Moss Biomonitoring of Transboundary Pollutants in Japan's Mountains

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Research Article

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Abstract

Long-range transported atmospheric pollutants (or transboundary pollutants) include trace metals with isotope ratios and compositions that vary from those of domestic pollutants, which threaten mountain ecosystems. These differences can be applied as indices (trace metal indices) to evaluate the influence of transboundary pollutants on mountain ecosystems. Mosses play important ecological functions in mountains and are sensitive to atmospheric deposition. Therefore, using these indices for moss biomonitoring can provide a more accurate indication of ecosystem health. However, few studies have examined which indices are appropriate for moss biomonitoring. In this study, the effectiveness of moss biomonitoring using trace metal indices was examined for evaluating transboundary pollutants in mountainous areas in Japan. Transboundary pollutants in these areas originate from mainland Asia and are characterized by high lead isotope ratios, lead to zinc (Pb/Zn) ratios, and arsenic to vanadium (As/V) ratios. Given that the abundance of transboundary pollutants decreases with distance from mainland Asia, moss isotope indices are also expected to vary with distance. Based on observations, Pb isotope ratios were found to decrease with distance from mainland Asia; in contrast, Pb/Zn and As/V ratios did not display any notable relationship with distance. These results are likely attributed to biological and environmental factors that affect trace metal contents in moss. Thus, moss Pb isotope ratios are useful indicators of transboundary pollutants in Japan's mountains, offering an important tool for comparable moss biomonitoring studies in East Asia.

Introduction

Long-range pollutants that are transported across national borders are termed 'transboundary pollutants.' These pollutants are transported long distances from their emission sources and threaten many of Earth's ecosystems. In particular, transboundary pollutants can become trapped in high mountains owing to low temperature that support dry deposition, high precipitation and fogs that increase wet deposition, and high organic content in soil that can retain large amounts of pollutants (Belis et al. 2009 ; Brahney et al 2015; Daly and Wania 2005; Camarero 2017). Atmospheric pollutants impact trophic status and species interactions, and cause loss of biodiversity in mountain ecosystems. For example, increased inputs of atmospheric phosphorous affect trophic status in alpine lakes as nutrient availability may be tightly coupled with the deposition of these pollutants (Brahney et al 2015). Nitrogen pollutants impact species interactions by providing a competitive advantage to nitrophytic species over nitrogen sensitive species (Sala et al 2000; Bobbink et al 2010). The influence of nitrogen pollutants is particularly severe in nitrogen-depleted ecosystems such as alpine ecosystems (Bobbink et al 2010). Hazardous pollutants transported to mountains are bio-magnified through food chains (Mazzoni et al 2020), being a driver of ecosystem degradation.

The impacts of transboundary pollutants on mountain ecosystems are of great concern in East Asia because of the rapidly increasing emissions of atmospheric pollutants (Wang et al. 2014). Such pollutants are transported to Japan from mainland Asia via westerly winds (Var et al. 2000; Aikawa et al. 2010). The mechanism of these pollutants reaching mountainous areas has been examined based on

analyses of aerosols (Suzuki et al. 2008), rain and fog (Uehara et al. 2015), snow (Yamamoto et al. 2018), lake sediments (Hosono et al. 2016), and alpine vegetation (Oishi 2018, 2019). Notably, transboundary pollutants are significantly associated with forest decline in Japan, having harmful effects on trees by injuring plant tissues, inhibiting the growth, and accelerating senescence (Takahashi et al. 2020). Despite these risks, pollutant monitoring in Japan's mountainous areas remains limited owing to the difficulties of installing measuring equipment in mountainous terrain. Therefore, simple monitoring techniques are urgently required.

Content of trace metals in plants are indicative for evaluation of levels of atmospheric pollution. Several types of plant groups have been used for biomonitoring, among which, mosses have been identified as good indicators for atmospheric pollutants including transboundary sources (Oishi 2018). Biomonitoring using moss (hereafter 'moss biomonitoring') is based on the recognition that mosses efficiently take up pollutants from atmospheric deposition and, thus, the pollutant content of moss reflects atmospheric deposition (Harmens et al. 2015). Thus, moss biomonitoring can be utilized as an effective indicator for ecosystem health given that mosses play important ecological roles and are sensitive to environmental changes (Frego 2007). Because of these characteristics, moss biomonitoring has been increasingly applied in Japan's mountainous areas in recent years, although the detection of transboundary pollutants varies according to pollutant concentrations and the efficiency of their uptake by mosses (Oishi 2019; Oishi et al. 2021). Therefore, further investigation of the application of moss biomonitoring for studying the influence of transboundary pollutants in this region is required.

Several indices, including Pb isotope ratio, Pb to Zn (Pb/Zn) ratios, and As to V (As/V) ratios (Hioki et al. 2008; Mukai et al. 1994; Yonemochi et al. 2018), have been applied for monitoring transboundary pollutants in Japan. Lead has four stable isotopes (^{204}Pb , ^{206}Pb , ^{207}Pb , and ^{208}Pb), the ratios of which are specific to source regions and do not change during industrial and environmental processes (Cheng and Hu 2010). Taking advantage of these characteristics, Pb isotope ratios have been successfully applied to detect transboundary pollutants in Japan (Mukai et al. 1999; Hosono et al. 2016). In comparison, Pb/Zn and As/V ratios are strongly linked to coal combustion, which is a major energy source in northern East Asia (Hayakawa 2009; Zhang et al. 2017). For example, concentrations of chalcophile elements, such as Pb and As, are elevated in the fly ash produced during coal combustion (Coles et al. 1979). Consequently, air masses transported from mainland Asia exhibit higher Pb/Zn and As/V ratios relative to those from within Japan (Mukai et al. 1994; Hioki et al. 2008; Yonemochi et al. 2018).

Based on these differences, trace metal indices (Pb isotope ratios and Pb/Zn and As/V ratios) offer great potential for moss biomonitoring of transboundary pollutants; however, the applicability of Pb/Zn and As/V ratios has not yet been tested in this context. Furthermore, existing research indicates that moss biomonitoring based on Pb isotope ratios may not be able to identify transboundary pollutants in Japan's mountains (Oishi et al. 2021), despite its usefulness having been reported elsewhere (Farmer et al. 2002; Xiang et al. 2017; Schnyder et al. 2018, Sucharová et al. 2014).

This study examines the usefulness and limitations of various trace metal indices for moss biomonitoring of transboundary pollutants in Japan's mountainous regions. Given that the concentrations of transboundary pollutants decrease with distance from mainland Asia (Toyama et al. 2013), a general hypothesis was applied assuming that the trace metal indices of moss will also change with distance. By exploring these relationships, this work contributes to the broader understanding of the applicability of moss biomonitoring in mountain ecosystems and supports the more widespread use of moss biomonitoring in East Asia as a relatively simple and useful technique.

Materials And Methods

Study site

The study sites were selected from mountainous areas in central Japan including, from north to south, the Osado Mountains, Mount Hakusan, the Japanese Northern Alps, the Japanese Central Alps, the Yatsugatake Mountains, and the Japanese Southern Alps (Fig. 1, Table S1). These mountains were grouped into four areas according to geographical location, i.e., the northernmost area (the Osado Mountains), the northern area (Mount Hakusan and the Northern Alps), the central area (the Central Alps, the Yatsugatake Mountains, and the northern part of the Southern Alps), and the southern area (the Southern Alps). Moss sampling was conducted in two or three mountains in each of the four areas (Table 1). The selection of these sites was based on the information of presence of a targeted moss species (*Racomitrium lanuginosum*). This species is widely distributed at high elevations throughout the world (Iwatsuki 1988) and is recommended as a bioindicator for metal contamination (Wojtuń et al. 2018). The moss samples were randomly collected in five, open locations at each site, and pooled to obtain homogenized samples. The samples were sealed in polyethylene bags and stored until Pb analysis.

Chemical analysis

Metal-free equipment was used in all chemical analyses to prevent contamination. First, detrital material was carefully removed from the moss samples using plastic tweezers. Then, the green parts of shoot tips (approximately 1–2 cm) were detached from the moss samples, oven-dried at 60 °C for 48 h, and their dry weight measured. The dried samples were homogenized and digested for the analysis of trace metal and Pb isotope ratios using inductively coupled plasma mass spectrometry (ICP-MS; 7500cx, Agilent Technologies, Tokyo, Japan) and multi-collector double-focusing ICP-MS (MCICP-MS) (Neptune Plus, Thermo Fisher Scientific, Bremen, Germany). The procedures of these analyses are described in detail by Oishi et al. (2021). Subsequently, $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ values were calculated based on the obtained Pb isotope values ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$). All analyses were conducted at the Research Institute for Humanity and Nature, Kyoto, Japan.

Analysis

The differences of $^{208}\text{Pb}/^{206}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$, Pb/Zn, and As/V values among the four areas were compared using Bonferroni's multiple-comparison.

Next, $^{208}\text{Pb}/^{206}\text{Pb}$ was plotted against $^{207}\text{Pb}/^{206}\text{Pb}$ to examine their relationships with transboundary pollutants. The values of these ratios for snow and precipitation of domestic and transboundary origin were used for comparison. The Pb isotope ratios in snow, transported via air mass from far-eastern Russia, Sino-Russian border region, Northern China, Central China and Korean Peninsula, and domestic regions are based on Mukai et al. (1999). Similarly, Pb isotope ratios in precipitation was are based on Mizoguchi et al. (2012). These Pb isotope ratios are listed in Table S2.

Finally, Pb/Zn was plotted against As/V to discuss the influence of transboundary pollutants on moss. Previous studies report that aerosols transported from mainland Asia exhibit Pb/Zn ratios >0.5 (Tsuji and Hioki 2013) and As/V ratios >1.0 (Yonemochi et al. 2018). Trace metal sources in moss were examined based on these values.

Results

Pb isotope ratios

The values of three indices (Pb isotope ratios and Pb/Zn and As/V ratios) are listed in Tables S3 and S4. The comparison of Pb isotope ratios is presented in Fig. 2. The $^{207}\text{Pb}/^{206}\text{Pb}$ (average $\pm$ SD) in the northernmost, northern, central, and southern area were 0.8656 ± 0.0006 , 0.8620 ± 0.0009 , 0.8609 ± 0.0006 , and 0.8590 ± 0.0008 , whereas the $^{208}\text{Pb}/^{206}\text{Pb}$ (average $\pm$ SD) were 2.114 ± 0.0012 , 2.1076 ± 0.0010 , 2.1074 ± 0.0007 , and 2.1062 ± 0.0012 , respectively. Both $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ values in the northernmost area were significantly higher than those of other areas.

Based on the Pb isotope ratios of transboundary pollutants (Table S2), it can be observed that the Pb isotope ratios of moss are similar to those for precipitation and snow transported from the Sino-Russian border region and Northern China. The scatterplot of Pb isotope ratios (Fig. 3) indicates that the Pb isotope ratios decrease with the distance from mainland Asia (northern–central–southern) and were similar to those of domestic pollutants (Table S2).

Pb/Zn and As/V ratios

In contrast to the Pb isotope ratios, the Pb/Zn vs. As/V scatterplot (Fig. 5) did not indicate any significant differences among study areas (Fig. 4) and notable geographic pattern. Except for one site (site 6), all of the Pb/Zn values fall within the reported range for air masses transported from within Japan (< 0.5 ; Tsuji and Hioki 2013). In all cases, the As/V ratios were lower than those reported for transboundary pollutants (> 1.0 ; Yonemochi et al. 2018).

Discussion

Pb isotope ratios and transboundary pollutants

Among the trace metal indices of transboundary pollutants, only the Pb isotope ratios decreased with distance from mainland Asia, corresponding to the abundance of transboundary pollutants. Based on this observation, the Pb isotope ratios of moss are considered good indicators of transboundary pollution. These results are noticeable in that Pb isotope ratios of moss in remote areas are primarily derived from domestic sources in other countries (Sucharová et al. 2014; Zhou et al. 2021). These differences imply that transboundary pollutants have a strong influence on mountain ecosystems in Japan.

Consequently, the contribution of transboundary sources to the Pb content of moss was successfully distinguished from domestic inputs based on their specific values. The similarity of the Pb isotope ratios suggest that the northernmost area could be strongly influenced by transboundary pollutants from the Sino-Russian border region and Northern China. These associations correspond to the results of back trajectory analysis of air mass by Mukai et al (1999) that reveal high susceptibility of the northernmost areas to air mass transported from these regions.

In contrast, the reduced influence of transboundary pollutants in other areas resulted in the dominance of domestic sources for the Pb contents in moss; this observation is supported by the significant lower values of Pb isotope ratios compared to the those in the northernmost area. Building on previous work (Oishi et al. 2021), these observations indicate that transboundary pollutants have a limited influence on moss Pb isotope ratios in these inland mountains.

Notably, these results vary from those obtained by evaluations of transboundary pollutants using abiotic materials. In contrast to Pb isotope ratios of moss, an abundance of transboundary Pb pollutants was detected in the precipitation (Mizoguchi et al. 2012), snow (Mukai et al. 1999), and lake sediment (Hosono et al 2016) across the northern–southern areas. These inconsistent results highlight the differences in the responses to transboundary Pb pollutants between mosses and abiotic factors. A possible cause for the varying responses could be the exposure period to transboundary pollutants. In the studied mountains, mosses are covered with snow from late-Autumn to Spring and protected from exposure to transboundary pollutants. Given that transboundary Pb pollutants are primarily carried to Japan during these periods (Hioki et al. 2006), the influence of transboundary pollutants on moss can be lower compared to that on abiotic factors.

Pb/Zn and As/V ratios and transboundary pollutants

The Pb isotope ratios, the Pb/Zn and As/V ratios did not decrease with increase in distance from emission sources (in this case mainland Asia). This can be explained by biological and environmental factors that affect the contents and sources of trace metals in mosses. For example, the trace metal content of moss changes with growth rate (Zechmeister 1998) and biomass (Gerdol et al. 2002), which can largely differ among mountains with varying climates. Furthermore, mosses take up trace metals from the soil via several pathways including the upward capillary movement of soil water, rain splash, and wind-blown soil particles (Bargagli et al. 1995; Økland et al. 1999; de Caritat et al. 2001). Thus, in addition to the biological factors such as growth rate and biomass, soil-derived trace metals can

influence the trace metal content of moss. Given that a terrestrial species of moss (*R. lanuginosum*) growing in open areas was sampled for this study, the influence of soil-derived trace metals may have been enhanced compared to that in abiotic indicators. In this regard, Pb/Zn and As/V ratios in epiphytic bryophytes that are less subjected to soil particles, might better reflect the influence of transboundary pollutants than terrestrial bryophytes.

However, the influence of soil-derived Pb on moss is small, which suggests a strong correlation of Pb isotope ratios between mosses and atmospheric deposition. This is because Pb in soil is immobilized due to binding with organic matter (Heinrichs and Mayer 1977); hence, the amount of soil-derived Pb available for uptake by moss, is insubstantial (Økland et al. 1999). Therefore, Pb isotope ratios can more reliably indicate the influence of transboundary pollutants on the ecosystem, as compared to Pb/Zn and As/V ratios.

Conclusions

The results of this study confirm that the Pb isotope ratios of mosses can serve as useful indicators of transboundary pollutants in the mountainous areas of Japan. The ratios decreased with distance from mainland Asia, reflecting the relative influence of transboundary pollutants in the inland mountain regions. However, Pb/Zn and As/V ratios in moss cannot be considered as reliable indicators owing to the strong influence of biological factors and soil-derived metals. The effect of this influence on analysis results may be mitigated by using epiphytic moss for biomonitoring; however, biological factors could continue to affect the ratios.

This study also compared the varying responses of Pb isotope ratios in moss and abiotic materials. These differences indicate the limitations of using moss biomonitoring as an alternative method to monitoring through physical equipment. Nevertheless, the results highlight the usefulness of moss biomonitoring as it facilitates the evaluation of ecosystem health from unique perspectives compared to that of abiotic factors. This evaluation method can be useful for detecting changes in ecosystems associated with ecological functions of bryophytes. For this purpose, further examination of the relationships between ecosystem health indicators and moss Pb isotope ratios should be conducted, which can also be used to assess the applicability of moss biomonitoring of transboundary pollutants in mountain ecosystems.

Finally, as the moss species used in this study is widely distributed across the high-elevation regions of East Asia (Iwatsuki 1988), the reported results offer an important reference for comparative studies of moss biomonitoring across this region.

Declarations

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Data availability: All data generated or analyzed during this study are included in this published article and its supplementary information files.

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Table

Table 1 is not available with this version.

Figures

Figure 1

(a) Location of the study site. The study sites (10 mountains) were grouped into the following four areas: northernmost, northern, central, and southern areas. 1. Mount Donden, 2. Mount Myoken, 3. Mount Hakuba-Norikura, 4. Mount Yakedake, 5. Mount Hakusan, 6. Mount Yokodake, 7. Mount Kisokomagatake, 8. Sensuitoge, 9. Mount Ainodake, and 10. Mount Okuhijiridake. (b) *Racomitrium lanuginosum* communities near the top of Mount Yakedake. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 2

Comparison of Pb isotope ratios. The differences in significance among the study areas are shown by the corresponding alphabet (multiple-comparison: $p < 0.05$)

Figure 3

Scatterplot of Pb isotope ratios ($^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$). The $^{207}Pb/^{206}Pb$ values of snow and precipitation from transboundary and domestic areas are listed in Table S2.

Figure 4

Comparison of trace metal ratios (Pb/Zn and As/V). The differences in significance among the study areas are shown by the corresponding alphabet (multiple-comparison: $p < 0.05$)

Figure 5

Scatterplot of trace metal ratios (Pb/Zn and As/V). Aerosol transported from mainland Asia exhibits Pb/Zn ratios > 0.5 (Tsuji and Hioki 2013) and As/V ratios > 1.0 (Yonemochi et al. 2018)

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