnement et les Matériaux using MC-ICP-MS (Nu Instrument, UK) combined with a desolvator nebulizer unit (DSN-100, Nu Instrument). The Pb isotopic compositions of the NIES CRM No. 28 and other aerosol samples are summarized in Table 6. Combining the isotope systematics ($^{206}$Pb/$^{204}$Pb and $^{87}$Sr/$^{86}$Sr) yield constraints for the source of the Beijing aerosols (see Figure 3), the results showed the impact of emissions from coal combustion and cement plants. The volume size distribution of the CRMs showed that particles with a diameter of ~7 μm were present with the highest frequency (Mori et al., 2008). According to Pb concentrations and Pb isotopic compositions, Widory et al. (2010) suggested that PM$_{2.5}$ samples are expected to be primarily influenced by activities such as lead refining, while the coarser TSP fraction is attributed to activities such as coal combustion or emissions from cement plants. The average $^{207}$Pb/$^{206}$Pb value and Pb concentration of aerosols from lead refining plants, coal combustion, and cement factories are 0.8868 ± 0.031 and 920,967 ± 165,969 ppm (2SD, $n = 6$), 0.8583 ± 0.179 and 109 ±
14 ppm (2SD, \( n = 4 \)), and 0.8616 ± 0.278 and 21 ± 16 ppm (2SD, \( n = 3 \)), respectively (Widory et al., 2010). The Pb concentration of lead refining plants is significantly high compared to that of CRM. The lead refining plants’ derived aerosols cannot be the major source, but a minor contribution to the CRM might be possible. A similarity in the Pb isotopic compositions of the CRM and TSPs in leaded gasoline vehicle exhaust from Shanghai was also reported (Chen et al., 2005). Since the complete ban on the use of alkyllead in 2000, atmospheric Pb emissions have significantly decreased. However, unleaded gasoline still contains a small amount of Pb, inherited from the crude oil, thus it could be a source of contamination (Wang et al., 2003; Bi et al., 2017). The original material for the CRM was recovered before this prohibition and during the phase-out of leaded gasoline, so the Pb isotopic ratio may record such environmental conditions.

**CONCLUSION**

NIES CRM No. 28 Urban Aerosols was originally prepared to certify mass fractions of major and minor elements. In this study, the Sr isotopic composition of the CRM was determined to provide an appropriate quality assurance/quality control tool for Sr isotopic analyses of atmospheric particles. To validate and ensure the accuracy of our method, secondary reference materials, JA-2 and JB-1b, were pretreated in the same way as the CRM, and Sr isotopic compositions were measured using MC-ICP-MS. According to our results regarding within- and between-bottle variations of CRM subsamples, the CRM was sufficiently homogenous to be used for Sr isotopic measurements. As part of
an interlaboratory CRM study, same sample aliquots were measured using TIMS. The results confirmed the consistency of the isotopic ratio using two instruments (MC-ICP-MS vs. TIMS). We also confirmed the consistency of the Sr isotopic composition using different digestion methods (hotplates vs. digestion bombs) and Sr separation (Sr spec resin vs. cation exchange resin). The results of our isotopic analysis contribute to the quality assurance of environmental aerosol monitoring studies.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

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AY, KY, DA, SB, and OD designed the research. AY, KN, MU, KO, KY, TS, KT, TK, and SB performed the analytical work. AY, KY, and TK wrote the manuscript. All the authors discussed the data and revised and approved the final form of the manuscript.

ACKNOWLEDGMENTS

We acknowledge Dr. M. Nishikawa and Dr. M. Sano of NIES for their helpful discussions. We especially thank two reviewers, Dr. J. R. Ferreira and Dr. A. Satkoski, for the thoughtful and constructive comments, and Prof. R. P. Mason for the efficient editorial handling of this paper. We would also like to thank Enago (www.enago.jp) for the English-language review.

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