Unexpected source of Fukushima-derived radiocesium to the coastal ocean of Japan

Virginie Sanial, Ken O. Buesseler, Matthew A. Charette, and Seiya Nagao

There are 440 operational nuclear reactors in the world, with approximately one-half situated along the coastline. This includes the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), which experienced multiple reactor meltdowns in March 2011 followed by the release of radioactivity to the marine environment. While surface inputs to the ocean via atmospheric deposition and rivers are usually well monitored after a nuclear accident, no study has focused on subterranean pathways. During our study period, we found the highest cesium-137 (137Cs) levels (up to 23,000 Bq·m⁻³) outside of the FDNPP site not in the ocean, rivers, or potable groundwater, but in groundwater beneath sand beaches over tens of kilometers away from the FDNPP. Here, we present evidence of a previously unknown, ongoing source of Fukushima-derived 137Cs to the coastal ocean. We postulate that these beach sands were contaminated in 2011 through wave- and tide-driven exchange and sorption of highly radioactive Cs from seawater. Subsequent desorption of 137Cs and fluid exchange from the beach sands was quantified using naturally occurring radium isotopes. This estimated ocean 137Cs source (0.6 TBq·y⁻¹) is of similar magnitude as the ongoing releases of 137Cs from the FDNPP site for 2013–2016, as well as the input of Fukushima-derived dissolved 137Cs via rivers. Although this ongoing source is not at present a public health issue for Japan, the release of Cs of this type and scale needs to be considered in nuclear power plant monitoring and scenarios involving future accidents.

Fukushima Dai-ichi Nuclear Power Plant accident | cesium | submarine groundwater discharge | radioactivity | radium

On March 11, 2011, a 9.0-magnitude earthquake triggered a 15-m tsunami that inundated the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), causing power loss, explosions, and reactor meltdowns, releasing a significant quantity of radionuclides into the atmosphere (1–3). More than 80% of the atmospheric fallout occurred over the ocean, with the highest deposition in the nearshore marine environment (4). In addition, direct liquid discharge of contaminated cooling water flowed into the ocean, making the FDNPP disaster the largest accidental input of radionuclides to the ocean (5).

Cesium-137 is an abundant fission product of nuclear power generation and nuclear weapons testing, which when released to the environment persists for decades due to its long half-life (30.2 y). The largest releases of Fukushima-derived 137Cs took place within the first month of the accident. The ongoing sources to the ocean that are known include rivers and groundwater flow beneath the FDNPP, but these are by comparison more than 1,000 times smaller than the 2011 releases, although they have persisted nearly 6 y after the accident (4). Submarine groundwater discharge has been recognized as an important pathway for the transport of materials from the land to the ocean (6), yet this process has not been evaluated as an ongoing source of radionuclides to the coastal environment outside of the vicinity of the FDNPP.

Here, we present 137Cs activities measured in groundwater collected underneath beaches up to 100 km away from the FDNPP (Fig. 1A). Eight beaches were visited between 2013 and 2015, with a more intensive sampling survey conducted in 2016 at Yotsukura beach, 35 km south of the FDNPP. Additional fresh groundwater and river samples were collected in the vicinity of the sandy beaches.

Results and Discussion

Dissolved (≤0.45 μm) 137Cs activities in beach groundwater spanned three orders of magnitude, with a maximum value of 23,000 ± 460 Bq·m⁻³ (Fig. 1B and Table S1). For perspective, three of the Yotsukura beach groundwater samples were higher than the Japanese drinking water limit of 10,000 Bq·m⁻³, although no one is either exposed to, or drinks, these waters, and thus public health is not of primary concern here. Beach groundwater 137Cs activities were generally higher than in nearby seawater, rivers, natural spring, and groundwater wells used for irrigation. The 137Cs activity in seawater rapidly decreased after the accident (7), and for the period of our study, the median seawater 137Cs activity within 100 km of the coastline (excluding the FDNPP harbor) was 14 Bq·m⁻³ (Fig. S1). In freshwater, dissolved 137Cs activities ranged from below detection to 5.8 ± 0.2 Bq·m⁻³ (Table S1). Thus, the 137Cs activities in beach groundwater cannot be explained by conservative mixing between any known freshwater and seawater sources (8). It must therefore be sourced from 137Cs-enriched beach sands.

Four sand cores were collected on Yotsukura beach (Table S2). In sand located between the beach surface and 40 cm, the 137Cs activity was relatively constant at 17 ± 4 Bq·kg⁻¹. Below 40 cm, the 137Cs activity increased with depth, reaching maximum values of 700 ± 60 Bq·kg⁻¹ (Fig. S2). The 137Cs inventory in the longest core was 4.8 ± 0.6 × 10⁶ Bq·m⁻², and is a minimum estimate since we did not reach the bottom of the high activity layer. This is one order of magnitude lower than the 137Cs activity in the ocean at this location, which spans 10⁷ Bq·m⁻³. Beach sands are therefore likely the primary source of cesium-137 to coastal ocean waters.

Significance

Five years after the Fukushima Dai-ichi Nuclear Power Plant accident, the highest radiocesium (137Cs) activities outside of the power plant site were observed in brackish groundwater underneath sand beaches. We hypothesize that the radiocesium was deposited on mineral surfaces in the days and weeks after the accident through wave- and tide-driven exchange of seawater through the beach face. As seawater radiocesium concentrations decreased, this radiocesium reentered the ocean via submarine groundwater discharge, at a rate on par with direct discharge from the power plant and river runoff. This new, unanticipated pathway for the storage and release of radionuclides to the ocean should be taken into account in the management of coastal areas where nuclear power plants are situated.

Author contributions: K.O.B. and M.A.C. designed research; V.S., K.O.B., M.A.C., and S.N. performed research; V.S., K.O.B., M.A.C., and S.N. analyzed data; and V.S., K.O.B., M.A.C., and S.N. wrote the paper.

Conflict of interest statement: K.O.B. has served in a consulting capacity related to radio-nuclides in Japanese fisheries products.

This article is a PNAS Direct Submission. Free access online through the PNAS open access option.

© 2017 The Author(s). Published by the National Academy of Sciences, USA.
magnitude higher than the largest recorded marine sediment inventory \[0.73 \pm 0.02 \times 10^3 \text{ Bq m}^{-2}\]; offshore of the FDNPP (9), within a factor of 4 of terrestrial soil cores \[20 \times 10^5 \text{ Bq m}^{-2}\] from the restricted access area, and in excess of soils in the Yotsukura region \[10^7 \text{ Bq m}^{-2}\] (10). Furthermore, the deep enrichment of \(^{137}\text{Cs}\) is inconsistent with a Fukushima atmospheric fallout source as \(^{137}\text{Cs}\) would have likely been trapped in the upper layers of the sand horizon as demonstrated in land soils (10).

Consequently, we need to consider an alternative source to explain the high \(^{137}\text{Cs}\) activities deeper in both beach sand and groundwater. In the days–weeks following the reactor melt-downs, dissolved \(^{137}\text{Cs}\) activities reached \(>60 \times 10^6 \text{ Bq m}^{-3}\) in the ocean closest to the FDNPP (5). Numerical modeling demonstrated that a net southward flowing coastal current transported this highly radioactive seawater along the shoreline (11). We hypothesize that seawater intrusion, driven in part by waves and tides (12–15), led to the storage of \(^{137}\text{Cs}\) by adsorption onto beach sands. In the years since the accident, falling ocean \(^{137}\text{Cs}\) activities and similar groundwater–surface water exchange processes would have led to the reverse reaction (desorption) and the elevated beach groundwater \(^{137}\text{Cs}\) activities observed today (Fig. 2). The \(^{137}\text{Cs}\)-enriched groundwater is then available to reproduce the salinity gradient observed in beach groundwater. Seawater solutions were spiked with \(^{137}\text{Cs}\) standard to reproduce the concentration of \(1 \times 10^5 \text{ Bq m}^{-3}\) observed in April 2011 in the ocean off Iwasawa beach, 16 km south of the FDNPP (5). The experiments showed that beach sands have an adequate ion exchange capacity for this level of \(^{137}\text{Cs}\), with an average adsorption fraction of 99% regardless of the salinity, sediment grain size, or mineralogy (Table S3). Desorption experiments involved recirculating \(^{137}\text{Cs}\) free seawater through the beach sand. The desorption fraction ranged from 1.4% to 11.4%, with the highest values found at intermediate salinities except for one low salinity treatment with 22% desorbed \(^{137}\text{Cs}\) (Fig. S4). These desorption rates were higher than those observed from riverine particles (18) or estuarine sediments (19), but consistent with a well-known property of \(\text{Cs}\): decreasing solid–solution partitioning with increasing ionic strength (20). Therefore, beach sands appear to be capable of storing a large inventory of \(^{137}\text{Cs}\) at depth that over time may be remobilized by seawater intrusion into beach aquifers and released to the coastal environment via groundwater–surface water exchange processes.

In beach settings, seawater intrusion generally follows two pathways: (i) an upper saline plume in the intertidal zone set up by waves and tides and (ii) a saltwater wedge at depth, a function of the density difference between fresh groundwater and seawater (refs. 13 and 15 and Fig. 2). The sand cores and the groundwater samples were collected at depths less than 2 m, along the boundary of influence of a typical upper saline plume, which is characterized
by dynamic mixing and exchange with seawater on timescales of days to weeks (13). The lower $^{137}$Cs concentration in the shallow cores could thus be the result of more frequent flushing with seawater in comparison with the deeper sand layer, or less exposure to high $^{137}$Cs activity seawater in 2011. The highly heterogeneous distribution of $^{137}$Cs in beach groundwater is also supported by the complexity of upper saline plume dynamics (15) as well as variability in sand physical and biogeochemical properties as shown by the desorption experiments.

Regardless of the mechanisms controlling the concentration of $^{137}$Cs in groundwater, beach aquifers in close proximity to FDNPP must be a source of $^{137}$Cs to the ocean, which is supported by surf zone $^{137}$Cs activities that were higher than offshore seawater during the study period (Fig. 1B). To quantify the magnitude of this $^{137}$Cs source, we used parallel measurements of radium isotopes ($^{223}$Ra, 11.4 d; $^{224}$Ra, 3.66 d), which are a well-established tool for providing regional scale estimates of submarine groundwater discharge (21, 22). Radium isotopes are continuously produced in aquifer sediments by the decay of insoluble thorium parent isotopes. Ra has a similar geochemical behavior to Cs, and Ra activities were also significantly higher in brackish groundwater than freshwater or seawater (Fig. 5S). Like $^{137}$Cs, Ra activities in the surf zone were inversely correlated with tidal stage (Fig. 5S). These characteristics are why Ra isotopes can be used as tracers of the ocean input of $^{137}$Cs from beach groundwater.

A Ra isotope mass-balance model (21–24) for the surf zone was used to estimate the volume of ocean water that exchanges with the beach aquifer on a daily basis. In the model, we assume that submarine groundwater discharge is the only source of Ra to the surf zone and that this source is balanced by Ra loss due to decay and mixing. The model was solved independently for the two Ra isotopes, which yielded a range of flux estimates between 0.07 and 0.51 m$^3$ m$^{-2}$ d$^{-1}$ for multiple beaches (Table S4).

To scale this estimate, we must make assumptions about the area over which the exchange of $^{137}$Cs-rich beach groundwater is occurring. The highest FDNPP $^{137}$Cs-contaminated marine sediments extend along 180 km of coastline (9). However, only ~45% of the coastline is covered by sandy beaches (80 km). Assuming groundwater–surface water exchange occurs over a (shore perpendicular) intertidal zone width of 50 m (calculated from beach slope and tidal range), the exchange of water was estimated to be on the order of 3.0–20 × 10$^3$ m$^3$ d$^{-1}$ (mean, 9.9 × 10$^3$ m$^3$ d$^{-1}$). Using the statistical mean of $^{137}$Cs activity in beach groundwater, 1.520 ± 0.570 Bq m$^{-3}$ (Fig. S6) and assuming a constant exchange of water over the period 2013–2016, we estimate that the amount of $^{137}$Cs delivered by submarine groundwater discharge along the Japanese coastline is on the order of 0.2–1.1 TBq y$^{-1}$, with an average of 0.6 TBq y$^{-1}$ (T = 10$^3$).

Over the course of 3 years, the main isotope of Cs carried by submarine groundwater discharge to the ocean from the main island of Japan (Honshu) has been estimated at 44.5 × 10$^3$ m$^3$ d$^{-1}$ km$^{-1}$ (25), which is equivalent to 3.5 × 10$^6$ m$^3$ d$^{-1}$ when scaled to our 80-km shoreline length. The $^{137}$Cs activities were relatively low in terrestrial groundwater, 2.3 Bq m$^{-3}$ on average in irrigation wells and in the natural spring, likely due to the strong affinity of Cs for clay particles in low ionic strength fluids, which limits its mobility in most inland freshwater aquifers (20). We estimate the amount of $^{137}$Cs carried to the ocean by terrestrial groundwater to be on the order of 3 × 10$^3$ TBq y$^{-1}$, which is <1% of the flux of $^{137}$Cs from brackish groundwater discharge. Thus, outside of the FDNPP site, the beach groundwater source dominates the flux of $^{137}$Cs released to the coastal ocean via submarine groundwater discharge.

In comparison with other sources, the ongoing releases of $^{137}$Cs at the FDNPP harbor were estimated to be 3 TBq y$^{-1}$ for the summer of 2012 (26). Harbor $^{137}$Cs activities decreased by a factor of 5 between 2013 and 2016 (27) (Fig. S1). Therefore, assuming that the flushing rate of the harbor has remained constant, the present-day FDNPP harbor flux should be ~0.6 TBq y$^{-1}$. Another ongoing source of FDNPP-derived $^{137}$Cs is from rivers, with release estimates for total $^{137}$Cs ranging from 2 to 12 TBq y$^{-1}$ (28–31). Typhoons and heavy rain events have been shown to increase the river runoff flux; the $^{137}$Cs input from this source is largely in the particulate phase (32), of which only a small amount is capable of entering the dissolved phase via desorption in the estuarine mixing zone (19). Thus, considering that ~90% of the Cs is irreversibly bound to riverine suspended sediments (19), the riverine flux of dissolved $^{137}$Cs is only ~0.2–1.2 TBq y$^{-1}$. Hence, the ocean input of $^{137}$Cs from groundwater below the sandy beaches is similar in magnitude as the other two major sources, namely ongoing FDNPP and dissolved river sources. Similar to projected decreases in $^{137}$Cs carried by runoff from land (30) and decreasing concentrations in the harbor at the FDNPP, the concentrations of $^{137}$Cs in sand will likely diminish over time due to desorption, and thereby the beach source to the ocean is expected to become depleted.

This study demonstrates that, aside from the aquifer beneath the FDNPP, the highest recorded present-day activities of $^{137}$Cs in the aqueous environment in Japan are associated with brackish groundwater underneath beaches. This finding suggests that the beach sands served as a reservoir for $^{137}$Cs, which is subsequently released via submarine groundwater discharge to the ocean. Using Ra isotopes, we were able to make an estimate of the magnitude of this flux and found that it is similar to other ongoing sources, including export from the FDNPP harbor. This unexpected and ongoing $^{137}$Cs source requires further investigation, in particular more systematic sampling in space and time given the variability in our groundwater and sand $^{137}$Cs observations. The implications of our study extend well beyond the FDNPP event to siting of all coastal NPPs, and this source will need to be considered when evaluating the fate of radionuclides in the ocean from both intentional (e.g., Sellafield) and unanticipated releases.
Methods

Sampling. Field trips for groundwater sampling were conducted in May 2013, September 2013, October 2014, October 2015, and November 2016. The river samples came from the Natsui, Iwasawa, Kido, Niido, Mano, and the Same River on 2015. The groundwater samples were collected at Nobiru, Nagahama, Funatsuke, Kawahata, Yotsukura, Karasuzaki, Iwasawa, and Nakoso beaches using peristaltic pumps and 1- to 2-m-long push point piezometers (M.H.E. Products). All water samples, including those from beaches, wells, rivers, and surf zones, were filtered through a 0.45-μm pore-sized filter. The water was then passed through a fiber impregnated with MnO₂ (“Mn-fiber”) to pre-concentrate radium (33) directly in the field. The water samples were then stored on dry ice, centrifuged to determine the Cs activities. Sediment profiles were obtained by digging pits on the beach and sampling the pit walls at 5-cm intervals.

Analysis. The beach sand samples were homogenized, dried, and counted on a gamma detector at the Low Level Radioactivity Laboratory in Kanazawa University (Table S2). The water samples were spiked with stable Cs and passed through columns filled with 5 mL of potassium hexacyanoferrate–polyacrylonitrile (KNiFC–PAN) ion-exchange resin (Czech Technical University, Prague, Czech Republic) for the extraction of radiocesium (34). Column Cs efficiency was determined by measuring the stable Cs in the filtrate via inductively coupled plasma mass spectrometry. Yields were on average 98.8 ± 1.4% (SD). The resin were then dried, transferred into vials, and counted on a well-type germanium gamma detector from Canberra Industries with a ceramic veto detector to discriminate the 137Cs activities using the gamma peaks at 661 keV and 134Cs using gamma peaks at 605 and 795 keV (35). All of the Cs activities were decay corrected to the sampling date. Gamma detectors were calibrated against international standards and an International Atomic Energy Agency Irish Sea water reference standard (IAEA-443 (36)). Although not discussed herein, 137Cs was detected in all samples (Table S1), indicative of a FDNNP source with a 1:1 release ratio due to the characteristics of the power plant such as reactor design, fuel cycle, or age (35). Therefore, once decay corrected to the accident date, the 137Cs activities are similar to the 137Cs activities, which is the reason that they are not discussed at length herein.

The Mn-fibers were rinsed with MQ water, partially dried, and counted on a radium delayed coincidence counting (RaDeCC) system (37) as soon as possible after collection to determine the short-lived radium isotopes (226Ra, 228Ra). Two additional counting sessions were conducted after 3 wk to determine the 226Ra supported by 228Th, and again after 3 mo to determine the 226Ra supported by 232Th. The RaDeCC system was calibrated with standards prepared in the same geometry as the Mn-fiber samples (37, 38).

Adsorption-Desorption Experiments Conducted in Beach Sand Samples. Filtered seawater (1-μm pore size) from Cape Cod Bay, Massachusetts, was used during the 137Cs adsorption and desorption experiments. The seawater was also processed through a KNiFC–PAN ion-exchange resin to remove the background radiation. The filtered seawater was de-ionized with Mill-Q water to produce four solutions of salinity 4, 10, 20, and 30. Sand samples were collected on two different beaches near the FDNNP (Table S3). The 137Cs activities in the sand were determined by gamma spectrometry.

The adsorption experiments were conducted on three sand samples (Table S3). Our experiment was designed to reproduce the infiltration of groundwater through the beach sand. Briefly, ~14 g of dry sand was placed into a column. Two 25-mL solutions, salinity 4 and 30, each with 137Cs activities of 1,000,000 Bq·L⁻¹ were gravity filtered through the sand columns twice with a filtration time of 15 min per sample. The filtrate was counted directly on a well-type germanium gamma detector to determine the fraction of 137Cs adsorbed by the sand, for the amount of water retained by the sand (~5 mL). The 137Cs adsorption fraction ranged from 97.9% to 100% with no correlation with salinity.

The desorption experiments were conducted on three sand samples (Fig. S4 and Table S3). Approximation system to quantify 14 g of dry sand was placed into a column. Four 1-L solutions of salinity 4, 10, 20, and 30 were recirculated for 4 h through the columns filled with sand using a peristaltic pump at <1 mL·min⁻¹ to reproduce groundwater infiltration. The desorption experiments were conducted in series on each sand, starting with the low-salinity solutions. The recovered solutions containing the Cs desorbed from the sand were filtered (0.45 μm) and the 137Cs preconcentrated onto KNiFC–PAN. The resin were then dried, transferred into vials, and counted on the gamma detector.

Beach Sand Properties. Grain size fractions and X-ray diffraction analysis were conducted on the sand samples used in the desorption experiments. The grain size fraction was relatively homogeneous throughout the deepest sand core (105 cm) with an average 50% of the grain sizes (d50) larger than 227 ± 14 μm (SD), which is classified as fine sand. Clay (<2 μm)- and silt (<60 μm)-size fractions in the beach sand were negligible, although X-ray diffraction analysis (Fig. S4 and Table S3) indicated that the largest Cs desorption was observed for the sand sample that had the greatest clay mineral content.

The distribution coefficient (Kd) for 137Cs in beach sand can be estimated from the desorption experiments using the following:

\[ K_d = \frac{V_m}{m} \left( 1 - e^{-\alpha_{GW} \cdot \tau} \right) / (M_m - \alpha), \]

where \( V_m \) is the volume of water used in the desorption experiment (1 L), \( M_m \) is the solid mass of sand (14 g), \( \alpha_{GW} \) is the desorption fraction. Considering a desorption fraction of 1.8-22% (Fig. S4), the \( K_d \) values of the beach sand are between 250 and 3,900 L·kg⁻¹, which agree with values previously observed for sediment–seawater interactions in Japanese coastal areas before the FDNPP accident (39). Applying these \( K_d \) values to the 137Cs concentrations measured in the sand cores collected at Yotsukura permits a prediction of the concentrations in groundwater from the following:

\[ \hat{C}_{GW} = \frac{C_{sand}}{K_d}. \]

where \( \hat{C}_{GW} \) is the dissolved 137Cs activity in groundwater and \( C_{sand} \) is the 137Cs activity in the sand. In the upper section, the mean 137Cs activity was 17 Bq·kg⁻¹, which leads to a \( \hat{C}_{GW} \) of 4–70 Bq·m⁻³. In the deeper section, the mean 137Cs activity was 700 Bq·kg⁻¹, which leads to a \( \hat{C}_{GW} \) of 180–2,800 Bq·m⁻³. The majority of the 137Cs activities measured in groundwater fall into the range of the \( \hat{C}_{GW} \) estimated from the \( K_d \) based on the desorption experiments (Fig. 1B), which supports our hypothesis that desorption from beach sand is the main mechanism releasing the 137Cs in groundwater.

Cesium-137 Flux from Beach Groundwater Using a Radium Mass-Balance Model. To estimate the flux of 137Cs-enriched water to the ocean through submarine groundwater discharge, we conducted a radium isotope mass-balance model. Submarine groundwater discharge is estimated for Iwasawa, Yotsukura, Karasuzaki, and Nakoso beaches, and was based on samples collected in 2015 and for Yotsukura beach in 2016 when both offshore and surf zone seawater Ra isotope data were available. The model assumes (i) that the system is at steady state, which is a fair assumption due to the fast production rate of short-lived radium isotopes (40); (ii) that submarine groundwater discharge is the primary source of Ra to the surf zone (23, 24), in other words, that diffusion from sediments is negligible, which we think is satisfied given the high-energy surf zone (41, 42); and (iii) that this input is balanced by the loss of Ra due to decay and mixing (ref. 22 and Eq. 3). For 226Ra or 228Ra, the model can be expressed as follows:

\[ F\cdot R_{GW} = F\cdot R_{SZ} (2) \cdot T, \]

where \( F\cdot R_{GW} \) is the groundwater flux of 226Ra or 228Ra, \( F\cdot R_{SZ} \) is the excess Ra inventory in the surf zone relative to the offshore water, \( \alpha \) is the decay constant of the Ra isotope considered, and \( T \) is the residence time of the water in the surf zone, which we assume to be equal to one tidal cycle (~0.5 d). The RaSZ were corrected for the average Ra activity in offshore water we measured from samples collected during two cruises conducted in 2015 (226Ra, 7.1 dpm·m⁻³; 228Ra, 80.5 dpm·m⁻³) and in 2016 (226Ra, 2.9 dpm·m⁻³; 228Ra, 48.1 dpm·m⁻³).

Based on Eq. 3, we calculated the area-normalized, volumetric flux of submarine groundwater discharge (VGW) independently with the two Ra isotopes following:

\[ V_GW = ( F\cdot R_{GW} x h) / R_{GW}, \]

where \( R_{GW} \) is the mean radium activity in groundwater and \( h \) is the tidal amplitude. The tide amplitude varied between ~1 and 179 cm during November 2016 at the Onahama tide gauge with an average of 90 ± 7 cm. We assumed that the tide amplitude, the residence time in the surf zone, as well as the RGW are the same for the different beaches. Our groundwater samples were well distributed across the gradient of salinity, thus, the Ra activities in groundwater are representative of fluids comprising submarine groundwater discharge (Fig. S5). In such a case, the mean Ra activity in groundwater is equal to the statistical mean (bootstrap method) used to determine the mean 137Cs activity in groundwater. Thus, we used the mean \( R_{GW} \) and \( R_{SZ} \) activities in groundwater for salinities between 5 and 30 (like for Cs) of 105 ± 16 and 1,990 ± 220 dpm·m⁻³, respectively, to quantify \( V_GW \). For the four beaches, the submarine groundwater discharge fell in the range of 0.07–0.51 m³·m⁻²·d⁻¹ (Table S4).
To scale these area-normalized $V_{GW}$ estimates to the affected region, we must calculate the area over which submarine groundwater discharge to the surf zone is occurring. As noted in Results and Discussion, we considered a coastal length of 180 km for which 80 km is covered by sand (estimated via Google Earth). During our sampling on November 15, 2016, the tide covered 90 m of the Yotsukura beach, which coincided with an extreme lunar tide of 150 cm predicted at Onahama station (Fig. 53). Based on this observation, we estimated the slope of the Yotsukura beach face at 0.96°. Assuming that the beach slope was constant over the coastline length, the width of the intertidal cell is thus ∼50 m for an average tide amplitude of 90 cm, and consistent with modeling studies of submarine groundwater discharge from beaches (15, 43). Note that this estimate could be a minimum width for the recirculation cell because submarine groundwater discharge occurs in the subtidal zone as well (44). This results in flux in the order of $3.0 \times 10^4 \text{ m}^2 \text{ d}^{-1}$ (using the min $V_{GW}$) and $20 \times 10^4 \text{ m}^2 \text{ d}^{-1}$ (using the max $V_{GW}$) with a mean flow of 9.9 m$^2$ d$^{-1}$.

Considering that the groundwater velocity flow was constant over the study period (2013–2016), the flux of $^{137}$Cs due to submarine groundwater discharge is determined as follows:


To scale these area-normalized $V_{GW}$ estimates to the affected region, we must calculate the area over which submarine groundwater discharge to the surf zone is occurring. As noted in Results and Discussion, we considered a coastal length of 180 km for which 80 km is covered by sand (estimated via Google Earth). During our sampling on November 15, 2016, the tide covered 90 m of the Yotsukura beach, which coincided with an extreme lunar tide of 150 cm predicted at Onahama station (Fig. 53). Based on this observation, we estimated the slope of the Yotsukura beach face at 0.96°. Assuming that the beach slope was constant over the coastline length, the width of the intertidal cell is thus ∼50 m for an average tide amplitude of 90 cm, and consistent with modeling studies of submarine groundwater discharge from beaches (15, 43). Note that this estimate could be a minimum width for the recirculation cell because submarine groundwater discharge occurs in the subtidal zone as well (44). This results in flux in the order of $3.0 \times 10^4 \text{ m}^2 \text{ d}^{-1}$ (using the min $V_{GW}$) and $20 \times 10^4 \text{ m}^2 \text{ d}^{-1}$ (using the max $V_{GW}$) with a mean flow of 9.9 m$^2$ d$^{-1}$.

Considering that the groundwater velocity flow was constant over the study period (2013–2016), the flux of $^{137}$Cs due to submarine groundwater discharge is determined as follows:


To scale these area-normalized $V_{GW}$ estimates to the affected region, we must calculate the area over which submarine groundwater discharge to the surf zone is occurring. As noted in Results and Discussion, we considered a coastal length of 180 km for which 80 km is covered by sand (estimated via Google Earth). During our sampling on November 15, 2016, the tide covered 90 m of the Yotsukura beach, which coincided with an extreme lunar tide of 150 cm predicted at Onahama station (Fig. 53). Based on this observation, we estimated the slope of the Yotsukura beach face at 0.96°. Assuming that the beach slope was constant over the coastline length, the width of the intertidal cell is thus ∼50 m for an average tide amplitude of 90 cm, and consistent with modeling studies of submarine groundwater discharge from beaches (15, 43). Note that this estimate could be a minimum width for the recirculation cell because submarine groundwater discharge occurs in the subtidal zone as well (44). This results in flux in the order of $3.0 \times 10^4 \text{ m}^2 \text{ d}^{-1}$ (using the min $V_{GW}$) and $20 \times 10^4 \text{ m}^2 \text{ d}^{-1}$ (using the max $V_{GW}$) with a mean flow of 9.9 m$^2$ d$^{-1}$.

Considering that the groundwater velocity flow was constant over the study period (2013–2016), the flux of $^{137}$Cs due to submarine groundwater discharge is determined as follows: