Submarine Groundwater Discharge helps making nearshore waters heterotrophic

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Submarine groundwater discharge (SGD) is the submarine seepage of all fluids from coastal sediments into the overlying coastal seas. It has been well documented that the SGD may contribute a great deal of allochthonous nutrients to the coastlines1–9. Excessive supply of nutrients may lead to eutrophication, hence affecting the sustainability of the coastal environment10. SGD also contains excess carbon11–14. It is, however, less known how nutrients and carbon interact after the SGD enters the oceans.

Because the groundwater has been in contact with the sediments for a long period of time it is expected that some of the particulate organic matter in the sediments would have decomposed thus consuming dissolved oxygen (DO) but releasing dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC) along with nutrients. The partial pressure of CO2 (pCO2) would also increase. Part of the DOC would decompose, further increase DIC and pCO2. Some of the CaCO3 in the sediments might also dissolve thus increase the total alkalinity (TA). The groundwater is isolated from the atmosphere but when the groundwater enters the oceans it is expected that the high pCO2 in the SGD might make the receiving coastal water a CO2 source for the atmosphere. Yet, the nutrient supply from the SGD would enhance primary productivity in coastal waters, hence drawing down the pCO2 of surface waters. Whether the SGD would eventually lead to a carbon source or sink into the receiving coastal waters does not have an a priori answer.

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Recently the salinity and major ions such as Ca, Mg, K, Na, Cl and SO₄ in the submarine groundwater samples around Taiwan have been measured. However, nutrients and carbon in the SGD have never been reported in the SGD from this part of the world. In fact, only a handful of studies have reported nutrients and carbon in the SGD in China. In this study, SGD samples were collected from 20 locations around the subtropical island of Taiwan at 5,486 m of coastline. This compares with our freshwater SGD component of 4.84 μM; Table 1). The NO₃ in the SGD increases when the salinity decreases (Fig. 2b; p < 0.001). In fact, the average NO₃ concentration in the local surface seawater is higher than those generally found in waters surrounding Taiwan (2.5 ± 4.0 μM) with waters surrounding Taiwan having the lowest value (2.5 ± 0.4 μM). Although there are other sources of NO₃ such as riverine input and acid rain, the SGD has likely played a role. NO₂, NH₄ and N₂O are all reduced forms of NO₃ and the average NH₄ concentration (92.4 ± 1.8 μM) is much higher than that of the average local surface seawater (4.84 ± 5.08 μM; Table 1). The presence of NO₃ and NO₂, however, indicates that much, but not all, of the nitrogen released as a result of organic matter decomposition has been reduced to NH₄.

Table 1. Concentrations of parameters measured in the submarine groundwater and local surface seawater. *

| Parameter      | Submarine Groundwater | Local Surface Seawater |
|----------------|------------------------|-------------------------|
| **Range**      | **Mean ± std**         | **n**                   | **Range**      | **Mean ± std** | **n** |
| S              | 0.008–34.8            | 21.92 ± 11.43           | 278            | 18.2–36.8     | 32.5 ± 2.42 | 125  |
| DO (%)         | 8.3–109               | 67.6 ± 21.9             | 218            | 72.6–106      | 95 ± 6.47  | 106  |
| NO₂ (μM)       | <0.02–280             | 27.4 ± 54.4             | 231            | <0.02–24.4    | 4.84 ± 5.08 | 110  |
| NO₃ (μM)       | <0.02–46.3            | 1.82 ± 4.95             | 218            | <0.02–4.19    | 0.69 ± 0.76 | 109  |
| NH₄⁺ (μM)      | 0.14–3042             | 92.4 ± 387              | 131            | 0.35–1653     | 48.38 ± 205 | 84   |
| PO₄ (μM)       | <0.02–33.7            | 0.88 ± 2.44             | 225            | <0.05–2.70    | 0.55 ± 0.48 | 107  |
| SiO₂ (μM)      | 0.01–221              | 64.2 ± 58.3             | 224            | 0.37–147      | 10.8 ± 17.9 | 110  |
| N₂O (nM)       | 3.56–91               | 10.6 ± 14.7             | 51             | 3.64–70.9     | 8.7 ± 14.0  | 22   |
| CH₄ (nM)       | 1.05–3994             | 523.1 ± 1231            | 13             | 3.84–1493     | 240 ± 554 | 7    |
| DOC [μmol/kg]  | 24–527                | 114 ± 112               | 31             | 26–130        | 84 ± 27 | 9    |
| pH             | 6.53–8.73             | 7.81 ± 0.29             | 166            | 7.27–8.40     | 8.10 ± 0.14 | 97   |
| TA [μmol/kg]   | 594–8579              | 3438 ± 1417             | 134            | 1505–4760     | 2343 ± 358 | 87   |
| DIC [μmol/kg]  | 352–8675             | 3193 ± 1373             | 122            | 1246–4108     | 2040 ± 363 | 86   |
| pCO₂ [atm]     | 221–112455            | 4729 ± 13163            | 122            | 145–3732      | 477 ± 479 | 86   |

* Taken from Chen et al.15.
(10.8 ± 17.9 μM; Table 1). Several high values above 50 μM are found in the local surface seawater compared with the generally low value of <5 μM found in waters surrounding Taiwan21. This is an indication that phytoplankton uptake is not fast enough to consume the SiO₂ released by the SGD near its source.

In the reduced environment CH₄ is generated. We do not have sufficient CH₄ data (n = 13) to see a clear trend relative to the salinity but the average CH₄ concentration (523.1 ± 1,231 nM; Table 1) in the SGD is clearly higher than that in the local surface seawater (240 ± 554 nM; Table 1). Note the CH₄ concentrations in waters around Taiwan are around only 5 nM28. This indicates that the SGD inputs of CH₄ do not have sufficient time to oxidize or to be released to the atmosphere near their sources.

Decomposition of organic matter generally lowers pH in the aerobic environment29 such as found in our case so the average pH (7.81 ± 0.29) of the SGD is slightly lower than that of the local surface seawater (8.10 ± 0.14; Table 1; Fig. 3a), which is similar to the pH of the waters surrounding Taiwan30–33. As for TA and DIC their values increase with the dissolution of calcareous rocks and decomposition of organic matter and it is indeed what was found. The average TA and DIC in the SGD (3,438 ± 1,471 and 3,193 ± 1,373 μmol/kg, respectively) are significantly higher than those found in the local surface seawater (2,343 ± 358 and 2,040 ± 363 μmol/kg, respectively; Table 1) and higher values are found at lower salinities (p < 0.001 in both cases; Fig. 3b,c). These TA and DIC values are more than 1,000 μmol/kg higher than those found in waters near Taiwan33–35.

The SGD has a high average pCO₂ of 4,729 ± 13,163 μatm (Table 1; Fig. 3d) compared with the average of the local surface seawater (477 ± 479 μatm). The pCO₂ shows a weak negative (p < 0.05; Fig. 3d) correlation with salinity. These values are higher than the pCO₂ of surface waters found near Taiwan31,32,35,36. But, whether the surface seawater receiving the SGD is heterotrophic, i.e., whether the water is a source or sink of CO₂ depends on the balance between carbon-consuming primary production and the excess DIC supplied by the SGD. The average C/N and C/P ratios of particulate matter in NW Pacific marginal seas are 8.8 and 152, respectively36,37. Based on this stoichiometry the average amount of nitrogen and phosphorus supplied by the SGD in Taiwan for each kg of water may consume 1,070 μmol/kg and 133 μmol/kg DIC, respectively. These values are much lower than the average excess DIC supported by the SGD. That is to say, primary production supported by the nutrient input from the SGD is insufficient to compensate for the high DIC and pCO₂ supplied by the SGD. As a result, the SGD around Taiwan leads to a CO₂ source for the atmosphere. Similar situation applies to the Jiulong and Pearl River Estuaries. Finally, decomposition of DOC also releases CO₂. Indeed, the higher pCO₂ values in local surface seawaters (477 ± 479 μatm; Table 1) relative to the atmosphere (~400 μatm) support this conclusion. Similar conclusion has been reported for the Pearl River Estuary38 and elsewhere36,39.

![Sampling locations.](image1.png)

Figure 1. Sampling locations.
The number of DOC data is also small (n = 31) but there seems to be a trend showing high values at low salinities (p < 0.25; Fig. 3e). The average DOC in the submarine groundwater (114 ± 112 μM) is slightly higher than that in the local seawater (84 ± 27 μM; Table 1). The waters surrounding Taiwan generally have a DOC concentration below 75 μM40,41. Note Fig. 3e seems to indicate that the DOC is removed, hence becoming a source of nutrients and pCO2.

It is critical to point out that the C/N and C/P values of the SGD (Fig. 3f,g) are much higher than the Redfield Ratio. To re-iterate, the excess nutrients supplied by the SGD are insufficient to consume the excess carbon thus the SGD helps making the coastal waters heterotrophic.

Of note is that the SGD-derived DIC flux is greater than the TA flux in the Pearl River estuary, indicating that the SGD serves to reduce the CO2 buffering capacity of the local seawater29. Yet, submarine groundwaters around Taiwan the TA flux is slightly higher than the DIC flux. As a result, the SGD from Taiwan serves to increase slightly the CO2 buffering capacity of the local seawater. Even so, the high pCO2 and the high C/N and C/P ratios of Taiwan’s SGD makes it a contributor of heterotrophic nearshore waters.

The percentage saturation for aragonite (Fig. 3h) and calcite (Fig. 3i) reaches a mean value of four and six, respectively for the local surface seawater but are slightly lower in the SGD. There is no doubt that the higher TA, DIC and pCO2 of the SGD compared to the local seawater is due to the dissolution of calcareous rocks and decomposition of organic matter in the groundwater. The decomposition of DOC must also be at play hence increasing pCO2. Since the submarine groundwaters do not become anoxic sulfate reduction probably has not occurred to a great extent. Figure 4 shows ΔHCO3 plotted vs ΔCa (local seawater is taken to be with HCO3 = 2.3 mM and Ca = 10.3 mM at a salinity of 35; Ca data taken from Chen et al.)16. The samples falling around the HCO3/Ca = 2 line reflect the dissolution of CaCO3. Much of the data shows an excess of HCO3 and Ca but the pattern is not obvious.

Figure 5 shows the saturation state of aragonite and calcite plotted vs pH. Lower saturation state corresponds to lower pH, indicating that the decomposition of organic matter leads to the dissolution of calcareous rocks. The
end result, however, is that the submarine groundwater is mostly highly super saturated, especially those with a pH above 7.5. The saturation state of aragonite and calcite even reach 12 and 20, respectively.

Fluxes of nutrients and carbon. Since the properties of groundwater are not expected to show much seasonal variation as compared to the flux (e.g. Szymczycha et al.39) the above conclusions represent reasonable averages. Based on the rudimentary SGD flux value reported by Chen et al.15 the annual amount of nitrogen, phosphorus, silicate, TA and DIC export due to the SGD around Taiwan are $1.18 \pm 0.83 \times 10^9$, $9.3 \pm 6.5 \times 10^6$, $0.68 \pm 0.48 \times 10^9$, $3.43 \pm 2.4 \times 10^{10}$ and $3.17 \pm 2.22 \times 10^{10}$ mol, respectively (Table 2). Based on the river flow (http://gweb.wra.gov.tw/wrweb/) and the N, P data (http://wgshow.epa.gov.tw/) of the 25 largest rivers in Taiwan the total N and P fluxes are $1.12 \times 10^{10}$ and $0.12 \times 10^{10}$ mol/a, respectively. Simply stated, the SGD outflow is as much as 10.5% of the river outflow for N but only 0.78% for P.

The Jiulong River catchment across the Taiwan Strait from Taiwan is 40.8% of Taiwan’s size. The annual SGD discharge of Jiulong River is $0.213 \pm 0.058 \times 10^{10}$ m$^3$ (Wang et al.)17 which is 21.3% of the total discharge for Taiwan estimated by Chen et al.15. By way of comparison, the annual discharge of N, P, Si, TA and DIC for Jiulong River are $0.58–1.21 \times 10^9$, $2.9–6.1 \times 10^5$, $0.96–2.0 \times 10^9$, $0.75–1.6 \times 10^{10}$ and $0.88–1.82 \times 10^{10}$ mol, respectively.
based on the concentration and water discharge data of Wang et al.17 (Table 2). Recently Hong et al.19 also presented annual fluxes for N, Si and DIC at 1.83–1.95 × 10⁹, 2.94–3.14 × 10⁹ and 2.14–2.65 × 10¹⁰ mol, respectively (Table 2), comparable to the results of Wang et al.17.

Our study in Taiwan (Table 2) results in a SGD discharge of about 1.18 ± 0.83 × 10⁹ mol/a N (NO₃ + NO₂ + NH₄), which is equivalent to a yield of 3.28 ± 2.3 × 10⁴ mol/a N per square kilometer of the total catchment area. This value is similar to the annual yield of Jiulong River at 3.90–8.23 × 10⁴ mol/km²/a calculated based on the size of the catchment area and the flux of N reported by Wang et al.17. The flux of P resulted from this study for Taiwan is 9.3 ± 6.5 × 10⁶ mol/a, and the yield is 260 ± 180 mol/km²/a. The P flux and yield for Jiulong River at 2.9–6.1 × 10⁵ and 20–41 mol/km²/a (Table 2), respectively, are surprisingly low. As reported above, the N/P ratio obtained from this study for the submarine groundwater is 136. The flux data of Wang et al.17 for Jiulong River translate to a N/P ratio of 2000 which is extremely high although we realize that P is removed from groundwater. Our work for the Jiulong River (Table 3) results in an N (n = 11) to P (n = 9) ratio of 51 in the river basin and an N (n = 7) to P (n = 7) ratio of 27 in the estuary. Of note is that the average groundwater N and P concentrations calculated from the data of Wang et al.17 are 495 and 22.75 μM, respectively. The resulting N/P ratio is only 21.8, similar to what we found in the Jiulong River estuary but way below the reported ratio of 2,000 for the SGD by these authors.

Liu et al.16 reported the total annual flux of P for the Pearl River at 30–680 × 10⁶ mol and Liu et al.42 obtained similar values. Liu et al.42 also reported the N fluxes for the Pearl River at 3.65–157 × 10⁹ and 0.95–40 × 10⁹ mol in summer and winter, respectively. They reported the Si fluxes at 1.9–91.3 × 10⁹ and 0.51–23.4 × 10⁹ mol in summer and winter, respectively (Table 2). The Pearl River (Fig. 1) has a large catchment area of 453,700 km² which is located at the same latitude as southern Taiwan. The P flux of Liu et al.16 translates to a yield of 66–1,500 mol/km²/a and the results of Liu et al.42 are similar (Table 2). These results are comparable to those from Taiwan but much higher than those from the Jiulong River. The annual DIC flux of Liu et al.16 for the Pearl River is 15.3–34.7 × 10¹⁰ mol and the yield is 3.37–7.65 × 10¹⁰ mol/km³ which is comparable with our result in Taiwan. The reported SGD N and P in the literature are also given in Table 2, and the ranges are high. Our fluxes per m² for N and P in Taiwan are at the low end of these reported values.
| Nutrient | Taiwan | Jiulong River | Pearl River | Elsewhere (Literature) |
|----------|---------|---------------|-------------|-----------------------|
| **Total Flux mol** | 1.18 ± 0.83 × 10⁸ | 0.98 ± 0.69 | 3.28 ± 2.3 × 10⁹ | 2.47 ± 2.16–2.63 ± 2.31 (winter, Wang et al.)³⁶ |
| **Flux mol/m²** | 0.58–1.21 × 10⁶ | 1.83–1.95 × 10⁶ | 3.90–8.23 × 10⁴ | 0.95–4.0 × 10⁵ |
| **Yield mol/km²** | 3.65–157 × 10⁴ | 0.95–40 × 10³ | 3.17–7.14 × 10⁴ | 0.043–1.8 × 10⁵ |

Table 2. Annual total fluxes (mol), fluxes per m² seepage area and yields (mol/km² catchment area) of nutrients, TA, DIC and DOC by submarine groundwater from Taiwan, as well as Jiulong and Pearl River estuaries. aCalculated based on the flux data of Wang et al.¹ and the catchment area of Jiulong River. bCalculated based on the flux data of Hong et al.¹⁷ and the catchment area of Jiulong River. cObtained from Liu et al.¹⁷ and the catchment area of Pearl River. dObtained from Liu et al.⁴³.

| Nutrient | Taiwan | Jiulong River | Pearl River | Elsewhere (Literature) |
|----------|---------|---------------|-------------|-----------------------|
| **Total Flux mol** | 3.17 ± 2.22 × 10⁹ | 26.4 ± 18.5 | 8.81 ± 6.17 × 10⁵ | 25.1 (Sadat-Noori et al., 2016)⁴⁸ |
| **Flux mol/m²** | 0.88–1.82 × 10⁶ | 2.14–2.65 × 10⁶ | 6.0–12.4 × 10⁵ | 1.48–3.0 × 10⁴ |
| **Yield mol/km²** | 15.3–34.7 × 10⁴ | 3.37–7.65 × 10⁴ | 15.9–30.4 × 10⁴ | 10.9–20.1 × 10⁴ |

Table 3. Concentrations of NO₃, NO₂, NH₄ and PO₄ for riverine (S < 2) and estuarine water (S ≥ 2) of Jiulong River.

| S < 2 | S ≥ 2 |
|-------|-------|
| range | mean ± std | n | range | mean ± std | n |
| NO₃ (µM) | 71–173 | 95 ± 28 | 11 | 14–50 | 23 ± 12 | 7 |
| NO₂ (µM) | 3.0–39 | 14 ± 13 | 11 | 1.0–4.0 | 2.2 ± 0.9 | 8 |
| NH₄⁺ (µM) | 11–211 | 55 ± 63 | 11 | 14–59 | 23 ± 18 | 6 |
| PO₄ (µM) | 1.3–11.0 | 3.2 ± 3.2 | 9 | 0.5–7.6 | 1.8 ± 2.4 | 7 |

The total flux of Si for Taiwan is 0.68 ± 0.48 × 10⁹ mol/a compared with the larger flux of 0.96–2.0 × 10⁹ mol/a (Wang et al.)¹⁷ or 2.94–3.14 × 10⁹ mol/a (Hong et al.)²⁰ reported for Jiulong River. As for the yield the value of 6.53–13.6 × 10⁴ mol/km²/a calculated based on the flux data of Wang et al.¹⁷, and the value of 0.65–0.69 × 10⁴ mol/km²/a based on the data of Hong et al.³⁵ bracket the yield of Taiwan at 1.89 ± 1.33 × 10³ mol/km²/a. Liu et al.⁴³
reported the total flux and yield of Si for the Pearl River. Although their total fluxes are high their yields also bracket our results for Taiwan. The Si fluxes reported for a subtropical bay in south China are slightly higher than those for Taiwan (Table 2).

The TA and DIC fluxes for Taiwan are 3.43 ± 2.4 × 10^10 and 3.17 ± 2.22 × 10^10 mol/m²/a, respectively. These values compare with 0.75–1.6 × 10^10 and 0.88–1.82 × 10^10 mol/m²/a, respectively, for Jilulong River based on the data of Wang et al.17 (Table 2). The yield of TA for Taiwan at 9.5 ± 6.7 × 10^9 mol/km²/a is slightly higher than those for Jilulong River, at 5.1–10.9 × 10^9 mol/km²/a. The reported TA fluxes elsewhere bracket our results (Table 2). The yield of DIC for Taiwan is 8.81 ± 6.17 × 10^9 mol/km²/a which falls between the slightly lower value of 6–12.4 × 10^9 mol/km²/a (Wang et al.)17 and the slightly higher value of 14.6–18 × 10^9 (Hong et al.)19 for the Jilulong River. The Pearl River basin also has an abundance of calcareous rocks. The DIC yield (3.37–7.65 × 10^9 mol/km²/a) nevertheless, is smaller compared to our result in Taiwan. This is perhaps because the weathering is weaker in the less steep Pearl River basin. This points to the difficulty of comparing the total flux or yield. It is yet not possible to compare data per unit area of the ocean floor or per unit length of the coastal line. In terms of flux per m² of the seepage area, however, our DIC flux falls in the range reported elsewhere as shown in Table 2. Our DOC flux (9.5 ± 9.3 mol/m²/a) is also comparable with those reported in the literature (Table 2).

Conclusions

The concentrations, fluxes and yields of N, P, Si, TA and DIC for the SGD in Taiwan have been reported for the first time, and these values are broadly comparable with the data in the literature. The nutrients supplied by the SGD are insufficient to compensate the DIC supported at the same time. As a result, the SGD around Taiwan leads to a source of CO2 for the atmosphere in the coastal seas. Similar situation exists in the Jilulong and Pearl River estuaries in Southeast China, and perhaps in other coastal regions around the world as well.

Methods

Geologically Taiwan is relatively young. The collision of the Philippine Arc and the Asian continent gave rise to the Central Range of Taiwan, and the orogenesis is still going. The population is 23 million. The western part of Taiwan is mainly covered by undeformed sediments, and is heavily populated. Less populated is Southern Taiwan where the coasts are largely covered by coral reefs. Eastern Taiwan has a coastal range, and the less populated coasts are mainly rocky.

Preliminary sampling of the SGD in Taiwan was performed from 2004 to 2016. Twenty sampling sites around the coastal areas Taiwan are shown in Fig. 1. Measurements of SGD fluxes were reported in Chen et al.15. Submarine groundwater samples for chemical analysis were drawn by a device designed by Zhang and Satake mostly on the sandy coast. At one site 350 m off SW Taiwan divers collected freshwater at a water depth of 8 m. The corresponding local surface seawater sample was also collected. Samples for NO₃, NO₂, NH₄ and PO₄ were collected in Jilulong River and its estuary in 2008. Preserved samples, with saturated HgCl₂ added, were brought back and measured in the laboratory with details given in Chen11, Yang et al.41 and Tseng et al.46 and 2017. The percentage DO saturation (DO (%)) was calculated based on the solubility equation of Chen.47 The HCO₃⁻ and pCO₂ were calculated based on pH and TA using the CO2SYS program.

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Author Contributions
S.L. Wang was in charge of the writing of the manuscript and took part in the sampling and measurements. C.T.A. Chen was in charge of the overall structure of the manuscript as well as field work and laboratory analysis. T.H.H., H.C.T., H.K.L. and T.R.P. took part in the field work, laboratory measurements and data analysis. S.K., J.Z., L.Y.Y., X.L.G., J.Y.L., F.W.K., X.G.C., Y.Y. and Y.J.L. took part in the field work and laboratory measurements. All authors reviewed the manuscript.

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