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Source analysis of radiocesium in river waters using road dust tracers

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HIGHLIGHTS

- Utility of metals as road dust tracers for source analysis of ^{137}Cs was evaluated.
- Road dust had more Zn and less Al and Fe than soils.
- Al, Fe, and Zn were detected frequently from suspended solids in river water.
- Combinations of metals and ^{137}Cs were useful in analyzing sources in river water.
- Contribution from road dust to radiocesium in river water was limited.

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ABSTRACT

Following the Fukushima Dai-ichi Nuclear Power Station accident, regional road dust, heavily contaminated with radiocesium, now represents a potential source of radiocesium pollution in river water. To promote effective countermeasures for reducing the risk from radiocesium pollution, it is important to understand its sources. This study evaluated the utility of metals, including Al, Fe, and Zn as road dust tracers, and applied them to analyze sources of ^{137}Cs in rivers around Fukushima during wet weather. Concentrations of Zn in road dust were higher than agricultural and forest soils, whereas concentrations of Fe and Al were the opposite. Concentrations of Zn were weakly but significantly correlated with benzothiazole, a molecular marker of tires, indicating Zn represents an effective tracer of road dust. Al, Fe, and Zn were frequently detected in suspended solids in river water during wet weather. Distribution coefficients of these metals and ^{137}Cs exceeded 10^4, suggesting sorptive behavior in water. Although concentrations of Al, Fe, Zn, and ^{137}Cs were higher in fine fractions of road dust and soils than in coarse fractions, use of ratios of ^{137}Cs to Al, Fe, or Zn showed smaller differences among size fractions. The results demonstrate that combinations of these metals and ^{137}Cs are useful for analyzing sources of radiocesium in water. These ratios in river water during wet weather were found to be comparable with or lower than during dry weather and were closer to soils than road dust, suggesting a limited contribution from road dust to radiocesium pollution in river water.

1. Introduction

After the 2011 Great East Japan Earthquake and tsunami, radiocesium (^{134}Cs and ^{137}Cs) emitted from the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Station were accumulated on the land surface via wet and dry deposition (Kinoshita

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et al., 2011). Subsequently, radiocesium washed off into streams and rivers has been detected in aquatic organisms (e.g., fish; Buesseler, 2012). Concerns regarding human health and ecosystems have driven decontamination programs and restrictions on the distribution of food including vegetables and fish (Murakami and Oki, 2012; Yasutaka et al., 2013). Although doses of radiocesium from dietary intake have been very small under the restricted food distribution (Murakami and Oki, 2014), continued suspension of the fish trade has caused economic damage to the fisheries industry. For effective risk management, comprehensive understanding of the sources, behavior, and fate of radiocesium is urgently required.

In Fukushima, the transport of radiocesium from land surfaces to water and its subsequent behavior in freshwater environments have been well studied (Evrard et al., 2015). For instance, in terms of the flux of radiocesium in rivers, the particulate form is the major contributor to total runoff, especially for heavy rainfall events (Murakami et al., 2016; Yamashiki et al., 2014). Furthermore, the amount of radiocesium wash-off has been investigated for different land use types (e.g., forests, agricultural fields, and urban areas; Koibuchi et al., 2015; Murakami et al., 2016; Yamashiki et al., 2014; Yamashita et al., 2015; Yoshimura et al., 2014). In particular, intensive station-based monitoring has been performed in the Abukuma River water system that drains through the Fukushima area (Tsuji et al., 2014; Yamashiki et al., 2014). A decrease of ^{137}Cs outflow from the Abukuma River water system has been predicted, although it is expected to continue over the long term (Yamaguchi et al., 2014). In this regard, identifying sources of radiocesium in freshwater is particularly valuable at the catchment scale.

Understanding the sources of radiocesium contamination is useful in the development and implementation of effective management strategies (e.g., identifying priority areas to be remediated). However, only a few investigations have analyzed the sources of radiocesium in river water in the Fukushima area (Yamashita et al., 2015; Lepage et al., 2016; Laceby et al., 2016). A useful approach in source analysis is the application of molecular markers or tracers, which are contained in a specific source. The utility of tracers can be evaluated thorough multiple factors (Nakada et al., 2008; Takada and Eganhouse, 1998): (1) occurrence in a specific source, (2) frequency of detection from samples, and (3) similarities in environmental behavior (e.g., adsorption, desorption, degradation, and wash-off behavior). Among the potential sources on land surfaces of a catchment, benzothiazolamines including benzothiazole (BT) and 2-(4-morpholinyl) benzothiazole have been used as possible markers for tire debris and/or road dust for source analysis (Kumata et al., 2002; Spies et al., 1987). In addition, metals such as Zn (tires; Funasaka et al., 2003), Cu (brake linings; Weckwerth, 2001), Pb, Cr (yellow road line markings; Murakami et al., 2007), and Pt (catalysts; Ash et al., 2014) originate from specific sources associated with automobiles and traffic activities, and have been detected in high concentrations in road dust and urban aerosols. However, the suitability of using metals as road dust tracers has not been investigated fully. Additionally, in using these tracers, it would be expected that the contributions of radiocesium from urban surface deposits (e.g., road dust) and other sources (e.g., forest and agricultural soils) to river water could be distinguished.

This study had two objectives. First, the utility of metals and a metalloid as tracers of road dust was evaluated from five viewpoints: (1) differences in concentrations between road dust and soils; (2) correlations with BT, a molecular marker of tires; (3) detection frequencies in river water; (4) similarities in the distribution coefficient K_d (the ratio of concentration in the particulate phase on a suspended solids [SS] weight basis to that in the dissolved phase on a liquid volume basis) with ^{137}Cs; and (5) similarities in the their concentrations and ratios in relation to particle size fractions. Second, the tracers were applied to evaluate the sources of radiocesium in SS in river water during wet weather in the tributaries of the Abukuma River. This is the first study to reveal the contribution of road dust to radiocesium pollution in river water through assessment using metals as road dust tracers.

2. Materials and methods

2.1. Sample collection

This study focused on the Abukuma River water system and its basin (Fig. 1). The Abukuma River basin has a basin area of approximately 5400 km^2 and its land use types are as follows: forests, 56%; paddy fields, 16%; fields, 14%; urban surfaces (including roads), 7%; and others, 7% (Kinouchi and Watanabe, 2011). Overall, 10 samples of river water were collected from two tributaries of the Abukuma River water system (i.e., the Gohyaku River [st.A] and the Ouse River [St.B]) on August 27, 2015 (wet

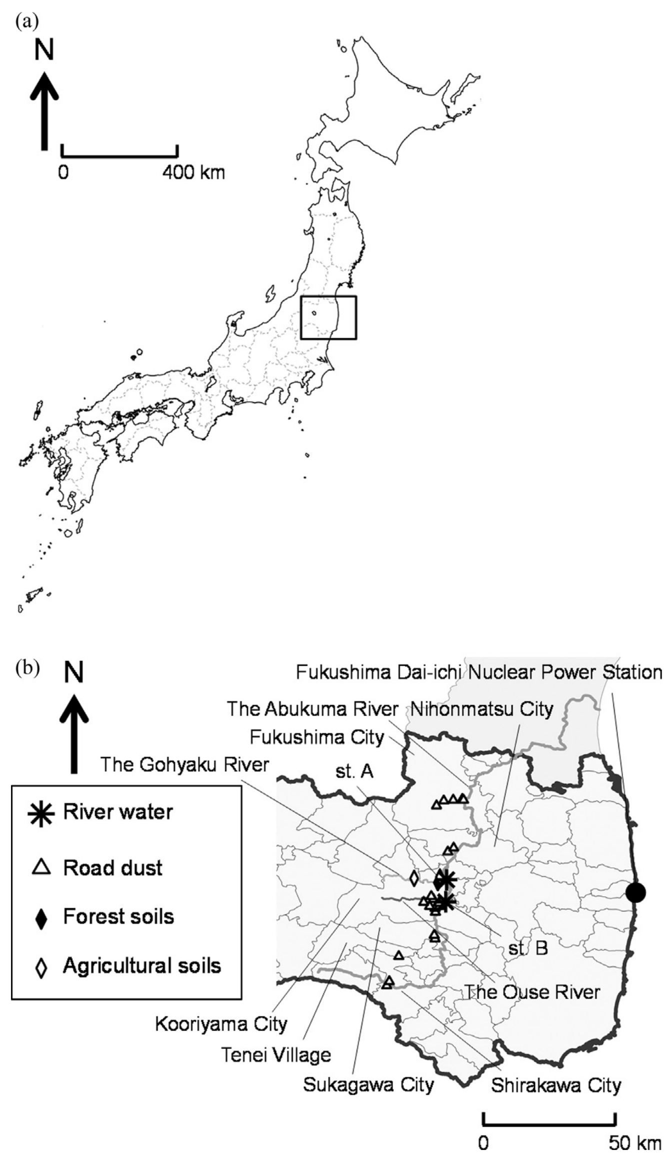

Fig. 1. Sampling locations. (a) Japan, (b) Fukushima Prefecture. Rivers comprise part of the Abukuma River water system.