Doubled the carbon flux as a result of increased fresh submarine groundwater discharge caused by half the "normal" snowfall over the past 20 years along the mid-latitude coast of Japan

Saki Katazakai[1] and Jing Zhang[2*]

[1.] Graduate School of Science and Engineering, University of Toyama, Japan.

[2.] Faculty of Science, Academic Assembly, University of Toyama, Japan.

Corresponding author: *Jing Zhang (jzhang@sci.u-toyama.ac.jp)

Key Points:

- ✓ FSGD increased due to snowfall being reduced, leading to its diluted chemical composition and residence time being reduced by one-third;
- ✓ Material supply via FSGD and river runoff increased by 65% in carbon flux but decreased by up to 80% in nutrients;
- ✓ Increased direct carbon flux and decreased primary productivity via reduced nutrients into the coasts resulted in twice as much excess-DIC;

Abstract

Fresh Submarine Groundwater Discharge (FSGD) is an important pathway for the transport of water and materials from land to ocean, but changes in the transport may occur as snowfall decreases. This study was conducted on Japan's mid-latitude western coast where FSDG is a quarter of the total riverine discharge and snowfall has decreased by ~50% since the 1990s. The altitude of the FSGD recharge area in 2018 has shifted 100–150 m higher than that in 2000, and the water residence time has decreased from 4-15 to 3-11 years. The pH of the groundwater dropped by 0.5, its CO_2 (aq) concentration doubled, and nitrogen and phosphorus decreased by 30–40% and 70–80%, respectively. These changes in nutrients reduced primary productivity in coastal waters and doubled the excess dissolved inorganic carbon flux. Our evidence highlights the sensitivity of FSGD carbon flux to climate change and of the urgency of carbon-related FSGD research worldwide.

Plain Language Summary

Submarine Groundwater Discharge (SGD) is an important pathway of water and materials from land to ocean, and it is a common phenomenon found globally in coastal oceans. However, recent climate change is causing continuing impact on freshwater systems, including Fresh Submarine Groundwater Discharge (FSGD). Many coastal areas of mid-latitude Japan are influenced by the Asian monsoon and have experienced remarkable decreases in snowfall (about 50% since the 1980s). In the study area, our long-term research on the terrestrial waters suggests that FSGD discharge has increased by 30% since 1985 due to the decrease in snowfall and the increase in rainfall in winter. This study found that the decreased ratio of snowfall to total precipitation in mountain areas has increased the direct carbon discharge and reduced primary productivity in the coastal ocean as a result of decreased nutrient flux. This in turn has doubled the dissolved inorganic carbon flux form land into the coastal waters. Our findings present direct evidence of the sensitivity of carbon flux via FSGD to the decreased snowfall in the mid-latitude coasts, and encourages better estimation of global carbon budget in consideration of climate change, especially global snow cover melting.

1 Introduction

Submarine groundwater discharge (SGD) is recognized as an important pathway of water and materials into the global ocean. However, its volume and chemical composition can be vulnerable to ongoing global warming. Fresh Submarine Groundwater Discharge (FSGD), which accounts for as much as 10% of all SGD flux (Kwon et al., 2014), is especially sensitive to changes in the terrestrial freshwater system (Moosdorf, 2017; Luijendijk et al., 2020). In a recent modeling study, Betts et al. (2018) simulated the future global increase in river runoff into the ocean, and they predicted riverine discharge would increase by up to 50% by 2030. Zhou et al. (2019) warned that FSGD runoff would increase at high latitudes due to thawing permafrost. These studies show that the changes in freshwater runoff through FSGD and rivers cannot be ignored under the effects of global warming. On the western Japanese coasts influenced by the Asian monsoon, the annual snowfall has dropped by 50–60% since the 1980s (Japan Meteorological Agency, 2018) and the winter precipitation in this area has doubled since 1990 (Yasunaga and Tomochika, 2017). This means that the ratio of snowfall to total precipitation decreased, whereas the ratio of rainfall increased. Zhang et al. (2017) estimated the change in FSGD volume flowing into Toyama Bay, one of the three largest and deepest bays in the Japanese islands, located in central western Japan. Their evaluation indicates that FSGD has increased up to 30% since the 1980s, resulting from the decline in snowfall. This suggests that a notable shift in FSGD due to climate change is already progressing on a regional scale, and it is suspected that the water and materials via freshwater pathways may be transforming worldwide as a consequence of global warming.

SGD is considered an important carbon source in the global ocean carbon budget (Cai et al., 2003; Cole et al., 2007; Moore, 2010; Zhang and Mandal. 2012; Porubskya et al., 2014; Taniguchi et al., 2019). Szymczycha et al. (2014) found that SGD carries as much carbon load as river runoff globally. The existence of excess dissolved inorganic carbon (excess-DIC) that is not consumed in primary productivity has been reported in some coastal zones in the world (Atkins et al., 2013; Liu et al., 2014; Wang et al., 2018). Their research highlights a remarkable increase in the amount of carbon supplied by FSGD and rivers to the ocean. However, research on the changes in fresh SGD-derived carbon load is quite scant. In a sophisticated case study, Wang et al. (2014) showed that the effect of climate change on SGD will have a negative impact on coastal coral reefs in the future. Therefore, a better understanding of the increase in carbon supply via FSGD due to global warming will provide new insights for interpreting the effects of climate change on global ocean carbon budget and coastal marine ecosystems.

In this study, the selected research area is the well-watered Katakai River Alluvial Fan and its coast located in western central Japan, a region seasonally influenced by the Asian monsoon. In this area, FSGD is linked to the terrestrial freshwater system, having exactly the same hydrological and geochemical characteristics as those of the shallow groundwater (Zhang and Satake, 2003). A series of observation/monitoring studies on FSGD and groundwater have been conducted since 2000, including (1) technique development of SGD-discharge survey and detection (Tokunaga et al., 2001; Zhang et al., 2005), (2) observation of FSGD discharge (Koyama et al., 2005), (3) origin identification and chemical characterization using geochemical proxies (Nakaguchi et al., 2005; Kameyama et al., 2005), and (4) estimation of water and material supply via terrestrial freshwater systems using a box model (Hatta et al., 2005; Hatta and Zhang, 2013). These studies showed that the amount of FSGD in the study area was substantially higher than the global average of FSGD flux (Taniguchi et al., 2002; Burnnet et al., 2003; Moore, 2010). To reveal the temporal changes in material fluxes supplied from the land, five-year (2000–2003 and 2017–2018) geochemical observational results combined with a decadal groundwater monitoring data (2005–2015) are discussed in this study. We evaluate an impact of the decreased snowfall in the mid-latitude on the material flux via FSGD and river runoff, paying special attention to the carbon flux.

2 Materials and Methods

2.1 Study area setting and water sampling

The study area and the water sampling sites are shown in Figure 1. The main geological characteristics are gravel and sand sediments on the Katakai River Alluvial Fan (see Text S1). Water samples were collected from shallow groundwater, river water, and FSGD in two periods (2000–2003 and 2017–2018). All shallow groundwater samples were collected from the same sandy aquifer in 2001-2002 and 2017-2018. FSGD samples were collected by scuba divers in 2000-2003 and 2018. Water from the six major rivers (Figure 1b) accounts for up to 95% of total riverine discharge into the Toyama Bay (see Text S4). The groundwater monitoring data at D1 station (in February and March in during the years 2005–2015) were obtained from The Water Information Database, Uozu City, Toyama Prefecture (2018, Table S5).

2.2 Analysis methods

Water temperature, pH (Shindengen, KS-701), and EC (Horiba, B-173) were measured at the sites. Alkalinity was measured and calculated using the standard titration with HCl. The major cations and anions in the filtered samples were measured using a chromatograph (Metrohm 761 Compact IC). SiO_2 and PO_4 were analyzed with a UV-VIS spectrophotometer (SHIMADZU UVmini-1240). Analytical uncertainty for dissolved concentrations was ± 5% in all measurements. δD and $\delta^{18}O$ values for H_2O were measured using a wavelength-scanned cavity ring-down spectrometer (L2130-i, Picarro) and a mass spectrometer (PRISM; Micromass). The values were determined based on V-SMOW. The analytical precisions were ±0.07‰ for δD and ±0.05‰ for $\delta^{18}O$. Tritium counting was estimated using a low background liquid scintillation counter. Total analytical precision for tritium concentration is ±0.1 TU.

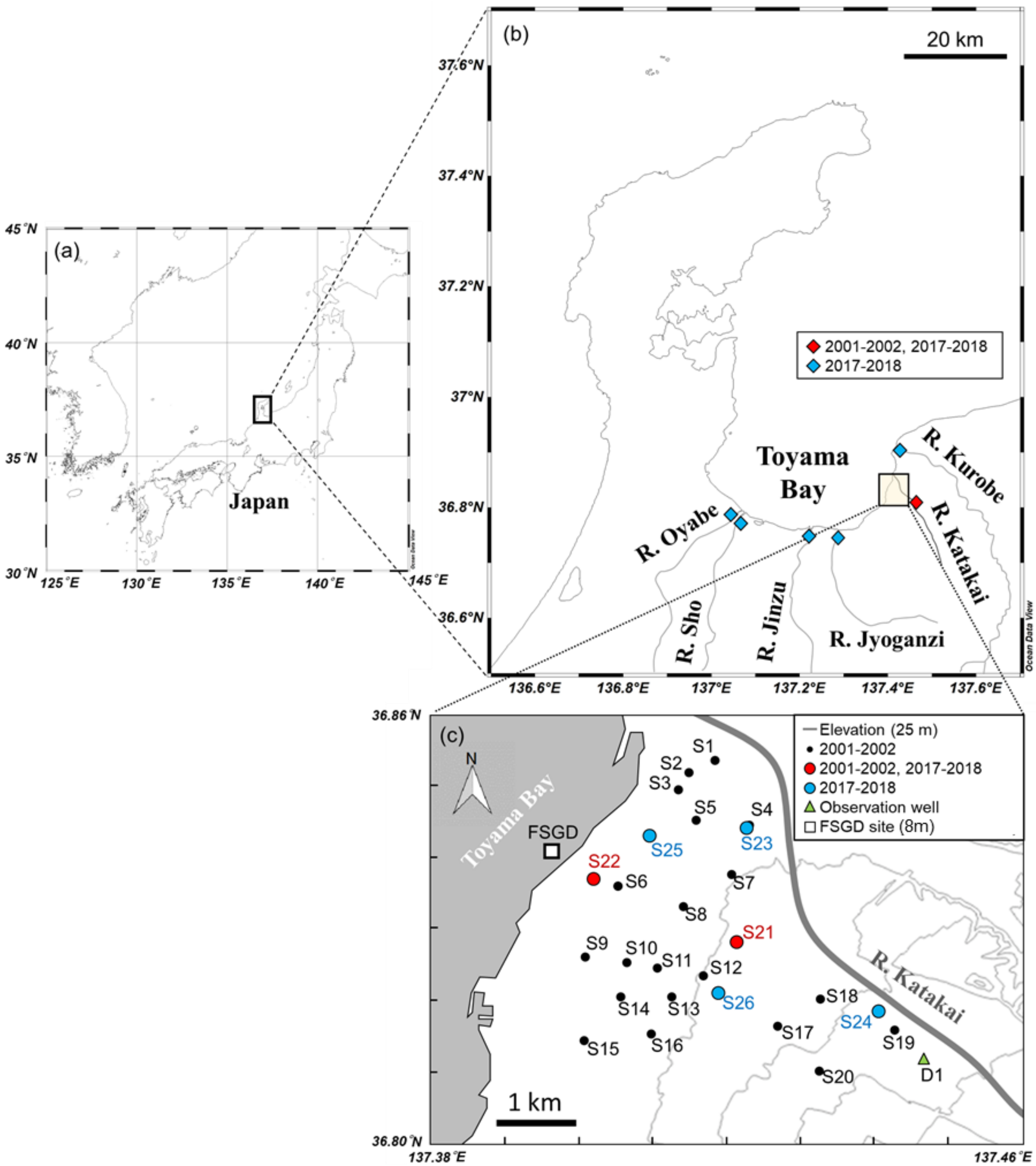

Figure 1. Sampling locations. Groundwater (dots) and river water (diamonds) samples were collected in 2001-2002 (black color), 2017-2018 (blue color), or both periods (red color). Green triangle indicates the observation well monitored in 2005–2015. White square shows the FSGD site.

3 Results and Discussion

3.1 Rise in recharge altitudes of FSGD and groundwater

All FSGD samples were freshwater, having EC values of 13.9 ± 5.4 mS/m in 2000–2003 and 11.7 ± 0.6 mS/m in 2018 (Table S3). Figure 2(a) shows the distribution of the average $\delta^{18}O$ value of groundwater and FSGD in the two periods (2000–2003 and 2017–2018). In both datasets, the $\delta^{18}O$ values of groundwater tend to be low at the stations in the center of the alluvial fan, which reflects the ancient river systems (Figure 2a). The $\delta^{18}O$ values of FSGD are also low and within the fluctuation range of the Katakai River (Tables S2 and S3). According to a well-known altitude effect, the $\delta^{18}O$ and δD values in precipitation and groundwater decrease toward higher altitudes of a water recharge area (e.g. Craig, 1961; Aravena et al., 1988). In this study area, Mizutani and Satake (1997) proposed the following empirical formulas between isotope compositions and elevation of the water recharge area (h, meters):

$$\delta D = -(0.0193 \pm 0.017)h - (4.84 \pm 2.3)\#(1)$$

$$\delta^{18}O = -(0.00236 \pm 0.00016)h - (8.68 \pm 0.22)\#(2)$$

Using these equations, it is revealed that the altitudes of FSGD recharge areas were about 800±50 m in 2000 (Zhang and Satake, 2003) and 850±50 m in 2018, and these altitudes were equivalent to the average catchment height of the Katakai River. This result shows that FSGD was recharged from high mountains and directly connected to the shallow groundwater system on the alluvial fan. In comparing the isotope compositions of the two periods (Figure 2b), most of our samples in 2017–2018 were lower than those of the early 2000s (Zhang and Satake, 2003; Suzuki and Zhang, 2003), and the altitudes of water recharge areas were higher by 100–150 m as well. This shift suggests that the water recharge altitudes throughout the entire shallow groundwater system have risen compared to 20 years ago.

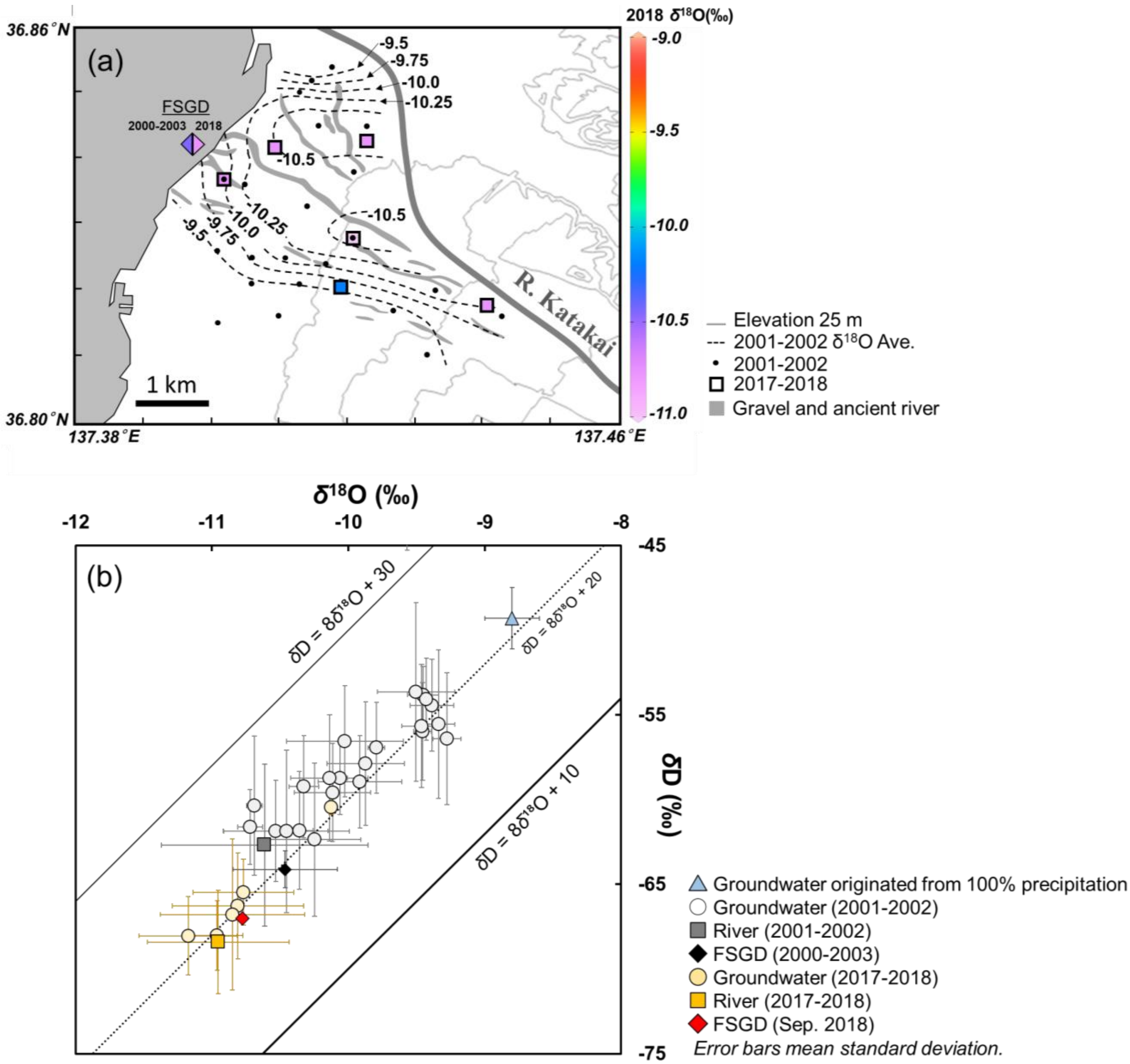

Figure 2. Average $\delta^{18}O$ and δD values in study area

(a) Distribution of $\delta^{18}O$ values in groundwater and FSGD. Dots show sampled stations in 2001–2002. The dashed contours indicate the distribution of average $\delta^{18}O$ values in 2001–2002. Squares show sampled stations in 2017–2018. The $\delta^{18}O$ values of FSGD are shown with a triangle in 2000–2003 and 2017–2018. The color bar represents the $\delta^{18}O$ value.

(b) Relationship between $\delta^{18}O$ and δD in all samples. Each plot shows an average value. Solid lines show standard deviation of each sample. A triangle shows the groundwater value recharged from 100% precipitation (Mizutani et al., 2001). Most of the water samples can be plotted along two meteoric water lines defined by Satake et al. (1983), where the Y-intercepts are 10 in summer and 30 in winter for this area's precipitation.

3.2 Diluted FSGD and its younger age

The decline in $\delta^{18}O$ and δD in 2018 described in section 3.1 can be explained by either (1) a change in the isotopic composition of precipitation or (2) a rise in the altitude of water recharge. Okakita et al. (2019) have analyzed the fluctuations in the δD and $\delta^{18}O$ values of precipitation, including those in our study area, since 1980, and they found that these isotopic values neither increased nor decreased significantly. Therefore, the decline in δD and $\delta^{18}O$ values possibly results from a rise in the water recharge altitude. If this is the case, the concentrations of dissolved components in 2018 should be lower than those in 2000–2003, since waters originating from the higher elevations of this area have lower concentrations of dissolved components (Yasuda et al., 1991).

Compared with the data of FSGD collected in summer 2000–2003 (Nakaguchi et al., 2005) (Figure S1), SiO_2 and NO_3 concentrations in 2018 decreased by 20±5% and 33±4%, respectively. These fluctuation ranges are nearly equivalent to the increase in FSGD and groundwater discharge (Zhang et al., 2017). In addition, NO_3 of the freshwater in this area is not affected by denitrification (Ohyama et al., 2012). These facts suggest that FSGD in this area is diluted as a result of the increase in water recharge from higher elevations.

Furthermore, in typical terrestrial water systems, the increase in water volume may lead to a shortened age of the average shallow groundwater (residence time) and reduce the efficiency of weathering, which would result in the dilution of dissolved components (Maher, 2011). SiO_2 concentration in groundwater can be used as a tracer for silicate weathering with time (Haines and Lloyd, 1985). Accordingly, the decline in SiO_2 concentration of FSGD in 2018 suggests that the time required for chemical weathering of FSGD has decreased and that the residence time of FSGD in this area might have become shorter. To verify our assumption, we compared the tritium concentrations and the age of FSGD in 2000 and 2018 (Table S4). The residence time of groundwater is determined by the following equation (cf. Zhang and Satake, 2003):

$$N = N_0 exp(-\lambda t)\#(3)$$

where λ is decay constant, N is tritium concentration in the groundwater, N_0 is the sum of tritium and helium-3 concentrations in the groundwater, and *t* is residence time. To estimate the residence time based on a piston flow model, we compared the tritium concentrations of our FSGD samples with those of the total precipitation of Japan (Saito et al., 2013; Environmental Radiation Database). The residence time in 2000, re-calculated by equation (3), was 4–15 years. However, the tritium concentration was 2.6 TU in the 2018 FSGD sample, and the residence time was estimated to be 3–11 years. This suggests that the water age of FSGD in Toyama Bay has become younger.

3.3 Decadal trend in compositions of the shallow groundwater connected to FSGD

To determine whether groundwater quality changed continuously during the period 2005–2015, the monitoring data at D1 station (Figure 1c) were used. The groundwater at D1 station, located at the top of the Katakai River Alluvial Fan, is connected to FSGD in the same terrestrial water system (Zhang and Satake, 2003). It is obvious that the values of Cl (Figure 3a), NO_3 (Figure 3b), and pH (Figure 3c) decreased at D1.

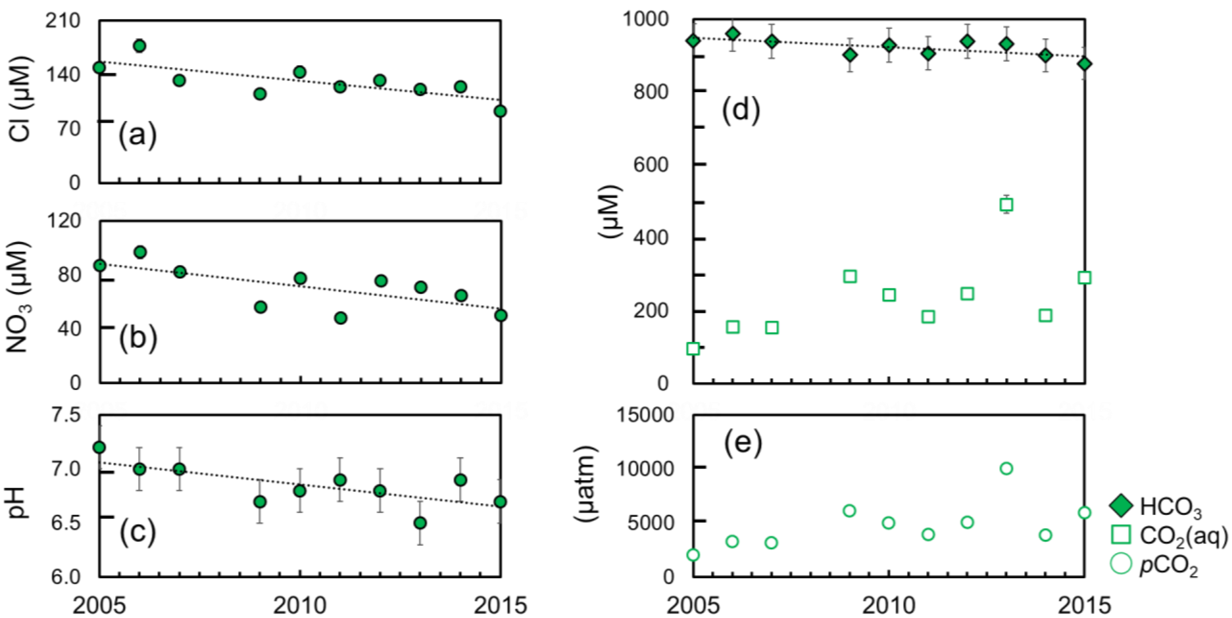

Figure 3. Ten year monitoring data of (a) Cl, (b) NO_3, (c) pH, (d) HCO_3^- and CO_2 (aq), and (e) pCO_2 at D1. The values of CO_2 (aq) and pCO_2 were calculated from the observation data. The measurement errors of Cl, NO_3, CO_2 (aq), and pCO_2 fall in each plot. Dashed lines indicate the linear regressions for each parameter (p<0.05). Details are listed given in Table S5.

Generally, a decrease in the pH value of groundwater is caused by influences such as (1) recharge of rainfall (Lång and Swedberg, 1990), (2) agricultural fertilizer usage (Beek et al., 1989), and (3) a dissolution of sediments (Preda and Cox, 2000). Factor (3) is neglected here due to the geological conditions in this area (see Text S1). It is well known that the nitrate concentration in groundwater increases due to the use of (2) agricultural fertilizer (Singh et al., 1995; Almasri and Kaluarachchi, 2004); however, NO_3 concentration at D1 (Figure 3b) declined for 10 consecutive years. Accordingly, the main reason for the decrease in pH can be attributed to the increase in (1) the recharge of rainfall. The decrease in groundwater pH with the increase in rainfall infiltration has also been reported in a different area that has the same geological conditions of this study's area (Zhou et al., 2015). In addition, nitrate and chloride concentrations decreased by about 40% from 2005 to 2015. These percentages are similar to the rate of decline in dissolved components of FSGD. Summarizing the discussion above, monitoring data at D1 indicate that the terrestrial water system connected to FSGD was diluted by the increase in rainfall infiltration from high mountains, resulting in the decreased pH.

The decrease in pH is also related to the carbonates in groundwater. In this area, bicarbonate (HCO_3^-) accounts for the majority of dissolved inorganic carbon (DIC) (Suzuki and Zhang, 2003), with very little dissolved organic carbon (DOC) (Nakaguchi et al., 2005). The lower pH value of groundwater can cause increased CO_2 (aq) in groundwater, resulting in higher carbon dioxide partial pressure (pCO_2). The concentrations of CO_3^{2-}, CO_2 (aq), and pCO_2 at D1 were calculated using the methods proposed by Lueker et al. (2000) and Weiss (1974) (see Text S2 and Text S3). Figure 3 (d) and Figure 3e show the values of HCO_3^-, CO_2 (aq), and pCO_2 at D1. It reveals that CO_2 (aq) concentrations in groundwater doubled from 2005 to 2015 and that pCO_2 also increased during the same period.

3.4 Estimating carbon and nutrient fluxes from the land and its impact on the coastal ocean

We calculated carbon and nutrient fluxes from the land to Toyama Bay via FSGD and rivers. Two characteristics of the fluxes are defined: V (volume) and C (concentration):

$$Flux_{FSGD} = V_{FSGD} \times C_{FSGD} \#(4)$$

$$Flux_{River} = V_{River} \times C_{River} \#(5)$$

where values for FSGD flux (V_{FSGD}) are taken from the water balance model (Zhang et al., 2017) with modification using the previous data of Hatta and Zhang (2013). The average nutrient and carbon concentrations in SGD and river water (C_{FSGD} and C_{River}), are based on the results of Suzuki and Zhang (2003), Nakaguchi et al. (2005), Tsujimoto (2009), Yanagi et al. (2019), and this study. Total riverine input (V_{River}) is taken from data of the Ministry of Land, Infrastructure and Transport, Government of Japan (http://www1.river.go.jp/). These details are provided from Table S6 to Table S9.

3.4.1 Increase in direct discharge of carbon into the coastal ocean over 20 years

Figure 4 shows the fluxes of DIC (the sum of CO_3^{2-}, HCO_3^-, and CO_2 (aq) concentrations) and nutrients (N and P) from the land in 2003 and 2018. Direct discharge of DIC via FSGD and river runoff increased by 65% in 15 years. It is known that the carbon input via groundwater increased in the wet season due to the increased rainfall infiltration and higher velocity of groundwater flow (Sadat-Noori et al., 2016). In this area, an increase in FSGD discharge and river flow (Zhang et al., 2017) and a shorter residence time in FSGD (this study) were observed due to the increase in the ratio of rainfall to total precipitation. Therefore, our data suggest that FSGD-derived DIC load increased due to climate change, especially the decline in snowfall. Furthermore, our calculations show that direct discharge of DIC via river runoff increased together with FSGD even if the error range were included.

3.4.2 Decrease in nutrient flux from land over 20 years

The nutrient supply via FSGD and river runoff has dropped dramatically since 2003: the nitrogen flux decreased by 30–40% and phosphorus flux decreased by 70–80% (Figure 4). The percentage of reduction in nitrogen corresponded with the increase in FSGD and river runoff volume reported by Zhang et al. (2017), but the phosphorus reduction was much higher. This is considered to have been caused by human activities on the land (e.g., enactment of new laws, land-use practices, and water treatment). As in many other regions of Japan, two trends have accelerated since the 1990s: the reduction of farmland (Ministry of Agriculture, Forestry and Fisheries, Japan) and the increased use of sewage treatment plants as technologies for removing phosphorus from wastewater (Kuba et al., 1996; Tsuneda et al., 2005). According to Nakaguchi et al. (2005), nitrogen and phosphorus in groundwater and river water in the area were mainly caused by the mineralization of organic matter in farmland soil. However, the decline in agricultural land in the area has been less than 5% since 1976 (Katazakai et al., 2019). Thus, the drop in phosphorus flux in our study was not mainly caused by decreased farmland. Regarding the dramatic reduction in phosphorus flux from land in Japan, there is related evidence in the Seto Inland Sea, one of the largest enclosed seas most seriously affected by oligotrophication in the world's coasts. Here, the phosphorus input from land has been halved in 1972–1994, whereas nitrogen has not changed much since 1972; this has been explained as a problem of the series of new Japanese laws aimed at limiting the nutrient discharge and improving water treatment technology (Yamamoto, 2003). Therefore, it is assumed that the drop in phosphorus (70–80%) in our study area is influenced by the changes in nutrient load due to human activities, in addition to the dilution of nutrients due to climate change (30%).

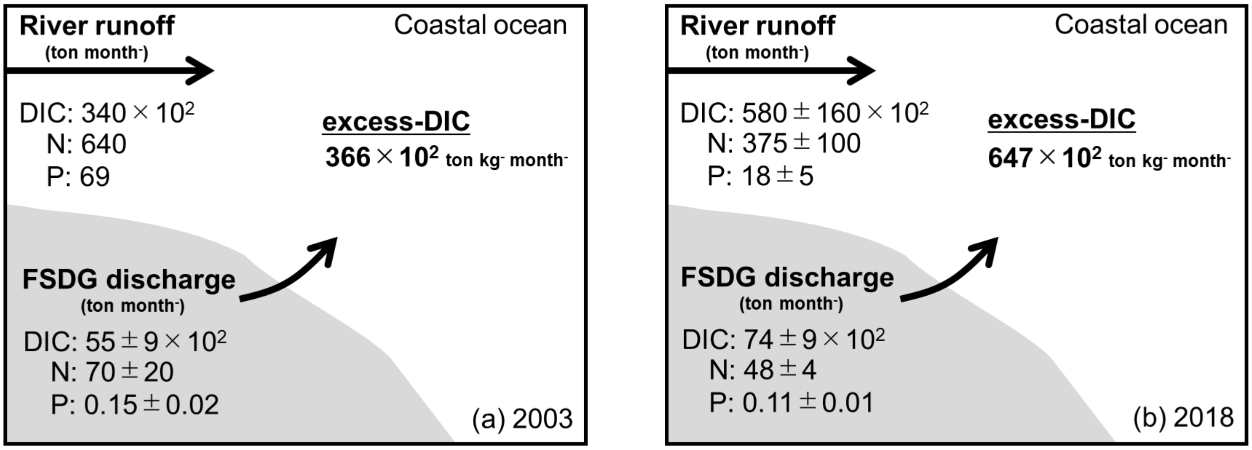

Figure 4. Carbon and nutrient fluxes from land to Toyama Bay via FSGD and river runoff in (a) 2003 and (b) 2018.

3.4. 3 Increase in carbon flux resulting from nutrients reduction and its impact on the coastal ocean

A decrease in nutrient input reduces the primary productivity of the coastal surface ocean, causing a decrease in the amount of carbon fixed by photosynthesis. This leads to an increase in excess-DIC in coastal surface waters. The amount of excess-DIC depends on the balance between carbon fixation in the primary productivity and DIC input from land (Borges and Abril, 2011; Maher et al., 2015). The primary productivity in the surface ocean is estimated using the Redfield ratio (C: N: P = 41.1: 7.2: 1, weight ratio; Redfield et al., 1963). Based on the Redfield ratio, the N/P ratios of our samples were 10 in 2003 and 22 in 2018. This suggests that Toyama Bay has had reductions in phosphorus over the past two decades, which is consistent with previous research (Hatta et al., 2005). Moreover, it is calculated that primary productivity with nutrient load from FSGD and river runoff in this area consumed 2,918±292 DIC ton/kg/month in 2003 and 740±250 DIC ton/kg/month in 2018. These amounts are much lower than the direct DIC discharge through FSGD and river runoff in both years. The excess-DIC in coastal waters was 366×10^2 ton/kg/month in 2003 (Figure 4a) and 646×10^2 ton/kg/month in 2018 (Figure 4b). This means that the excess-DIC in the coastal waters has doubled in 15 years. More importantly, the increase in excess-DIC is much larger than the increase in direct DIC flux. We believe this remarkable increase in excess-DIC was caused by the increasing direct DIC flux from the land, along with the decreasing carbon consumption in primary productivity due to the reduced supply of nutrients through FSGD and rivers. The greatest cause of increase in the area’s excess-DIC is assumed to be related to the decrease in the ratio of snowfall to total precipitation, followed by the changes in human activities.

DIC supply from land to oceans can be easily interpreted as having increased up to the present in many coastal zones globally. In the Northern Hemisphere (excluding the Arctic region), snow cover has been decreasing in all recorded seasons during the past four decades (1978–2015), and the rate of snow cover melting in summer and autumn is particularly serious (Hori et al., 2017). The ratio of snowfall to total precipitation has gone down over half a century (1949–2005) in many areas of the United States facing coastal zones (Feng and Hu, 2007). These observations show that the terrestrial water system can change in terms of the water quality and the material transfer. Consequently, it is possible that DIC supply from the land to global oceans is increasing in all coastal zones, as our observations indicate. Our findings provide a direct evidence, especially from the perspective of FSGD, of how sensitively the carbon budget responds to global climate change, and they demonstrate the importance and even urgency of carbon-related FSGD research worldwide.

Acknowledgments

The tritium concentration data is available at Environmental Radiation Database (https://search.kankyo-hoshano.go.jp/servlet/search.top). The data of land-use in Japan can be downloaded at Ministry of Agriculture, Forestry and Fisheries, Japan (https://www.maff.go.jp/j/tokei/kouhyou/nougyou_sansyutu/tyouki/). Riverine inflow dataset is available at Ministry of Land, Infrastructure and Transport, Government of Japan (http://www1.river.go.jp/). The authors would like to express their sincere thanks to K. Asai (Chikyu Kagaku Kenkyusho Inc.), members of the Uozu Fisheries Cooperative Association, and Uozu Aquarium for help in collecting FSGD samples. We are grateful to M. Suzuki (University of Toyama) and K. Sawada (Uozu City) for help in gathering data. We are also grateful to K. Asai for help in the analysis of tritium concentration. This work was supported by a Grant-in-Aid for Scientific Research (KAKENHI) (20H04319, 15H00973, and 19310007), a Sasakawa Scientific Research Grant from The Japan Science Society (2018-7040), the Environment Research and Technology Development Fund (S-13) of the Ministry of the Environment, Japan, the Support Project for Japan Seaology Promotion Organization in 2016-2018, ERAN's Collaborative Research Project (Y-19-24 and Y-20-27), the cooperative research program of the Institute of Nature and Environmental Technology, Kanazawa University (18043), Interdisciplinary Project on Environmental Transfer of Radionuclides (HF-18-10), and Joint Research Grant for the Environmental Isotope Study of Research Institute for Humanity and Nature.

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