

## Sources and conservative mixing of uranium in the Taiwan Strait

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### Abstract

Seawater samples are collected in the spring of 2013 from the Taiwan Strait for the analysis of uranium (U) concentrations and isotopic compositions using MC-ICP-MS, and the geochemical behavior patterns of U in the Taiwan Strait are then investigated. Average concentrations of individual U isotopes are  $(3.23 \pm 0.14) \mu\text{g}/\text{kg}$  for  $^{238}\text{U}$ ,  $(2.34 \pm 0.09) \times 10^{-2} \mu\text{g}/\text{kg}$  for  $^{235}\text{U}$  and  $(2.05 \pm 0.07) \times 10^{-4} \mu\text{g}/\text{kg}$  for  $^{234}\text{U}$ . Correspondingly, the U isotopic compositions are  $155 \pm 18$  for  $\delta^{234}\text{U}$  and  $138 \pm 2$  for  $^{238}\text{U}:^{235}\text{U}$ . The U concentrations and isotopic ratios in the Taiwan Strait are similar to those of open ocean seawater, suggesting the dominance of the open ocean input to the strait's U pool. However, river input, as suggested by the slightly lower salinity than that of the open ocean, also affected the U concentrations and isotopic compositions in the strait. From a compilation of U concentrations in the Taiwan Strait and adjacent areas, including the Jiulong Estuary and Zhujiang Estuary, the Xiamen Bay and the northern South China Sea, a strong and significant relationship between U concentration and salinity [ $U:S; U=(0.0934 \pm 0.0024)S + (0.0920 \pm 0.0615)$ ] is revealed, suggesting conservative mixing of U in the Taiwan Strait. To better understand the U geochemistry in the Taiwan Strait, a multiple endmembers mixing model is applied to estimate the contributions of potential sources. The open ocean seawater contributed 69%–95% of U in the Taiwan Strait, with river water approximately 2%, and dust deposition only around 0.13%. Therefore, the model results supported the open ocean input source and the conservative mixing behavior of U derived from the observation of U concentrations and isotopic ratios and U:S ratios. The sediment interstitial water may be an important source of U to the Taiwan Strait with a possible contribution of 3%–29%, consistent with previous investigations based on radium isotopes. However, further investigations are warranted to examine the U concentration in the sediment interstitial water and its input to the overlying seawater in the Taiwan Strait.

**Key words:** conservative mixing, uranium, seawater, Taiwan Strait

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### 1 Introduction

Uranium (U) in the ocean is derived mostly from terrestrial sources through river transport, aerosol deposition and submarine groundwater discharge (Dunk et al., 2002). Of these pathways, river transport of about 10.0 kt/a accounts for approximately 90% of the total U export into the ocean, while submarine groundwater discharge and aerosol deposition contribute only 10% of the total U in the ocean (Cochran, 1992; Palmer and Edmond, 1993). In addition, human activities, such as the use of phosphate fertilizer in agricultural production, may affect U concentration in the coastal ocean although its contribution is not well quantified (Dunk et al., 2002; Henderson and Anderson, 2003). Approximately 50% of the U concentration is removed from the water column by the reduction of the highly soluble hexavalent uranyl-carbonate complex,  $[\text{UO}_2(\text{CO}_3)_3]^{4-}$ , into the

insoluble tetravalent uraninite,  $\text{UO}_2$ , in the sediment interstitial water, which then enters the anoxic and suboxic sediments (Barnes and Cochran, 1990; Morford and Emerson, 1999). The adsorption by amorphous iron oxyhydroxides derived from the low temperature basalt alteration, hydrothermal scavenging and biogenic carbonate immobilization removes another 50% of U from the ocean (Staudigel et al., 1996; Chen et al., 1986b). Overall, U concentration in the ocean barely maintains a steady state between its sources and sinks (Dunk et al., 2002).

Uranium has an oceanic residence time of about  $4 \times 10^5$  a, far longer than the ocean circulation timescale (Ku et al., 1977; Dunk et al., 2002). Accordingly, U is largely a conservative element in the open ocean with quite stable U to salinity (U:S) and  $^{234}\text{U}:^{238}\text{U}$  ratios (Ku et al., 1977; Chen et al., 1986a). However, a non-conservative mixing behavior of U is observed in some estuaries and

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coastal waters. For example, annually, a significant amount of U (33.0 t) is released into the plume water through desorption from suspended particles in the turbid Huanghe (Yellow River) Estuary and this causes the apparent addition in the medium salinity region (Jiang et al., 2007). On the contrary, pronounced U removal is observed in the Amazon River plume due to inorganic and organic colloidal flocculation and scavenging and the formation of authigenic minerals (Swarzenski et al., 2004). The adsorption of dissolved U onto suspended particles and subsequent microbial reduction also results in significant U removal during estuarine mixing in the Ganges-Brahmaputra River Estuary (Carroll and Moore, 1993). In the Tampa Bay Estuary of Florida, U exhibits removal in the low salinity and enrichment in the high salinity zones, likely the result of interaction between surface water and groundwater altering the U reactivity (Swarzenski and Baskaran, 2007).

So far, investigations of U geochemistry in the Taiwan Strait and surrounding water body are very limited, and most focused on the Jiulong Estuary and Zhujiang Estuary and the northern South China Sea. Cai et al. (1996) note that the dissolved  $^{238}\text{U}$  in the Jiulong Estuary generally shows a conservative mixing behavior. The conservative behavior of U is further confirmed by a recent investigation into U size-fractionation in the Jiulong Estuary (Lü et al., 2008). The concentrations in the Zhujiang Estuary water range from 0.21 to 2.25  $\mu\text{g}/\text{kg}$  and generally follow a binary mixing between the river water and seawater endmembers (Sun et al., 1987). Meanwhile, the average U concentration is 2.98  $\mu\text{g}/\text{kg}$  in the South China Sea, lower than that in the open ocean, probably owing to dilution by rainfall (Chen et al., 1997). The U:S ratio in the South China Sea is also about 10% lower than that in the Jiulong Estuary (Chen et al., 1997). However, no U concentration or geochemical behavior has been reported in the Taiwan Strait, a confluence area of water masses from the Jiulong Estuary and Zhujiang Estuary and the South China Sea.

The objectives of this study were to investigate the concentration and isotopic compositions of U in the Taiwan Strait to assess its mixing behavior and budget, and to provide new insights into the potential roles of submarine groundwater and oxic sediments in the oceanic U budget. To achieve these ends, a fast and simple MC-ICP-MS analytical procedure was utilized for the pre-

cise determination of U concentrations and isotopic compositions in the Taiwan Strait in April 2013. The high quality data of U concentrations and isotopic compositions obtained in this study enabled us to explore the mixing behavior and sources of U in the Taiwan Strait in a regional context.

## 2 Methods

### 2.1 Site description

The Taiwan Strait is located between China's Mainland and Taiwan Island of China with about three-quarters of its area shallower than 60 m (Fig. 1). However, the water is deeper than 1 000 m in the southeastern Taiwan Strait bordering the northern South China Sea (Hong et al., 2011). The Taiwan Strait forms the sole water passageway between the South China Sea and the East China Sea (Hu et al., 2011). Hydrology in the Taiwan Strait is affected by the East Asia monsoon system, in which the southwest monsoons prevails in summer and the northeast monsoon does in winter (Hong et al., 2011). The water masses in the Taiwan Strait are well characterized based on salinity and temperature properties in previous hydrologic investigations (Naik and Chen, 2008; Hu et al., 2011). In winter, the Zhe-Min Coastal Current flows southward along the west coast of the Taiwan Strait while the South China Sea waters and the Kuroshio Current flow northward in summer with seasonal upwelling occurring near the Minnan-Taiwan Bank. Spring is a transition period between the northeast and southwest monsoons. The Zhe-Min Coastal Water therefore gradually retreats with increasing intrusion of South China Sea waters and the Kuroshio Current into the Taiwan Strait.

The southern Taiwan Strait receives fresh water input from the Zhujiang River, the Hanjiang River, the Jiulong River and some small coastal rivers. River plumes during spring rainy season can be clearly observed during spring to summer in the Taiwan Strait (Hu et al., 2010; Jan et al., 2010).

### 2.2 Sample collection

Surface water samples along Transect C and from a profile at Sta. C11 were collected from the southern Taiwan Strait in April 2013 (Fig. 1, Table 1). Surface water was pumped through a pre-cleaned 0.45  $\mu\text{m}$  pore size polycarbonate filter cartridge (Osmon-

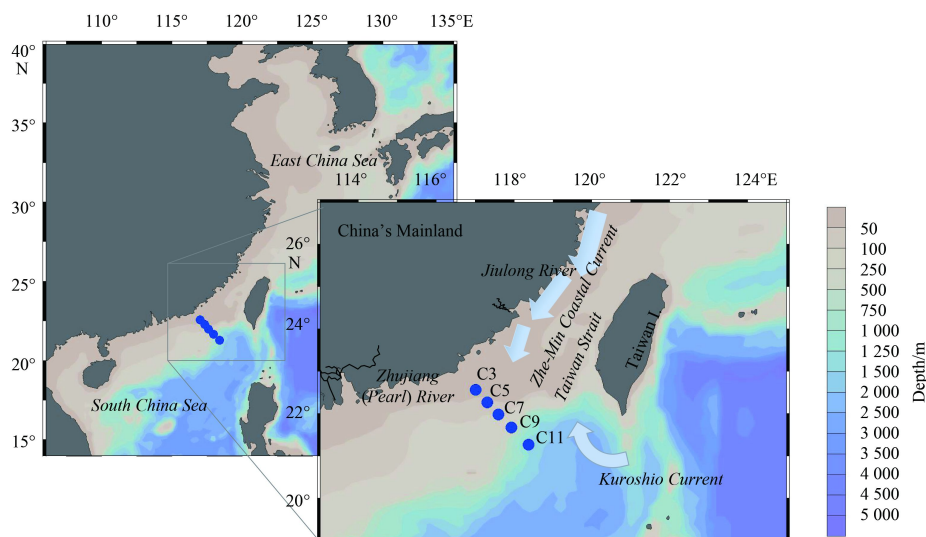


Fig. 1. Sampling stations in the Taiwan Strait.

**Table 1.** Sampling information in the Taiwan Strait

Station	North latitude/(°)	East longitude/(°)	Depth/m	Date	Layer/m	Salinity	Temperature/°C
C3	22.557	117.014	48	2013/04/24	1	34.21	24.0
C5	22.270	117.311	64	2013/04/24	1	34.07	25.2
C7	21.977	117.603	153	2013/04/24	1	34.06	25.9
C9	21.670	117.938	1 370	2013/04/24	1	34.06	26.0
C11	21.267	118.369	2 373	2013/04/24	1	34.03	26.1
					25	34.10	25.3
					50	34.12	24.9
					75	34.30	23.3
					100	34.58	21.2
					150	34.63	17.4
					200	34.59	14.9
					300	34.45	11.6
					400	34.41	9.5
					500	34.42	8.2
					600	34.43	7.0
					800	34.47	5.7
					1 000	34.52	4.4

ics). Water samples at Sta. C11 profile were collected using 12 L Niskin bottles mounted on a rosette sampler with a Seabird CTD and were also filtered with the Osmonics filter cartridge. The filtrates were collected in trace metal clean polypropylene containers and acidified to pH 2 with 6 mol/L double quartz-distilled HCl.

### 2.3 Column chemistry

Water samples were weighed and spiked with  $^{236}\text{U}$ . After 24 h equilibration,  $\text{NH}_3\cdot\text{H}_2\text{O}$  was then added to adjust the pH value to approximately 10 to co-precipitate the actinides with Mg-oxyhydroxides precipitate (Okubo et al., 2012). After 2 d of precipitate settling, the majority of the water were siphoned off and the precipitate was centrifuged and washed with alkaline Milli-Q water. The obtained precipitate was dissolved with concentrated  $\text{HNO}_3$  and then concentrated  $\text{NH}_3\cdot\text{H}_2\text{O}$  was added to form the secondary  $\text{Mg}(\text{OH})_2$  precipitate. By doing so, the amount of precipitate could be reduced for subsequent column chemistry (Scholten et al., 2008; Auro et al., 2012). After double  $\text{Mg}(\text{OH})_2$  coprecipitation, the resultant precipitate was dissolved in concentrated  $\text{HNO}_3$  and transferred to a clean fluorinated ethylene propylene beaker. Concentrated HF,  $\text{HNO}_3$  and  $\text{HClO}_4$  were added to the beaker to decompose organic matter. After the digestion solution was removed by drying, the samples were then redissolved in 8 mol/L  $\text{HNO}_3$  for column purification.

The U was purified with Bio-Rad AG1-X8 100–200 mesh anion exchange resin (Thomas et al., 2006; Zheng et al., 2006). Samples in 8 mol/L  $\text{HNO}_3$  solution were loaded onto the column with a further 8 mol/L  $\text{HNO}_3$  rinse to wash off metal impurities. Th was eluted with 9 mol/L HCl. Part of the U was also lost in the two steps above. The remaining U was eluted with 0.1 mol/L HCl. Total U yield during the  $\text{Mg}(\text{OH})_2$  coprecipitation and column chemistry, assessed using recovery of the  $^{236}\text{U}$  spike, was approximately 20%. The  $\text{Mg}(\text{OH})_2$  coprecipitation recovered only 40% U from the water samples whereas half of the recovered U was lost during column chemistry.

### 2.4 MC-ICP-MS analysis

U was measured on a Nu Instruments MC-ICP-MS with 12 faraday collectors and three ion counters (Table 2). The standard-sample bracketing protocol was designed to assess the mass

**Table 2.** Collector array of uranium measurements in seawater

	Step	F9 <sup>1)</sup>	F10 <sup>1)</sup>	F11 <sup>1)</sup>	F12 <sup>1)</sup>	IC0 <sup>2)</sup>	IC1 <sup>2)</sup>	IC2 <sup>2)</sup>
U-standard	1	$^{238}\text{U}$				$^{235}\text{U}$		
	2				$^{238}\text{U}$	$^{235}\text{U}$	$^{234}\text{U}$	
	3						$^{235}\text{U}$	$^{234}\text{U}$
U-sample	4				$^{238}\text{U}$	$^{235}\text{U}$	$^{234}\text{U}$	
	5					$^{236}\text{U}$	$^{235}\text{U}$	$^{234}\text{U}$

Note: <sup>1)</sup> F9–12: faraday collectors; and <sup>2)</sup> IC0–2: ion counters.

fractionation coefficient and the gain between faraday cups and electron multipliers. An integration time of 10 s with 15 to 30 counting cycles was adopted to reduce the error from the beam drift.

Mass discrimination and ion-counter gain were corrected by measurement of the CRM-145 U standard ( $^{235}\text{U}:^{238}\text{U} = 0.007\ 253$ ,  $^{234}\text{U}:^{238}\text{U} = 0.000\ 055\ 4$ ) preceding each sample analysis. Abundance sensitivity, background and memory effect of the instrument were also corrected by measuring the CRM-145 U standard (Robinson et al., 2004). Long-term monitoring over three months indicated that the analysis precision of  $^{235}\text{U}:^{238}\text{U}$  and  $^{234}\text{U}:^{238}\text{U}$  was within 0.2%–0.8%, and the accuracy was better than 0.2%.

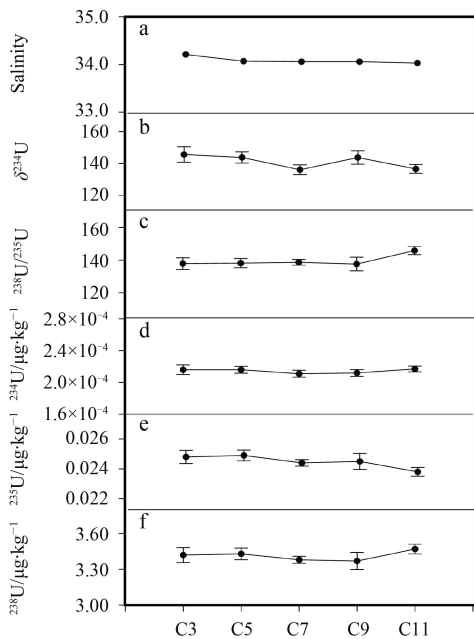
## 3 Results

### 3.1 Concentrations of U in the Taiwan Strait

All three U isotope ( $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$ ) concentrations varied within a small range in this study. The  $^{238}\text{U}$  concentration ranged between 3.04 and 3.47  $\mu\text{g}/\text{kg}$  with an average of  $(3.42\pm 0.04)\ \mu\text{g}/\text{kg}$  in the surface water (Table 3, Fig. 2). At Sta. C11, the  $^{238}\text{U}$  concentration slightly decreased from 3.47  $\mu\text{g}/\text{kg}$  in surface water to 3.04  $\mu\text{g}/\text{kg}$  at the 1 000 m depth, with an average of  $(3.18\pm 0.12)\ \mu\text{g}/\text{kg}$  (Table 3, Fig. 3). Overall,  $^{238}\text{U}$  concentrations in the Taiwan Strait were close to those of the open ocean, although the salinity is slightly lower (Ku et al., 1977). Therefore, U in the Taiwan Strait was likely to be dominated by the input from the open ocean. The  $^{235}\text{U}$  concentrations ranged from  $2.20\times 10^{-2}$  to  $2.49\times 10^{-2}\ \mu\text{g}/\text{kg}$ , and were three orders of magnitude lower than the  $^{238}\text{U}$  concentrations. The average concentration was  $(2.45\pm 0.04)\times 10^{-2}\ \mu\text{g}/\text{kg}$  in the surface water and  $(2.31\pm 0.07)\times 10^{-2}\ \mu\text{g}/\text{kg}$  in the profile of Sta. C11 (Table 3, Figs 2 and 3). The  $^{234}\text{U}$  varied from  $1.95\times 10^{-4}$  to

**Table 3.** Uranium concentrations and isotopic ratios of Transect C in the Taiwan Strait

Station	Depth/m	<sup>238</sup> U		<sup>235</sup> U		<sup>234</sup> U		$\delta^{234}\text{U}$		<sup>238</sup> U: <sup>235</sup> U	
		$\mu\text{g}/\text{kg}$	$1\sigma$	$10^{-2} \mu\text{g}/\text{kg}$	$1\sigma$	$10^{-4} \mu\text{g}/\text{kg}$	$1\sigma$	Ratio	$1\sigma$	Ratio	$1\sigma$
C3	1	3.42	0.06	2.48	0.05	2.16	0.06	146	5	138	4
C5	1	3.43	0.05	2.49	0.04	2.16	0.04	144	4	138	3
C7	1	3.38	0.03	2.44	0.02	2.11	0.04	136	3	139	2
C9	1	3.37	0.07	2.45	0.05	2.12	0.04	144	4	138	4
C11	1	3.47	0.04	2.38	0.03	2.17	0.04	137	3	146	2
	25	3.29	0.04	2.37	0.03	2.05	0.04	133	3	139	2
	50	3.23	0.07	2.37	0.14	2.04	0.05	147	5	137	9
	75	3.11	0.28	2.33	0.12	1.95	0.17	143	18	134	14
	100	3.17	0.19	2.34	0.09	2.00	0.12	147	13	136	10
	150	3.17	0.17	2.31	0.27	2.01	0.11	151	11	137	18
	200	3.1	0.07	2.23	0.11	2.02	0.04	182	6	139	7
	300	3.1	0.05	2.23	0.08	2.01	0.04	181	4	139	6
	400	3.3	0.19	2.41	0.14	2.10	0.12	154	13	137	11
	500	3.22	0.25	2.34	0.18	2.05	0.16	161	18	138	15
600	3.11	0.07	2.26	0.02	2.04	0.05	192	6	138	3	
800	3.06	0.02	2.20	0.11	1.97	0.03	176	3	139	7	
1 000	3.04	0.04	2.23	0.05	1.95	0.03	168	4	136	4	
Average		3.23	0.14	2.34	0.09	2.05	0.07	155	18	138	2

**Fig. 2.** Distribution of uranium concentration, <sup>238</sup>U:<sup>235</sup>U,  $\delta^{234}\text{U}$  and salinity in the C surface transect.

$2.17 \times 10^{-4} \mu\text{g}/\text{kg}$  with averages of  $(2.14 \pm 0.04) \times 10^{-4} \mu\text{g}/\text{kg}$  in the surface water and  $(2.03 \pm 0.06) \times 10^{-4} \mu\text{g}/\text{kg}$  in the C11 profile.

### 3.2 Isotopic ratios of U in the Taiwan Strait

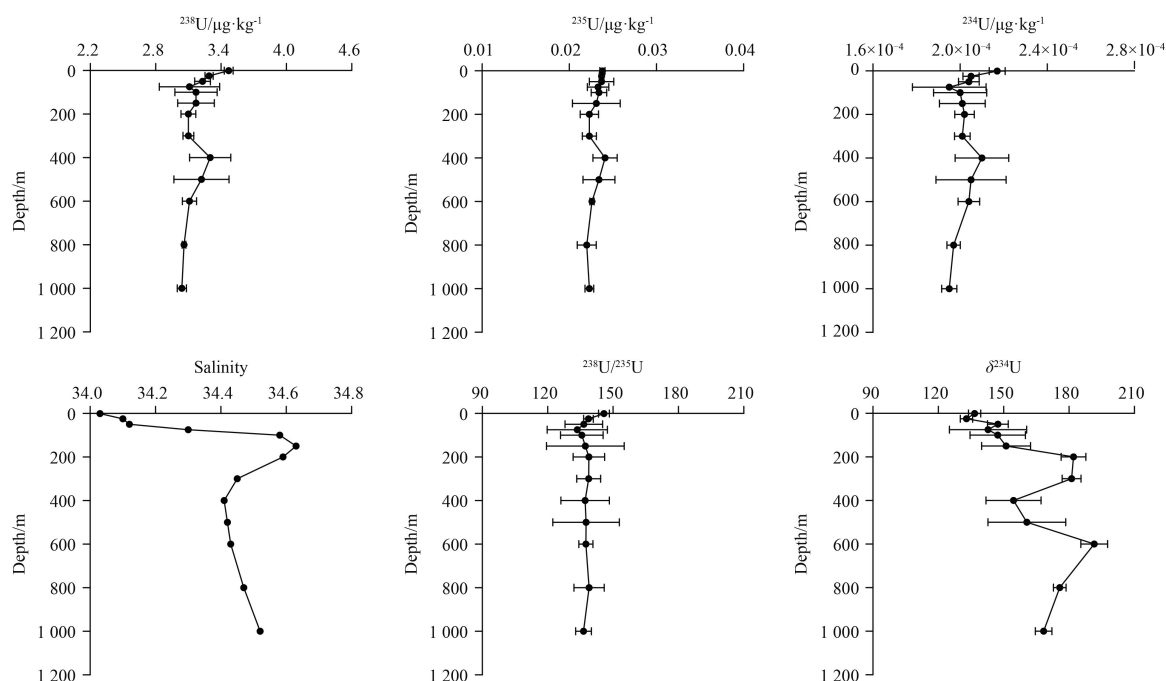
U isotopic ratios remained quite stable in the southern Taiwan Strait (Table 3, Fig. 2). The <sup>238</sup>U:<sup>235</sup>U ratio ranged between 134 and 146 with an average value of  $138 \pm 2$ , which was highly consistent with the open ocean seawater ratio of 137.82 (Hiess et al., 2012). The average value of  $\delta^{234}\text{U}$ , defined as  $[(^{234}\text{U}:^{238}\text{U})_{\text{A.R.}} - 1] \times 10^3$ , was  $(156 \pm 18)$  in the southern Taiwan Strait, which was slightly higher than the open ocean seawater value of 146.8 (Anderson et al., 2010), but showed no significant difference considering the analysis error (Table 4). The consistency of both U isotopic ratios between this study and the open

ocean datasets suggested dominant U sources from the open ocean in the southern Taiwan Strait. In addition, the average  $\delta^{234}\text{U}$  in the southern Taiwan Strait was higher than those in the Jiulong Estuary and Zhujiang Estuary (Sun et al., 1987; Chen et al., 1997; Lü et al., 2008) and the northeastern South China Sea (Chen et al., 1997), implying limited riverine U contribution. The low U concentrations in the Jiulong Estuary and Zhujiang Estuary further excluded local rivers as the major U sources in the southern Taiwan Strait. Overall, the U provenance in the southern Taiwan Strait was largely controlled by its main water sources, that is, the open South China Sea and the Kuroshio Current as revealed by the observed high salinity (34.03–34.63) (Table 1).

## 4 Discussion

### 4.1 Hydrology

The temperature increased from onshore to offshore water in the southern Taiwan Strait while the low salinity water was limited to the area close to the Jiulong Estuary and coastline (Fig. 4). The patterns of the temperature and the salinity indicate that the onshore waters are influenced by the Jiulong Dilution Water and the Zhe-Min Coastal Current, while the offshore waters with high temperature and high salinity are impacted by the South China Sea Water and the Kuroshio Current (Naik and Chen, 2008; Hu et al., 2011). In the present study, Sta. C11 was the only station located on a continental slope deeper than 1 000 m. As indicated by the salinity and temperature profiles, typical water masses from the South China Sea are well distinguished at Sta. C11 (Figs 4 and 5) (Wong et al., 2007; Dai et al., 2009). Therefore, the water samples from the water column of Sta. C11 were used to represent the open ocean endmember for the physical mixing model quantification. The temperature gradually decreased with increasing depth from 26 (surface) to 4.4°C (1 000 m). The salinity remained relatively stable (34.1) in the upper 75 m, consistent with the mixed layer depth (20–80 m, Fig. 5) (Wong et al., 2007). A salinity maximum (34.6) and minimum (34.4) existed at 150 and 500 m, respectively, which suggested their sources from the North Pacific Tropical Water and the North Pacific Intermediate



**Fig. 3.** Distribution of uranium concentration,  $^{238}\text{U}:^{235}\text{U}$ ,  $\delta^{234}\text{U}$  and salinity in the C11 profile.

**Table 4.** Comparison of the  $\delta^{234}\text{U}$  value between this study and previous studies

Area	$\delta^{234}\text{U}$	Detection method	Preconcentration method	References
Atlantic, Pacific, Arctic and southern Oceans	$144 \pm 2$	TIMS	Fe co-precipitation	Chen et al. (1986)
Bahamas	$143 \pm 0.5$	ICP-MS	Fe co-precipitation	Henderson et al. (1999)
Mediterranean Sea	$149.4 \pm 0.6$	TIMS	Fe co-precipitation	Delanghe et al. (2002)
North Atlantic Ocean	$149.6 \pm 1$	TIMS	Fe co-precipitation	Delanghe et al. (2002)
Pacific Ocean	$150.9 \pm 1.7$	TIMS	Fe co-precipitation	Delanghe et al. (2002)
Indian Ocean	$149.3 \pm 3.2$	TIMS	Fe co-precipitation	Delanghe et al. (2002)
Atlantic Ocean	$146.6 \pm 2.5$	MC-ICP-MS	Fe co-precipitation	Robinson et al. (2004)
East Indian Ocean, western Pacific Ocean, South China Sea	$147.4 \pm 2$	SF-ICP-MS	Fe co-precipitation	Shen et al. (2008)
Pacific Ocean	$146.8 \pm 0.1$	MC-ICP-MS	Fe co-precipitation	Anderson et al. (2010)
Zhujiang Estuary	70	$\alpha$	Fe co-precipitation	Liu et al. (1988a)
Changjiang Estuary	$375 \pm 13$	$\alpha$	Fe co-precipitation	Liu et al. (1988b)
Zhujiang Estuary	$120 \pm 4.3$	$\alpha$	Fe co-precipitation	Sun et al. (1987)
Jiulong Estuary	$130 \pm 1.2$	$\alpha$	Fe co-precipitation	Lü et al. (2008)
Nansha waters of South China Sea	$135 \pm 1.2$	$\alpha$	Fe co-precipitation	Chen et al. (1997)
Northeast of South China Sea	$120 \pm 1.1$	$\alpha$	Fe co-precipitation	Chen et al. (1997)
Xiamen Bay	$140 \pm 1.2$	$\alpha$	Fe co-precipitation	Chen et al. (1997)
Jiulong Estuary	$120 \pm 1.1$	$\alpha$	Fe co-precipitation	Chen et al. (1997)
Huanghe Estuary	$320 \pm 26$	$\alpha$	Fe co-precipitation	Jiang et al. (2007)
Taiwan Strait	$155 \pm 18$	MC-ICP-MS	Mg co-precipitation	this study

Note: TIMS represents thermal ionization mass spectrometry, and  $\alpha$  represents  $\alpha$  spectrometry.

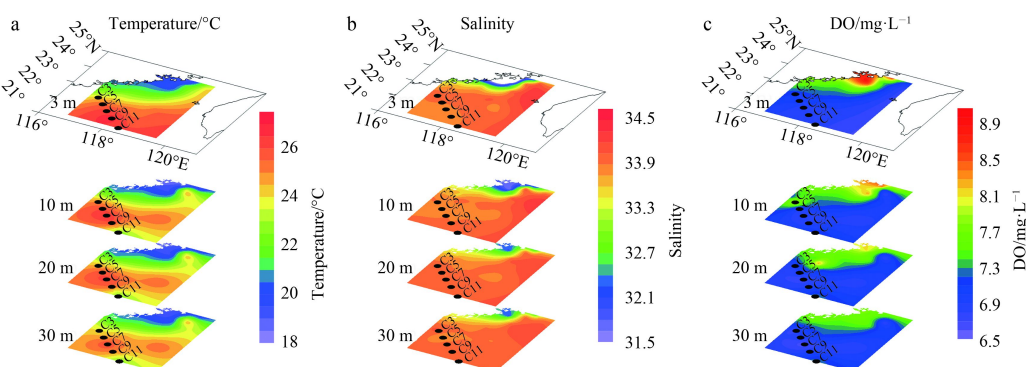
Water, respectively. In the water column deeper than 500 m, the salinity increased gradually with depth and was closer to the South China Sea Deep Water and the northern Pacific Deep Water (Wong et al., 2007).

Dissolved oxygen concentration obtained during the cruise ranged from 6.5 to 9.0 mg/L (Fig. 4), suggesting an oxic status in the water column of the Taiwan Strait.

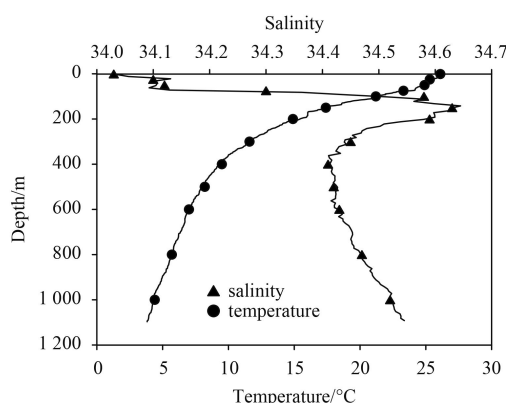
#### 4.2 Conservative behavior of U in the Taiwan Strait

The average  $^{238}\text{U}:^{235}\text{U}$  ratio in the southern Taiwan Strait was ( $0.0943 \pm 0.0046$ ), within the range for the open ocean seawater

( $0.0919\text{--}0.1000$ ) (Table 5 and references therein). Such consistency, together with the U isotopic ratios being close to the open ocean seawater, indicated the conservative geochemical behavior of U in the southern Taiwan Strait. The conservative behavior of U was also supported by the oxic seawater environment in the Taiwan Strait as suggested by its high dissolved oxygen level (Fig. 4). However, Lü et al. (2008) obtained a  $^{238}\text{U}:^{235}\text{U}$  ratio of ( $0.0900$ ) in the Jiulong Estuary with only five samples along a salinity gradient. This ratio is significantly lower than that in the southern Taiwan Strait. To better examine the U mixing behavior and constrain  $^{238}\text{U}:^{235}\text{U}$  ratio in the southern Taiwan Strait and northern



**Fig. 4.** Distribution of temperatur (a), salinity (b) and dissolved oxygen (c) in the Taiwan Strait during late spring in 2013.



**Fig. 5.** Distributions of temperature and salinity in the Sta. C11 profile during late spring in 2013.

South China Sea and adjacent estuaries, we compiled available U concentrations in these areas (Fig. 6). The U concentration in the southern Taiwan Strait was similar to those in the Xiamen Bay and the northeastern South China Sea, but higher than those in the Zhujiang and Jiulong estuaries with lower salinities (Cai et al., 1996; Chen et al., 1997; Lü et al., 2008; Sun et al., 1987). Taken as a whole, U is significantly correlated with the salinity ( $R^2=0.978$ ,  $p<0.0001$ ) with a  $^{238}\text{U}:S$  ratio of  $(0.0934 \pm 0.0024)$  in the coastal systems including the southern Taiwan Strait, the northern South

China Sea and the Zhujiang and Jiulong estuaries (Fig. 6). The significance of the regression between U concentration and salinity revealed a conservative geochemical behavior of U in the Taiwan Strait. Furthermore, the identical  $^{238}\text{U}:S$  ratio obtained from the whole dataset in the broad Taiwan Strait to those of the global open ocean also indicated that the mixing of U between the riverine input and open ocean sources in the Taiwan Strait followed the global pattern (Table 5 and references therein). Unlike the Huanghe Estuary, which shows apparent U addition during estuarine mixing (Jiang, 2008), the conservative U mixing in the southern Taiwan Strait and the northeastern South China Sea coastal system makes it possible to estimate U concentrations based on the available salinity, given the generic  $^{238}\text{U}:S$  ratio of  $(0.0934)$ . This was extremely helpful to simplify the laboratory analysis and accelerated the data output in the investigations on the U-series nuclide geochemistry as well as the estimation of particulate organic carbon export and particle dynamics in the subtropical and tropical marginal seas using the  $^{238}\text{U}\text{-}^{234}\text{Th}$  and  $^{234}\text{U}\text{-}^{230}\text{Th}$  disequilibria.

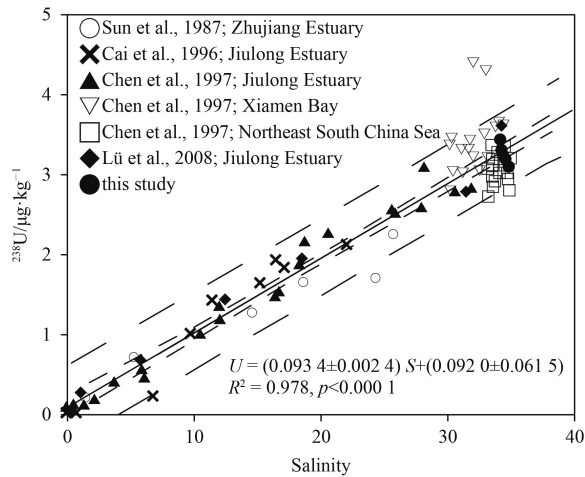
#### 4.3 Source contribution of U in the surface Taiwan Strait

The possible sources of U in the Taiwan Strait included the transport of open ocean water masses, riverine inputs, diffusion of sediment interstitial water, and dust precipitation. To quantitatively determine the contribution of individual endmembers in the Taiwan Strait, a physical mixing model was applied in this study:

**Table 5.** Comparison of the U:S ratio in this study and previous studies

Area	Salinity	U/S	Detection method	Preconcentration method	Sampling time	References
Atlantic, Pacific, Arctic and southern Oceans	30.3–36.2	$0.0934 \pm 0.0056$		Fe co-precipitation	April 1968	Ku et al. (1977)
Atlantic, Pacific, Arctic and southern Oceans	34.1–36.1	$0.0919 \pm 0.0005$	TIMS	Fe co-precipitation	—	Chen et al. (1986)
Mediterranean Sea, northeast Atlantic Ocean and Indian Ocean	34.7–38.6	$0.0923 \pm 0.0019$	TIMS	Fe co-precipitation	—	Delanghe et al. (2002)
Atlantic Ocean	35.4–37.3	$0.0953 \pm 0.0004$	MC-ICP-MS	Fe co-precipitation	July 2000	Robinson et al. (2004)
Pacific Ocean	33.0–34.4	$0.0920 \pm 0.0018$	ICP-MS	Fe co-precipitation	—	Zheng et al. (2006)
Mediterranean Sea	37.3–39.1	$0.0933 \pm 0.0013$	ICP-MS	Fe co-precipitation	March 1997, February 2002	Pates et al. (2007)
Subtropical Atlantic Ocean	32.7–37.1	$0.1000^{(1)} \pm 0.0060$	ICP-MS	Fe co-precipitation	2007–2010	Owens et al. (2011)
Arctic Ocean	0–35	$0.0908 \pm 0.0005$	TIMS	Fe co-precipitation	2008	Not et al. (2012)
Jiulong Estuary	1.1–31.4	0.0900	Fe co-precipitation	March 2002	Lü et al. (2008)	
Taiwan Strait	0.2–34.6	$0.0934 \pm 0.0024$	MC-ICP-MS	Mg co-precipitation	April 2013	this study

Note:  $^{1)}^{238}\text{U}(10^{-9}) = (0.100 \pm 0.0006) \times S - (0.326 \pm 0.206)$ .



**Fig. 6.** Uranium in the Taiwan Strait and adjacent area as a function of salinity. The lines involve data of previous studies and the present work, being the regression (solid), 95% confidence interval (dash), and 95% prediction interval (dot) lines.

$$U_o \cdot f_o + U_r \cdot f_r + U_i \cdot f_i + U_d \cdot f_d = U, \quad (1)$$

$$S_o \cdot f_o + S_r \cdot f_r + S_i \cdot f_i = S, \quad (2)$$

$$f_o + f_r + f_i + f_d = 1, \quad (3)$$

where  $f$ ,  $U$ ,  $S$  denote the fraction, uranium concentration and salinity of the individual contributing source, respectively and the subscripts  $o$ ,  $r$ ,  $i$  and  $d$  represent the open ocean water masses, rivers, sediment interstitial water and dust precipitation, respectively. The salinity of dust precipitation is 0 and, therefore, can be eliminated from Eq. (2).

Recently, it has been found that atmospheric precipitation plays an important role in the  $U$  budget in soils. For example, Pett-Ridge et al. (2007) note that dust precipitation dominates the  $U$  pool in Hawaiian soils; and Pelt et al. (2013) further note that the Sahara dust contributes up to 25% of the total  $U$  in Mount Cameroon volcano soil. However, the dust contribution to the seawater  $U$  still remains unclear (Dunk et al., 2002; Henderson and Anderson, 2003). In the Taiwan Strait, the aerosol deposition flux is estimated as about  $50 \text{ g}/(\text{m}^2 \cdot \text{a})$  during 1997–2007 (Tan et al., 2013). Given the average  $U$  concentration in the upper continental crust ( $2.4 \pm 0.6 \text{ } \mu\text{g}/\text{g}$ ) in the upper continental crust (Taylor et al., 1981), the  $U$  dust input can be estimated as  $(120 \pm 30) \text{ } \mu\text{g}/(\text{m}^2 \cdot \text{a})$  in the Taiwan Strait.

Although the anthropogenic activities cause severe haze episodes and dust storms in northern China, the atmospheric environment in southern China remains generally undisturbed (Tan et al., 2013; Guo et al., 2014). Therefore, the average  $U$  concentration in the upper continental crust can reasonably represent the  $U$  concentration of dust to the Taiwan Strait.

The contribution of dust deposition to the seawater  $U$  can be estimated as follows:

Dust uranium contribution equals dust uranium deposition multiplied by water residence time/(mixing layer depth multiplied by seawater density).

While the water residence time in the southern Taiwan Strait is not available, the water residence time in the surface mixing layer of the northern South China Sea is determined as 1.3 a (Cai et al., 2004). Assuming a similar water residence time as in the

northern South China Sea, the contribution of dust precipitation to the seawater  $U$  in the southern Taiwan Strait can be estimated as  $(4.35 \pm 1.09) \text{ ng}/\text{kg}$  with a 35 m mixing layer depth, based on the salinity and water temperature profiles at Sta. C11. Apparently, the dust contribution was insignificant and accounted for only  $(0.13 \pm 0.03)\%$  of the total seawater  $U$  in the southern Taiwan Strait.

Surface water in the C transect had lower salinity than the open ocean due to the influence of the Zhe-Min Coastal Current, which receives a large amount of fresh water from the coastal rivers. Among the rivers, the Jiulong River is closest to the study area and directly influence the salinity and  $U$  levels in the southern Taiwan Strait during spring. Therefore, the Jiulong River water was selected to represent the river water endmember in the physical mixing model. The  $U$  concentration in the Jiulong River is  $(0.064 \pm 0.007) \text{ } \mu\text{g}/\text{kg}$  at 0.2 salinity (Cai et al., 1996). In contrast, the  $U$  concentration in the open ocean is  $3.33 \text{ } \mu\text{g}/\text{kg}$  with a salinity value of 35 (Ku et al., 1977).

Sediment interstitial water is a potential source of  $U$  to the overlying seawater (Dunk et al., 2002). The high dissolved oxygen level throughout the whole water column, especially near the sediment/water interface, minimized the reduction of  $(\text{UO}_2(\text{CO}_3)_3)^{2+}$  in the water column and further benefited the desorption of soluble  $U$  from the shelf sediment (Fig. 4). The shallow water depth and the well mixed water column observed in the Taiwan Strait shelf area favored the entrainment of the shelf sediment interstitial water and the diffusion of  $U$  into the overlying seawater (Fig. 4). On the basis of radium isotopic balance, Liu et al. (2012) estimate that the submarine groundwater discharge, replenished by the recycled seawater, accounts for 12%–21% of the Zhujiang River discharge into the area including the northern South China Sea shelf and our study sites. While there is no available  $U$  concentration data for the sediment interstitial water in the Taiwan Strait, Henderson et al. (1999) reveal that the  $U$  concentration in the carbonate sediment interstitial water shallower than 50 m below the surface is in the range  $0.93\text{--}8.75 \text{ } \mu\text{g}/\text{kg}$ . This is similar to the  $U$  concentrations normally found in groundwater and matches the reported sediment interstitial water  $U$  concentrations (Cochran et al., 1986; Gariépy et al., 1994; Dunk et al., 2002). However, when the  $U$  concentration was below or close to the seawater  $U$  concentration, the mixing model derived a sediment interstitial water fraction less than 0 or unreasonably high (close to 100%). Based on the results of previous investigations of submarine groundwater contribution and the oxic state of the seawater (Hong et al., 2011; Liu et al., 2012), we assumed that the sediment interstitial water in the Taiwan Strait had a  $U$  concentration range of  $3.75\text{--}8.75 \text{ } \mu\text{g}/\text{kg}$  with a salinity similar to the overlying bottom water (34.0). The minimum  $U$  concentration was 10% larger than the seawater concentration for a better correction of the model error.

With the salinity and  $U$  concentrations available for different endmembers as discussed above, the contribution of each endmember to the surface seawater  $U$  pool in the southern Taiwan Strait was estimated (Table 6). The contributions of open ocean seawater, sediment interstitial water, and the river water were 69%–95%, 3%–29%, and around 2%, respectively. Overall, the open ocean seawater was the dominant source of  $U$  in the Taiwan Strait while the river water and the atmospheric deposition accounted for only an insignificant fraction. This was consistent with the observed high salinity and conservative mixing behavior of  $U$  (Fig. 6). The sediment interstitial water could supply up to 29% of the  $U$  to the Taiwan Strait when a  $U$  concentration close to that of seawater was selected. Such high input from

**Table 6.** Summary of endmember values applied in the three endmembers mixing model

Parameter	Source				Surface water
	Open ocean	River water	Sediment interstitial water	Atmospheric precipitation	
Salinity	35.0	0.2	34.0	0	34.09±0.07
$^{238}\text{U}/\mu\text{g}\cdot\text{kg}^{-1}$	3.33	0.064	0.93–8.75	0.004 4	3.41±0.04
$f(U_i=8.75 \mu\text{g}/\text{kg})$	94.72%	2.41%	2.74%	0.13%	–
$f(U_i=3.75 \mu\text{g}/\text{kg})$	68.84%	1.64%	29.39%	0.13%	–

sediment interstitial water was possible as suggested by the significant submarine groundwater discharge from the recycled seawater (Liu et al., 2012). However, the U concentration in the sediment interstitial water warranted further measurement to better understand the role of sediment interstitial water or submarine groundwater discharge in the U geochemistry of the Taiwan Strait.

In a recent review of the oceanic U budget, the contribution of recycled submarine groundwater discharge was ignored owing to the lack of evidence of U addition during the recycling of seawater (Dunk et al., 2002). However, our result shows a significant contribution of sediment interstitial water to the seawater U budget through recycled submarine groundwater discharge. It not only provides an indirect evidence for the enrichment of U during the interaction between submarine groundwater and sediment, but also implies the potential role of recycled submarine groundwater discharge in the oceanic U budget, something which has long been ignored. In addition, our results suggested that the oxic sediments could act as a U source in contrast to the removal of U in suboxic and anoxic sediments (Barnes and Cochran, 1990; Morford and Emerson, 1999; Dunk et al., 2002). Since oxic sediments are widely spread in coastal regions and continental shelves, a reevaluation of the role of oxic sediments in the oceanic U budget is greatly needed.

## 5 Conclusions

U shows various mixing behaviors in estuaries and coastal oceans, which hinders precise estimation of U mass balance and geochemistry in the oceans. In this study, we reported high precision U concentrations and isotopic compositions in the Taiwan Strait, and investigated the mixing behavior and sources of U in the Taiwan Strait. Average concentrations of individual U isotopes were  $3.23\pm 0.14 \mu\text{g}/\text{kg}$  for  $^{238}\text{U}$ ,  $(2.34\pm 0.09)\times 10^{-2} \mu\text{g}/\text{kg}$  for  $^{235}\text{U}$  and  $(2.05\pm 0.07)\times 10^{-4} \mu\text{g}/\text{kg}$  for  $^{234}\text{U}$ , and the average U isotopic compositions were  $155\pm 18$  and  $138\pm 2$  for  $\delta^{234}\text{U}$  and  $^{238}\text{U}/^{235}\text{U}$ . Although the salinity was slightly lower than 35 due to the influence of the river water, both the U concentrations and isotopic ratios in the Taiwan Strait were close to those of the open ocean seawater, suggesting the dominance of open ocean input to its U pool. However, the river input might be responsible for the variation of U concentrations and isotopic compositions in the Taiwan Strait. A compilation of U concentrations in the Taiwan Strait and the Jiulong Estuary and Zhujiang Estuary, Xiamen Bay and the northern South China Sea revealed a strong relationship between U concentration and salinity, which suggests the conservative mixing behavior of U in the Taiwan Strait. The U:S ratio obtained in this study ( $0.093 4\pm 0.002 4$ ) is similar to the open ocean ratio, further supporting the dominant influence of open ocean U and the conservative mixing behavior of U in the Taiwan Strait. To better understand U geochemistry in the Taiwan Strait, a multiple endmembers mixing model, including dust deposition, open ocean seawater, river water, and sediment interstitial water, was applied to estimate the contributions of these potential sources. The open ocean seawater contributed

69%–95% of U to the Taiwan Strait, while the contributions of the river water and dust deposition were only 2% and 0.13%, respectively, which reconfirmed the conservative mixing behavior of U and the open ocean source water as the dominant source. The sediment interstitial water was identified as a potential source of U to the Taiwan Strait with a possible contribution of 3%–29%, consistent with previous investigations based on radium isotopes. However, further investigations on U concentrations in the sediment interstitial water are required to precisely determine its input to the overlying seawater in the Taiwan Strait.

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