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Oxygen and hydrogen isotope characteristics of seawater in Otsuchi Bay and meteoric water of inflowing rivers

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Abstract — Oxygen isotopes of biogenic calcium carbonate can be used to reconstruct salinity if the oxygen isotope–salinity (δ^{18}O–S) relationship for the region of interest is known. In Otsuchi Bay, located on the northeast coast of Japan, δ^{18}O–S relationship has been reported; however, the data were only collected from the mouth of the bay and covered a limited salinity range, which restricts the applicability of the relationship in reconstructions of salinity. This study is the first to establish a δ^{18}O–S relationship for Otsuchi Bay that can be applied to a wide range of salinities, from zero (fresh water) to 34 (open ocean water), using water samples collected from September 2014 to September 2015 from the middle of the bay (Hourai-jima): The obtained relationship was $\delta^{18}O=(0.28\pm0.01)\ S-(9.31\pm0.41)$, and the slope was shallower than that of the previous study. We also sampled waters in the lower reaches of the three main rivers that flow into Otsuchi Bay: the Otsuchi, Kozuchi and Unosumai rivers, and determined their oxygen and hydrogen isotope compositions. No statistically significant differences were detected between the rivers, and no distinct seasonality was observed. Calculated d-excess values of these newly obtained data are consistent with previously reported values of stream water collected in the region.

Key words: oxygen isotope, hydrogen isotope, salinity, d-excess

Introduction

Otsuchi Bay is a rectangular-shaped bay located on the northeast coast of Japan, measuring 8 km from east to west and 2 km north to south (Fig. 1). The water depth increases gradually towards the bay mouth and reaches 70 m at the mouth of the bay. Three main rivers (the Otsuchi, Kozuchi, and Unosumai rivers) flow into the bay. Each river carries fresh water at a rate of 3–10 m^3/s. The salinity in Otsuchi Bay is lower than that in the open ocean through most of the year. There are three major currents that potentially affect the open ocean water adjacent to Otsuchi Bay: the warm Kuroshio Current from the subtropical North Pacific, the cold Oyashio Current from the subarctic North Pacific, and the Tsugaru Warm Current originating from the Tsushima Current that flows through the Sea of Japan (Hanawa 1983, Hanawa and Mitsudera 1986, Oguma et al. 2002, Yasuda, 2003, Ishizu et al. 2016).

The aim of this study is to establish an oxygen isotope–salinity (hereafter δ^{18}O–S) relationship in Otsuchi Bay that is applicable to a range of salinities from brackish water to seawater in this region. Establishing such a relationship is important for two reasons. Firstly, past salinity can be reconstructed using δ^{18}O of biogenically precipitated calcium carbonate, such as the shells of bivalves and brachiopods. δ^{18}O of calcium carbonate varies as a function of temperature (4°C increase corresponds 1‰ decrease) and δ^{18}O of the water, and if temperature is reconstructed independently, δ^{18}O of the water mass in which the organism lived can be reconstructed. Subsequently, salinity of the water mass can be reconstructed using the modern δ^{18}O–S relationship (e.g., Grossmann and Ku 1986, Felis et al. 2009, Yamamoto et al. 2011, Kubota et al. 2017). A regional salinity reconstruction would allow for a deeper understanding of climate variability on seasonal to decadal timescales (e.g., frequency of occurrence of heavy rain and snowfall). Secondly, the migratory behavior of amphidromous fish can be modeled using oxygen isotope data of fish otoliths, which are paired ear stones (Campana 1999, Kubota et al. 2015, Amekawa et al. 2016). A deeper understanding of fish ecology is important for the conservation of fish habitats and ecosystems, which in turn benefits local fisheries and people. The pioneering work of Yamamoto et al. (2011) was the first to report a δ^{18}O–S relationship for Otsuchi Bay; however, the relationship was determined from seawater collected from the mouth of the bay at 70 m water depth, where salinity only varies from 33.5 to 34.5. The salinity range of Otsuchi Bay overall is much larger, from zero (freshwater) to ∼34 (open ocean water); therefore, the δ^{18}O–S relationship reported by Yamamoto et

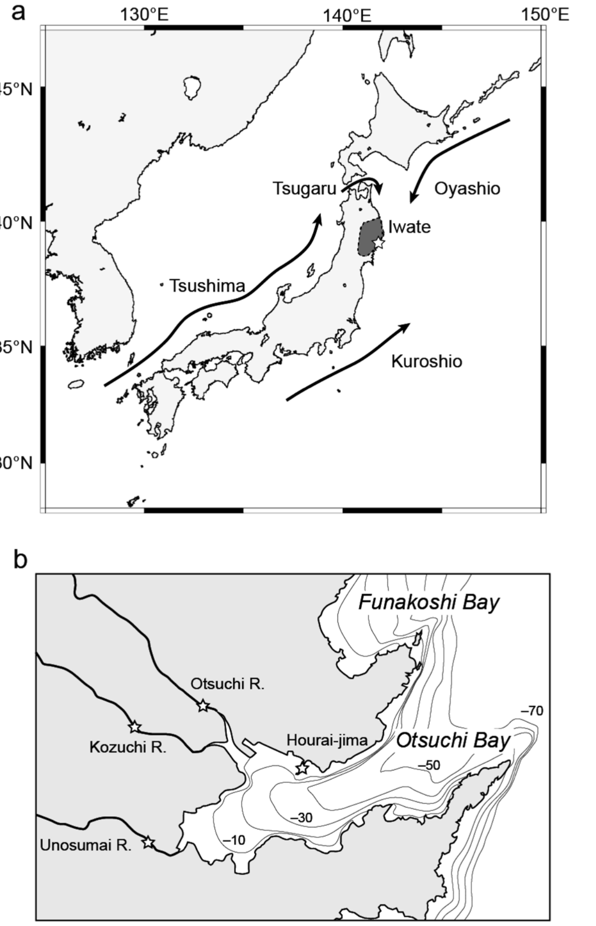

Fig. 1. **(a)** Location of the study area, with shading indicating the Iwate region. The star signifies the location of Otsuchi Bay. Major ocean currents flowing along the Japanese coast are also shown. **(b)** Geographical map of Otsuchi bay, with bathymetric contours at 10 m intervals. Water sampling locations are indicated by stars.

al. (2011) is not applicable to all salinity range of the surface water. To establish a more useful δ^{18}O–S relationship, a time-series dataset of oxygen isotopes and salinity is needed at fixed stations that cover the range of values observed in the bay. In addition, seasonal variations in the δ^{18}O of river water flowing into the bay need to be constrained, to determine the effects of river input (the zero salinity end-member) on the seawater δ^{18}O–S relationship. If large seasonal variations in the δ^{18}O value of river water were to be observed, the δ^{18}O–S relationship for Otsuchi Bay would become more complex, which would limit the applicability of the obtained equation in reconstructions of salinity.

To achieve these aims, we repeatedly collected seawater near the center of the bay, as well as freshwater from the lower reaches of the Otsuchi, Kozuchi and Unosumai rivers (Fig. 1). By collecting water in each of these locations, and measuring the oxygen isotopes and salinity of the water, we monitored seasonal variability of these two variables and established a δ^{18}O–S relationship for Otsuchi Bay. In tandem with oxygen isotope measurements of the water, hydrogen isotope (δD) measurements were also obtained, and we have included a preliminary interpretation of these data.

Materials and Methods

From 11 September 2014 to 15 September 2015, we collected 10 water samples at each of the four fixed locations (star symbols in Fig. 1b). Surface seawater (< 1 m) was collected at Hourai-jima, which is located near the middle of the bay. Surface freshwater was collected in the lower reaches of the Otsuchi, Kozuchi and Unosumai rivers. Weather (daily mean air temperature and daily precipitation, data from Japan Meteorological Agency, available at http://www.data.jma.go.jp) of the sampling days is listed in Table 1. Water samples were stored in tight glass containers and refrigerated until analysis. The salinity of the samples was measured using a portable pH and salinity meter (LAQUA act; Horiba, Japan) calibrated with IAPSO standard seawater (Batch: P153; OSIL, United Kingdom). The salinity measurement of IAPSO standard seawater yielded 34.91, consistent with the certified value of 34.992 (Bacon et al. 2007). We measured only three freshwater samples (the sample set collected on 17 December 2014) and confirmed the salinity to be essentially zero; thus, we did not measure the salinity of the other samples. Water samples were analyzed for δ^{18}O and δD using a wavelength-scanned cavity ring-down spectroscopy isotopic water analyzer (L2120-i; Picarro, USA) at the Atmosphere and Ocean Research Institute, The University of Tokyo, Japan. Repeated analyses of Milli-Q water yielded −12.93±0.10‰ (1 standard deviation (SD)) for δ^{18}O and −72.86±1.31‰ (1 SD) for δD. These standard deviations are used as the analytical uncertainty for isotopic measurements. Isotopic values of water samples are reported relative to Vienna Standard Mean Ocean Water (VSMOW). In the case of δ^{18}O measurements, ±0.10‰ analytical precision is enough for the scope of this study, given the large difference of δ^{18}O values between the seawater (ca. 0‰) and the terrestrial water (ca. −9‰ in this region).

Results

The salinity and isotope measurements are listed in Table 1.

River water

Average δ^{18}O values of the Otsuchi, Kozuchi, and Unosumai rivers were −9.3‰ (range −9.6‰ to −9.0‰), −9.3‰ (range −9.5‰ to −9.2‰), and −9.4‰ (range −9.7‰ to −9.2‰), respectively. The range of values for each river is in good agreement, and there is no clear seasonality (Fig. 2a).

Table 1. Salinity, $\delta^{18}O$, δD, and d-excess values of water collected at Hourai-jima and from the Otsuchi, Kozuchi and Unosumai rivers. Daily mean air temperature and daily precipitation data are from Kamaishi and Otsuchi, respectively.

Fresh water	Otsuchi R.			Kozuchi R.			Unosumai R.		
Date (Daily mean air temperature /daily precipitation)	$\delta^{18}O$ (‰)	δD (‰)	d-excess (‰)	$\delta^{18}O$ (‰)	δD (‰)	d-excess (‰)	$\delta^{18}O$ (‰)	δD (‰)	d-excess (‰)
2014/9/11 (19.7°C / 2.0 mm)	−9.0	−59.7	12.5	−9.2	−59.4	14.2	−9.4	−59.1	16.1
2014/10/5 (13.5°C / 0.5 mm)	−9.1	−59.3	13.8	−9.2	−59.1	14.4	−9.2	−59.5	14.2
2014/10/21 (16.9°C / 16.0 mm)	−9.2	−60.0	13.6	−9.2	−59.4	14.2	−9.3	−60.3	13.8
2014/12/17 (0.7°C / 0.0 mm)	−9.2	−60.1	13.3	−9.5	−60.8	15.2	−9.6	−61.5	15.3
2015/3/18 (8.6°C / 0.0 mm)	−9.3	−60.3	14.1	−9.4	−60.4	14.7	−9.4	−61.1	14.2
2015/5/12 (14.9°C / 27.5 mm)	−9.2	−60.8	12.8	−9.4	−60.2	14.8	−9.5	−61.3	14.4
2015/6/16 (18.2°C / 0.0 mm)	−9.6	−59.9	16.9	−9.5	−59.1	16.9	−9.6	−60.5	16.3
2015/7/1 (18.3°C / 5.5 mm)	−9.4	−58.4	16.5	−9.5	−58.3	17.4	−9.4	−59.1	16.5
2015/7/17 (18.7°C / 1.0 mm)	−9.4	−59.6	15.8	−9.5	−59.0	16.9	−9.7	−59.8	17.5
2015/9/15 (17.6°C / 0.0 mm)	−9.3	−57.2	17.1	−9.2	−56.0	17.4	−9.3	−56.9	17.6
Average	−9.3	−59.5	14.6	−9.3	−59.2	15.6	−9.4	−59.9	15.6
SD	0.2	1.0	1.8	0.1	1.4	1.4	0.2	1.4	1.4

Seawater	Hourai-jima		
Date	Salinity	$\delta^{18}O$ (‰)	δD (‰)
2014/9/11	31.7	−0.5	−4.4
2014/10/5	34.4	0.2	1.5
2014/10/21	25.2	−2.3	−14.4
2014/12/18	34.4	0.3	3.0
2015/3/18	31.2	−1.0	−5.1
2015/5/12	32.6	0.0	0.4
2015/6/16	32.7	−0.5	−0.5
2015/7/1	23.0	−3.0	−16.7
2015/7/17	30.5	−0.9	−4.1
2015/9/15	27.8	−1.6	−7.9
Average	30.3	−0.9	−4.8
SD	3.9	1.1	6.6

Average δD values of the three rivers were −59.5‰ (range −60.8‰ to −57.2‰), −59.2‰ (range −60.8‰ to −56.0‰), and −59.9‰ (range −61.5‰ to −56.9‰), respectively. These values are also in good agreement, except for data collected on 15 September 2015, which is slightly higher than the other values (Fig. 2b). Deuterium excess (d-excess, d=8δD − $\delta^{18}O$; Dansgaard 1964) provides information on kinetic isotope fractionation during phase changes of water. Average d-excess values of the three rivers are 14.6‰ (range 12.5‰ to 17.1‰), 15.6‰ (range 14.2‰ to 17.4‰), and 15.6‰ (range 13.8‰ to 17.6‰), respectively. These are generally in good agreement (Fig. 2c). In a cross-plot of $\delta^{18}O$ (horizontal axis) versus δD (vertical axis) (Fig. 2d), all data plot above the global meteoric water line (Dansgaard 1964), meaning d-excess values are always positive.

Seawater

Seawater salinity varied from 23.0 to 34.4 (Fig. 3a). $\delta^{18}O$ and δD varied from −3.0‰ to 0.3‰ and −16.7‰ to 3.0‰, respectively (Fig. 3b, c). Salinity lower than 28 was observed on 2014/10/21, 2015/7/1, and 2015/9/15, which was equivalent to the day when rain heavier than 50 mm/24 h occurred within 8 days (Fig. 3). There were distinct positive linear relationships between δD and salinity (Fig. 4a), and $\delta^{18}O$ and salinity of the seawater samples (Fig. 4b). Linear least-squares regression (with 95% confidence level) gives the following fits:

$$\delta D=(1.68\pm0.11)\ S-(55.81\pm3.27) \quad (r=0.99,\ p<0.0001,\ n=10)$$

$$\delta^{18}O=(0.28\pm0.01)\ S-(9.31\pm0.41) \quad (r=0.98,\ p<0.0001,\ n=10)$$

The δD and $\delta^{18}O$ values of the river water samples fall within the 95% confidence envelope of the δD–salinity and the $\delta^{18}O$–salinity relationships (Figs. 4a, b).

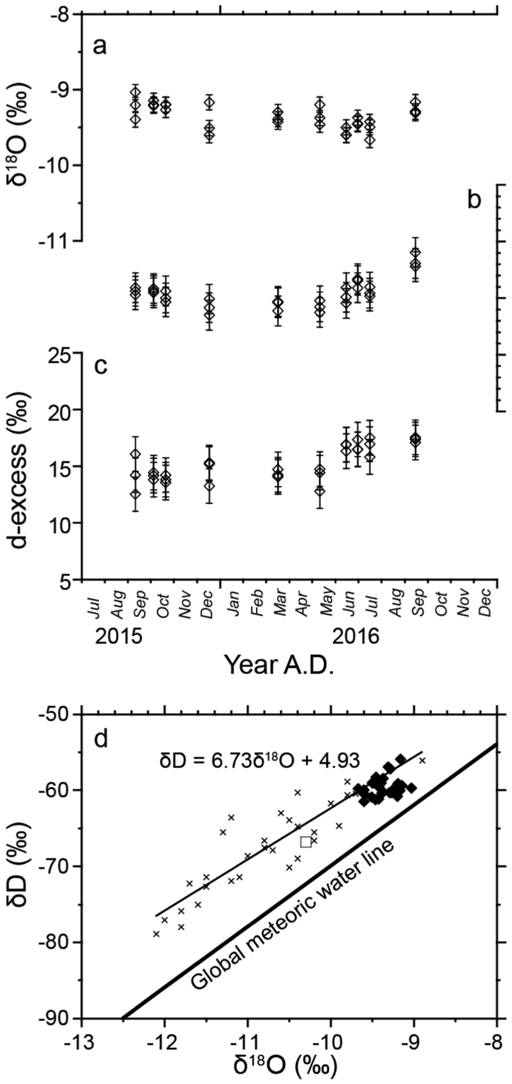

Fig. 2. **(a–c)** δ^{18}O, δD, and d-excess values of water from the Otsuchi, Kozuchi and Unosumai rivers collected over one year from September 2014 to September 2015. Error bars are 1 standard deviation. **(d)** Cross-plot of δD versus δ^{18}O. Diamonds indicate data of the present study, and crosses and a rectangle indicate data from previous studies that measured stream water in the Iwate region (Yabusaki and Shimano 2009 and Katsuyama et al. 2015, respectively). Also shown are the Global Meteoric Water Line (Dansgaard 1964; thick line) and a least-squares linear regression of stream water in the Iwate region (Katsuyama et al. 2015; thin line).

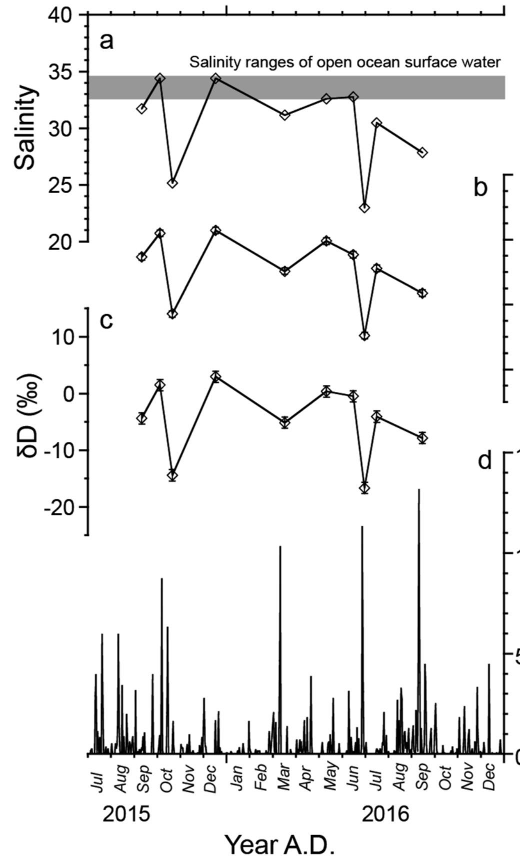

Fig. 3. **(a)** Salinity of seawater collected at Hourai-jima over one year from September 2014 to September 2015. Gray bar represents salinity ranges of the open ocean water (± 2 SD from the average since January 1971 to September 2011), which is calculated from monthly observations at 0 m depth offshore from Ozaki (39°15'N, 141°59'E), located 15 km south–southeast from Otsuchi Bay, using the vessel Iwate-maru (available at http://www2.pref.iwate.jp). **(b, c)** Same as a, but for δ^{18}O and δD values. Error bars are 1 SD. **(d)** Daily precipitation observed at Otsuchi (Data from Japan Meteorological Agency, available at http://www.data.jma.go.jp).

Discussion

No statistically significant differences in δ^{18}O and δD values were observed between the three rivers ($p > 0.05$, ANOVA). Similarly, no clear seasonality was seen in δ^{18}O and δD. Therefore, the freshwater entering Otsuchi Bay can be viewed as a single water mass.

The calculated d-excess values (12.5–17.6‰) of the three rivers are consistent with values reported previously for stream waters in the Iwate region (13.8–26.0‰) (Yabusaki and Shimano 2009, Katsuyama et al. 2015) (Figs. 1a and 2d). The new data of the present study are in good agreement with, and plot on the higher end of, the least-squares linear regression line reported by Katsuyama et al. (2015): δD=6.73 δ^{18}O+4.93; r=1.00, p<0.00001, n=52 (cross symbols in Fig. 2d). Data of Katsuyama et al. (2015) has large range of variability, which is likely a result of wide geographical range of sampling points. The likely reason why our δ^{18}O and δD data have relatively high values is proximity to the ocean, since δ^{18}O and δD of raindrop becomes lower as the water vapor is transported to inland, high altitude areas (Katsuyama et al. 2015). It is well established that d-excess values of precipitation on the Pacific coast of Japan vary seasonally by ～20‰ (Tanoue and Ichiyanagi 2016). Despite this, no clear seasonality is seen in the d-excess values of the three rivers monitored in this study between September 2014 and September 2015 (Table 1; Fig. 2c). This finding indicates that ground-

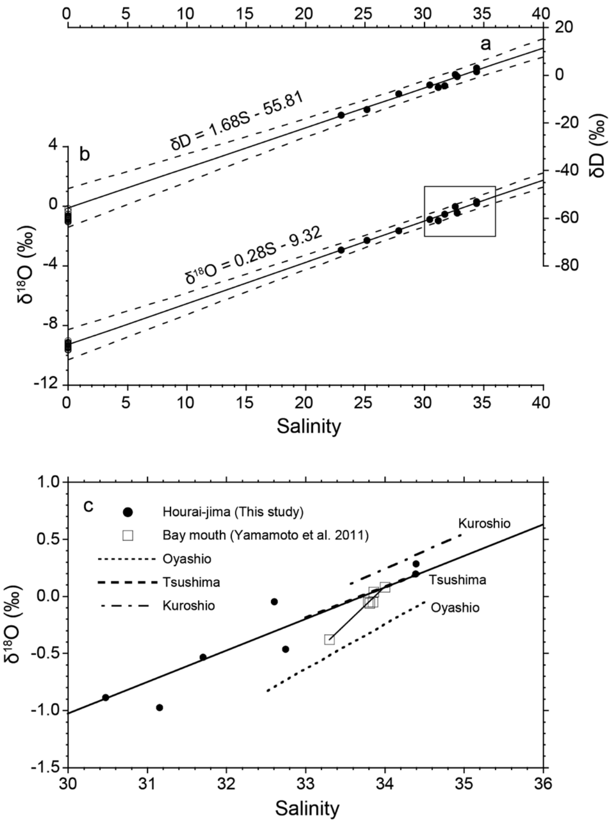

Fig. 4. Relationship between **(a)** δD values and salinity, and **(b)** δ^{18}O values and salinity, of freshwater (open circles) and seawater (closed circles) samples collected around Otsuchi Bay. Solid lines are least-squares linear regressions of each variable of seawater sample. The dotted lines delineate the 95% confidence bands of the regression. **(c)** Enlarged view of b, also showing additional data and the least-squares linear regression line of Yamamoto et al. (2011). The additional data represent bottom water collected at 70 m depth at the mouth of Otsuchi Bay. Also shown are the δ^{18}O–S relationships of the Kuroshio Current (Abe et al. 2009), the Tsushima Current (Kodaira et al. 2016), and the Oyashio Current (Yamamoto et al. 2001) as representative data of open ocean water that influences the bay.

water reservoirs in the catchment areas of the three rivers are large enough to smooth the seasonality of d-excess observed in rainfall. Such evidence of smoothing has been reported in other stream waters in the Iwate region (Katsuyama et al. 2015, Yabusaki and Shimano 2009) (Fig. 1a).

The obtained δ^{18}O–S equation can be applied to the salinity range 0–34.4, which covers the observed salinity in Otsuchi Bay and the precision of salinity estimation is less than 7 at the 95% confidence limit given uncertainties derived from δ^{18}O measurements and regression (Fig. 4b). In a previous study, Yamamoto et al. (2011) obtained the following δ^{18}O–S relationship based on water sampling at the mouth of Otsuchi Bay: δ^{18}O=0.66 S − 22.46. The newly obtained relationship presented in the current study has a shallower slope than that of Yamamoto et al. (2011) (Fig. 4c). Offshore of Otsuchi Bay, three ocean water masses mix in a complex way and have different δ^{18}O–S relationships to each other. These are the Kuroshio Current (δ^{18}O=0.31 S − 10.3; Abe et al. 2009), the Oyashio Current (δ^{18}O=0.39 S − 13.56; Yamamoto et al. 2001), and the Tsugaru Warm Current that originates from the Tsushima Current (δ^{18}O=0.27 S − 9.1; Kodaira et al. 2016) (Fig. 4c). It follows that the δ^{18}O–S relationship of open ocean water adjacent to Otsuchi Bay can vary in accordance with the mixing ratio of the three currents, and may not be regarded as a single end-member. However, at least for the surface seawater, this variation in the seawater end-member has relatively smaller influence on the δ^{18}O–S relationship within Otsuchi Bay compared with the freshwater end-member. Thus, as a first-order approximation, mixing of the surface seawater in Otsuchi Bay can be explained by a simple two end-member mixing model, with salty open ocean water mixing with fresh water from the rivers. As we did not collect subsurface water, vertical profile of the δ^{18}O–S relationship should be evaluated further in future works. It is also worth noting that the δ^{18}O–S relationship presented by Yamamoto et al. (2011) has a much steeper slope than the others (Fig. 4c). This is perhaps due to the limited number of samples and the narrow salinity range.

In general, δ^{18}O values of biogenic calcium carbonate vary as a function of water temperature and the δ^{18}O value of the water in which the calcifiers live (e.g., Grossmann and Ku 1986). Therefore, if the δ^{18}O–S relationship of a region is known, and if we can measure past water temperature independently, we can reconstruct the past δ^{18}O of the water, and thus past salinity using the δ^{18}O–S relationship. The new δ^{18}O–S relationship presented in this study is a valuable tool that can be used to reconstruct past salinity from calcifying organisms living in Otsuchi Bay, such as bivalves and brachiopods, and can also be used to reconstruct the routes of fish migration from fish otoliths, as this equation covers the full range of salinity observed in the bay. In particular, bivalve shells of Stimpson's hard clam (*Mercenaria stimpsoni*), found in Otsuchi Bay as well as in Funakoshi Bay to the north of Otsuchi Bay (Fig. 1b), are of great importance because this animal lives for nearly 100 years and is widely distributed on the sandy seafloors of these bays (Seike et al. 2013, Kubota et al. 2017, Shirai et al. 2018).

We observed no statistically significant differences in the oxygen and hydrogen isotopes, or d-excess values of water flowing into Otsuchi Bay from the Otsuchi, Kozuchi and Unosumai rivers. We also found no distinct seasonality in the isotopic characteristics of the fresh water entering the bay. Clear linear relationships were observed between δ^{18}O and salinity, and δD and salinity, indicating that the seawater δ^{18}O–S relationship in Otsuchi Bay is governed by simple mixing between open ocean water and river inflow. We also show that d-excess values of river water agree well with those of other stream waters in the Iwate region. We have established for the first time a seawater δ^{18}O–S relationship for

Otsuchi Bay that is applicable over a wide salinity range, from 0 to 34.4. Such a relationship will enable past salinity to be reconstructed on seasonal to decadal timescales, and will yield a deeper understanding of the migratory behavior of fish, using biogenic hard parts that consist of calcium carbonate.

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