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Radioactive cesium dynamics derived from hydrographic observations in the Abukuma River Estuary, Japan

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ABSTRACT

Large quantities of radioactive materials were released into the air and the ocean as a result of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, caused by the 2011 Tohoku earthquake and the subsequent major tsunami off the Pacific coast. There is much concern about radioactive contamination in both the watershed of the Abukuma River, which flows through Fukushima Prefecture, and its estuary, where it discharges into the sea in Miyagi Prefecture. We investigated radioactive cesium dynamics using mixing diagrams obtained from hydrographic observations of the Abukuma River Estuary. Particulate radioactive cesium dominates the cesium load in the river, whereas the dissolved form dominates in the sea. As the salinity increased from <0.1 to 0.1–2.3, the mixing diagram showed that dissolved radioactive cesium concentrations increased, because of desorption. Desorption from suspended particles explained 36% of dissolved radioactive cesium in estuarine water. However, the dissolved and particulate radioactive cesium concentrations in the sea decreased sharply because of dilution. It is thought that more than 80% of the discharged particulate radioactive cesium was deposited off the river mouth, where the radioactive cesium concentrations in sediment were relatively high (217–2440 Bq kg^{-1}). Radioactive cesium that was discharged to the sea was transported southward by currents driven by the density distribution.

1. Introduction

The Tohoku earthquake and subsequent major tsunami off the Pacific coast of Japan on 11 March 2011 triggered the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, which resulted in the release of large quantities of radioactive materials to the air and ocean (Buesseler et al., 2011; Chino et al., 2011; Yasunari et al., 2011; Tsumune et al., 2012; Kumamoto et al., 2015). Although a variety of radioactive materials were emitted into the environment (Povinec et al., 2013), ^{134}Cs and ^{137}Cs have received much attention because of their potential impacts on public health and ecosystems, and their relatively long half-lives (2.07 y and 30.07 y, respectively). The total amount of radioactive cesium directly discharged into the sea was estimated to be between 3 and 6 PBq (Kawamura et al., 2011; Estournel et al., 2012; Tsumune et al., 2013). Moreover, more than 10 PBq of radioactive cesium was deposited into the ocean from the atmosphere (Morino et al., 2013; Miyazawa et al., 2013).

The Abukuma River, a major river in the Tohoku Region of Japan, is 234 km long. It has a watershed area of 5400 km^2, and has an average discharge of 200 $m^3 s^{-1}$ in its lower reaches. While it flows through both Fukushima and Miyagi Prefectures (Fig. 1), most of its catchment is in Fukushima Prefecture. Relatively higher ^{137}Cs concentrations than before the FDNPP accident have been observed in river water and sediments (Iguchi et al., 2013; Sakaguchi et al., 2014), and ^{137}Cs has been detected in freshwater fish caught in the river (Iguchi et al., 2013; Mizuno and Kubo, 2013). The river flows into Sendai Bay, where relatively higher ^{137}Cs concentrations than the oceanic region off the bay have been observed in seawater and zooplankton (Kaeriyama et al., 2015). The ^{137}Cs concentration of 300 Bq kg^{-1} dry was recorded for coast and off-shore marine sediments in the Abukuma River estuary by a towed gamma-ray spectrometer (Hirao et al., 2014). In contrast, however, it is reported that radioactive cesium concentrations in marine sediments were lower in the area north of the FDNPP than south (Ambe et al.,

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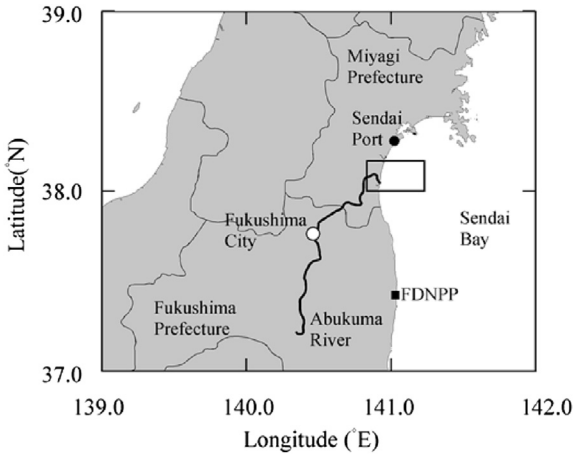

Fig. 1. Locations of the Abukuma River and the observation area (rectangle) shown in Fig. 2. The closed square, closed circle, and open circle indicate the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), the tide gauge station of Sendai Port, and the meteorological station (AMeDAS) of Fukushima city, respectively.

2014). It is thought therefore, that the peak in ^{137}Cs concentrations off the Abukuma River Estuary reported by Hirao et al. (2014) reflects deposition and accumulation of radioactive cesium that was transported to the estuary zone by the river.

To evaluate the influence of any substance on the environment, including radioactive materials, its dynamics must be thoroughly understood. However, even though various studies have been conducted since the tsunami disaster, including examinations of water quality and radioactivity, the dynamics of radioactive substances in the Abukuma River Estuary remain poorly understood (e.g., Kakehi et al., 2012; Kaeriyama et al., 2015; Kakehi et al., 2015). Dilution by seawater is a conservative process that changes the concentrations of the substances discharged from the river. However, in estuaries, where there is a sudden increase in salinity, substances may also be influenced by non-conservative processes such as flocculation, adsorption, release, and desorption (e.g., Sholkovitz, 1976, 1978; Edmond et al., 1985; Burban et al., 1989). Takata et al. (2015) demonstrated experimentally that ^{137}Cs was desorbed from riverine particles when they were mixed with seawater. Mixing diagrams, which plot target substances and salinity, can be used to determine whether the dominant process is conservative or non-conservative (Liss, 1976; Kemp and Boynton, 1984). In the present study, we used a mixing diagram obtained from hydrographic observations in the Abukama River Estuary to examine both types of processes for radioactive cesium dynamics.

2. Materials and methods

2.1. Hydrographic observations

Hydrographic observations were conducted on 22 August 2013 and 5 July 2014 in the Abukuma Estuary (Table 1). In 2013, sampling stations were established in the river at intervals of approximately 0.8 km and in the sea at intervals of between 1.5 and 4 km (Fig. 2a). Samples were mainly collected in the vicinity of the river mouth in 2014 (Fig. 2b and c). Temperature and salinity in the water column were measured with a water quality sensor (AAQ−1186; JFE Advantech, Hyogo, Japan). Water samples for salinity and radioactive cesium analysis were collected at depth of 1 m at all stations with a 6 L Van Dorn water sampler. At station 10, samples were also collected at depths of 20 m (half the water depth) and 38 m (2 m above the bottom). In 2014, three water samples were collected at a depth of 1 m at station R6 in the river and station C5 in the sea for suspended substance (SS) analysis. Bottom sediment samples were collected at all stations in 2013 with an Ekman-Birge sediment sampler (Rigo-sha) that had a sampling area of 0.04 m^2.

Table 1
Summary of sampling station and date.

Station	Latitude	Longitude	Date (year/month/day)
RE1	38° 5.754′	140° 52.287′	2013/8/22
RE2	38° 5.797′	140° 52.836′	2013/8/22
RE3	38° 5.690′	140° 53.269′	2013/8/22
RE4	38° 5.207′	140° 53.961′	2013/8/22
RE5	38° 4.867′	140° 54.315′	2013/8/22
RE6	38° 4.353′	140° 54.629′	2013/8/22
RE7	38° 3.806′	140° 54.472′	2013/8/22
RE8	38° 3.342′	140° 54.348′	2013/8/22
RE9	38° 3.112′	140° 54.674′	2013/8/22
RE10	38° 3.004′	140° 54.942′	2013/8/22
SE1	38° 2.866′	140° 55.922′	2013/8/22
SE2	38° 2.863′	140° 56.820′	2013/8/22
SE3	38° 2.866′	140° 57.821′	2013/8/22
SE4	38° 2.871′	140° 58.812′	2013/8/22
SE5	38° 2.866′	141° 0.815′	2013/8/22
SE6	38° 2.872′	141° 2.809′	2013/8/22
SE7	38° 2.872′	141° 4.807′	2013/8/22
SE8	38° 2.870′	141° 6.805′	2013/8/22
SE9	38° 2.857′	141° 9.819′	2013/8/22
SE10	38° 2.870′	141° 12.820′	2013/8/22
C5	38° 1.830′	141° 4.980′	2014/6/28
R1	38° 3.290′	140° 54.270′	2014/7/5
R2	38° 3.137′	140° 54.708′	2014/7/5
R3	38° 3.057′	140° 54.973′	2014/7/5
R4	38° 3.042′	140° 54.901′	2014/7/5
R5	38° 3.039′	140° 54.897′	2014/7/5
R6	38° 5.785′	140° 52.618′	2014/7/5
RE6	38° 4.346′	140° 54.624′	2014/7/5
RE8	38° 3.302′	140° 54.275′	2014/7/5
S1	38° 2.876′	140° 55.806′	2014/7/5
S2	38° 2.890′	140° 55.888′	2014/7/5
S3	38° 3.031′	140° 55.711′	2014/7/5
S4	38° 3.107′	140° 55.786′	2014/7/5

Current measurement data were collected onboard a fishing boat equipped with an Acoustic Doppler Current Profiler (ADCP; Workhorse Sentinel 600 kHz; Teledyne RD Instruments) on 22 August 2013. The boat traversed repeatedly between stations SE4 and SE9 from sunrise to sunset. The boat speed was reduced to <1 m s^{-1} at sampling stations, and the vertical current distribution was measured in depth increments of 1 m from 2 m below the surface to 2 m above the bottom for 2–10 min. Current measurements were not recorded between stations when the boat was traveling at full speed. Current data were recorded by ADCP eight or nine times in a 12-h period.

2.2. Analytical methods

Salinity was measured with a salinometer (AUTOSAL 8400B; Guildline Instruments). The salinity values from the AAQ were calibrated with those obtained by the salinometer. Before measuring the SS concentrations, samples of river water and seawater (0.8 and 11.5 L, respectively) were filtered through preweighed 47 mm Whatman GF/F glass fiber filters. After filtration, the filters were rapidly rinsed with MiliQ® water to eliminate salt. The filters were dried for a few days in a desiccator with silica gel at room temperature. The SS concentrations were determined as the difference between the weight of the filter before and after filtration. The average and standard deviations of the SS concentrations were calculated from the three samples collected at each station.

We measured dissolved and particulate radioactive cesium concentrations per liter of water (mBq L^{-1}) as follows. We collected