

Measurements of atmospheric $\Delta^{14}\text{CO}_2$ along the R/V *Xuelong* cruise track from Zhongshan Station (Antarctica) to Shanghai

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ABSTRACT

During January–March 2011, 29 atmospheric $\Delta^{14}\text{CO}_2$ samples were measured along the R/V *Xuelong* cruise track from Zhongshan Station (69.37°S, 76.38°E; Antarctica) to Shanghai. The extent of fossil fuel contamination in the samples was evaluated by comparison with contemporary hemispheric-scale averages of $\Delta^{14}\text{CO}_2$, together with transport analysis and consideration of the $\delta^{13}\text{CO}_2$ levels in the samples. Generally, the $\Delta^{14}\text{CO}_2$ levels along the route south to 40.67°S were lower than the hemispheric-scale average. Circumpolar deep water (CDW) upwelling, stratosphere–troposphere transport (STT), and regional transport associated with the Fukushima nuclear power plant (NPP) accident in Japan on 12 March 2011 were found to have influenced $\Delta^{14}\text{CO}_2$ levels at the sampling sites. In comparison with the atmospheric $\Delta^{14}\text{CO}_2$ level ($45.7 \pm 3.5\text{‰}$ (1σ)) measured in the coastal region of East Antarctica, CDW upwelling caused reduction of $\sim 10\text{‰}$ in $\Delta^{14}\text{CO}_2$ in two cross sections: 62.0°–64.0°S along 74.0°E and 51.7°–54.0°S along 80.5°–84.0°E. Conversely, STT-enhanced $\Delta^{14}\text{CO}_2$ was $\sim 7\text{‰}$ ($\sim 3\text{‰}$) at Zhongshan (around 45.0°S). The linkage was supported by a certain Model-3/CMAQ simulation that indicated dispersion of radionuclides from the Fukushima NPP accident over the East Asian region, which explained the individual high $\Delta^{14}\text{CO}_2$ (52‰) level on 29 March 2011 in Shanghai.

Keywords: atmospheric $\Delta^{14}\text{CO}_2$, *Xuelong* cruise, CDW upwelling dilution, STT, Fukushima NPP accident

1. Introduction

Both fossil fuel emissions and ocean–atmosphere exchange have played critical roles in the declining trend of global atmospheric $\Delta^{14}\text{CO}_2$ since its peak in the 1950s–1960s, which was attributable to the detonation of nuclear bombs. This global-scale trend has been evidenced by the long-term sampling networks established by the University of Heidelberg (Levin et al., 1985) and the unique long-term measurements taken at Baring Head (41.4°S, 174.9°E), New Zealand (Turnbull et al., 2017). Observations have also revealed a negative hemispheric gradient of atmospheric $\Delta^{14}\text{CO}_2$ from south to north since 2000, in contrast to the trend existing

previously (Levin et al., 2010; Graven et al., 2012a). This change in gradient is attributable primarily to the global continuous growth of fossil fuel emissions (mainly in the Northern Hemisphere) and to the oceanic negative input of atmospheric $\Delta^{14}\text{CO}_2$, including the uptake of atmospheric $\Delta^{14}\text{CO}_2$ and the emissions of depleted $\Delta^{14}\text{CO}_2$. The impact of fossil fuel emissions on atmospheric $\Delta^{14}\text{CO}_2$ was estimated at approximately $-9 \pm 1\text{‰ year}^{-1}$ in 1995, exceeding that of the ocean ($-7 \pm 2\text{‰ year}^{-1}$) (Turnbull et al., 2016).

Hydrographic surveys showed that ^{14}C in the surface water of the subtropical ocean had reached near-equilibrium levels with tropospheric $^{14}\text{CO}_2$ by 1995 owing to shallow upwelling (Key et al., 2004), and that this equilibrium subsequently changed to a positive ^{14}C flux from

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the ocean to the atmosphere (Graven et al., 2012b). Contrastingly, large-scale upwelling of ocean deep water at high latitudes causes mixing of $\Delta^{14}\text{CO}_2$ -depleted old water (that is generally sequestered in the deep ocean) with surface water parcels, which results in oceanic uptake of atmospheric $\Delta^{14}\text{CO}_2$ and emission of depleted $\Delta^{14}\text{CO}_2$. Such effects on atmospheric $\Delta^{14}\text{CO}_2$ spread in both the Southern Ocean and the North Pacific Ocean. In Rozanski et al. (1995), observational evidence was reported that indicated the reduction of atmospheric $\Delta^{14}\text{CO}_2$ levels at equatorial sites was due to ocean deep water upwelling during the 1992–1993 El Niño–Southern Oscillation event. It was also proposed that large-scale upwelling of the Circumpolar Deep Water (CDW) in the Southern Ocean could account for the lower level of atmospheric $\Delta^{14}\text{CO}_2$ (by 2–5‰) at Cape Grim (41.0°S, 145.0°E, 95 m a.s.l.), Tasmania, compared with that at equatorial sites. Furthermore, in comparison with the levels at Cape Grim and Neumayer (70.65°S, 8.25°E), Antarctica, Levin et al. (2010) discovered a lower atmospheric $\Delta^{14}\text{CO}_2$ value at Macquarie Island (54.5°S, 158.9°E), again confirming that depletion of atmospheric $\Delta^{14}\text{CO}_2$ in the Southern Ocean is closely associated with CDW upwelling. It has been argued that upwelling of deep ocean water in the North Pacific Ocean influenced the $\Delta^{14}\text{C}$ level of tree rings archived in Washington and Arizona (Jirikowic and Kalin, 1993). The influence of such upwelling is evidenced by the rapid decline of atmospheric O_2/N_2 at around 2000, which occurred in phase with the variation of atmospheric $\Delta^{14}\text{CO}_2$ recorded at La Jolla, California, USA (32.87°N, 117.25°W) (Graven et al., 2012c). The Southern Ocean is the globally dominant region of oceanic impact on atmospheric $\Delta^{14}\text{CO}_2$, with negative ocean–atmosphere gradients of >100‰, whereas the gradients over smaller areas in the North Pacific Ocean are 50‰–100‰ (Turnbull et al., 2016).

In contrast to the abovementioned negative influencing factors, obvious factors with positive influence on atmospheric $\Delta^{14}\text{CO}_2$ during the post-bomb period include cosmogenic production that occurs mainly in the stratosphere, nuclear power plants (NPPs), and the terrestrial biosphere within the boundary layer. Terrestrial respiration might return bomb-related ^{14}C back to the atmosphere (Randerson et al., 2002), and the impact of this process has been estimated at $2 \pm 2\%$ year^{−1} (Turnbull et al., 2016). As in the terrestrial biosphere, subtropical surface water has released bomb-related ^{14}C back to the atmosphere since 1995. However, the substantial global fossil fuel emissions offset the positive input of atmospheric $\Delta^{14}\text{CO}_2$ (Graven et al., 2012b).

Stratosphere–troposphere transport (STT) is the primary pathway via which cosmogenic $\Delta^{14}\text{CO}_2$ is conveyed

to the troposphere. The longer residence time of air masses with less-reduced ^{14}C in the stratosphere than in the troposphere also causes higher stratospheric $\Delta^{14}\text{CO}_2$ levels. The impact of the stratospheric input on the trend of atmospheric $\Delta^{14}\text{CO}_2$ was estimated at approximately $7 \pm 1\%$ year^{−1} in 1995 (Turnbull et al., 2016). At the synoptic scale, STT occurs at the downward branch of a mid- or high-latitude frontogenetic circulation. This process enhances tropospheric ozone, an important trace gas originating partly in the stratosphere that can be traced extensively by other cosmogenic radioactive isotopes, e.g. ^7Be or ^{10}Be (Zheng et al., 2011). However, there is scant observational evidence of the coincident enhancement of $\Delta^{14}\text{CO}_2$ and surface ozone in association with STT.

The main industrial source of atmospheric ^{14}C in the post-bomb period is NPPs, which represent a global impact of approximately 0.5‰–1.0‰ year^{−1} (Turnbull et al., 2016). Simulation has suggested that NPP enrichment might counterbalance at least 20% of the fossil fuel dilution of $\Delta^{14}\text{CO}_2$ at the continental scale (Graven and Gruber, 2011). This counterbalance is expected to increase the uncertainty of the CO_2 budget quantified through measurements of atmospheric $\Delta^{14}\text{CO}_2$ (Kuderer et al., 2018). The magnitude of this offset could be strengthened following an NPP accident, e.g. Chernobyl in the former Soviet Union in 1986 (Buzinny et al., 1997). The Japan Fukushima NPP accident on 12 March 2011 released substantial amounts of $\Delta^{14}\text{CO}_2$ (Xu et al., 2016), and this release is believed to have had regional influence on atmospheric $\Delta^{14}\text{CO}_2$ variations observed on the Korean Peninsula (Park et al., 2015).

Through measurement of short-lived aerosol particle-borne radionuclides (e.g. ^{137}Cs and ^{131}I), it has been proven that the nuclear plume associated with the Japan Fukushima NPP accident could have been transported to eastern parts of China including Taiwan (Hsu et al., 2012) and Heilongjiang Province (Xiangwang et al., 2012). Coincidentally, at the time of the Fukushima NPP accident, the R/V *Xuelong* was traversing the seas to the southeast of China during the final days of a cruise. This provided an opportunity to examine the possible impact of the Fukushima NPP accident on atmospheric $\Delta^{14}\text{CO}_2$ levels in the region.

This study presents atmospheric $\Delta^{14}\text{CO}_2$ levels measured in the marine boundary layer (MBL) along the cruise track of the R/V *Xuelong* during the 27th CHINA National Antarctica Research Expedition (CHINARE). The cruise from Zhongshan Station (Antarctica) to Shanghai (31.36°N, 121.65°E; China) during January–March 2011 passed the coastline of Western Australia and through the islands of Southeast Asia. Most sampling sites along this route were far from the effects of the terrestrial biosphere, particularly those in

Table 1. Reproducibility tests of laboratory $^{14}\text{CO}_2$ measurements.

#Cylinder-bottle. No.	Lab. code	YYYY-MM	FM	$\Delta^{14}\text{CO}_2/\text{‰}$
#4-1	GZ8458	2013.4	0.9749 ± 0.0025	-32.9 ± 2.5
#4-2	GZ8459	2013.4	0.9759 ± 0.0034	-31.9 ± 3.4
#2-1	GZ8460	2013.11	0.8716 ± 0.0031	-135.3 ± 3.1
#2-2	GZ8461	2013.11	0.8710 ± 0.0027	-136.0 ± 2.7
#6-1	GZ8462	2013.11	0.6343 ± 0.0018	-370.8 ± 1.8
#6-2	GZ8463	2013.11	0.6365 ± 0.0018	-368.5 ± 1.8
#4-1	GZ8464	2013.11	0.9787 ± 0.0024	-29.1 ± 2.4
#4-2	GZ8465	2013.11	0.9739 ± 0.0027	-33.8 ± 2.7

the Southern Indian Ocean and in the coastal region of Antarctica, which is considered a proxy of the global atmospheric baseline condition. However, the cruise track also passed through waters near the highly populated regions of the equator and subtropical western Pacific Ocean with exposure to substantial fossil fuel emissions. Additionally, as the sampling campaign was conducted on a moving platform powered by fossil fuel, local contamination of the $\Delta^{14}\text{CO}_2$ measurements was a possibility under certain wind conditions. Therefore, the extent of contamination was evaluated prior to performing the analysis on the $\Delta^{14}\text{CO}_2$ levels measured in the MBL.

2. Air parcel sampling and laboratory measurement

The sampling unit and the stainless steel flasks (1.0 L) used in this study were obtained from Tohoku University, Japan (Zhang et al., 2008). The sampling unit was equipped with a diaphragm pump that directly forces ambient air into the flasks. The sample inlet placed ~ 5 m above the surface of the ship's top forecandle deck was connected to the inlets of the flasks. The forecandle deck was ~ 80 m from the ship's funnel at the stern. All flasks were evacuated in a laboratory before the sampling was conducted. To ensure collection of completely exchanged air, each flask was first flushed by air passing through at a rate of 10 L min^{-1} for ~ 10 min. Then, the flask's outlet was tightened and the air pressure inside the flask started to rise. Once the air pressure reached 0.1 MPa or slightly higher, the inlet was tightened.

The laboratory extraction procedures for CO_2 conversion and graphite involved three principal steps. i) The sample flasks were first introduced to a dry ice-ethanol trap (-90°C) at a fixed flow rate of 120 mL min^{-1} to eliminate water vapour. Then, they were introduced to a liquid nitrogen trap (-196°C) to cryogenically trap and purify the atmospheric CO_2 concentrations. ii) The purified CO_2 was divided into two parts with consideration of the total sample volume. The part with the greater fraction was retained for the $\Delta^{14}\text{CO}_2$ measurements, while

the part with the smaller fraction was used for the $\delta^{13}\text{CO}_2$ measurements. iii) The purified CO_2 was converted to graphite in a sealed glass tube with a zinc, iron, and TiH_2 catalyst using the method of Xu et al. (2007). The synthesised graphite targets were then sent to the NEC 0.5 MeV Accelerator Mass Spectrometer (AMS) at Peking University (China) for ^{14}C analyses. The above steps for CO_2 purification, separation, and target synthesis have been described in detail in previous literature (e.g. Ding et al., 2013). The levels of ^{14}C in CO_2 are expressed as $\Delta^{14}\text{C}$ per mil (‰) in the following equation (Stuiver and Polach, 1977):

$$\Delta^{14}\text{C} = \left[\frac{(^{14}\text{C}/^{12}\text{C})_{\text{samm}}}{(^{14}\text{C}/^{12}\text{C})_{\text{abs}}} - 1 \right] \times 1000 \text{ ‰}$$

where $(^{14}\text{C}/^{12}\text{C})_{\text{abs}}$ is the $^{14}\text{C}:^{12}\text{C}$ ratio of the absolute radiocarbon reference standard corrected for isotopic fractionation and for radiocarbon decay (Stuiver and Polach, 1977). The standards used in this study were oxalic acid standard I and oxalic acid standard II. Here, $(^{14}\text{C}/^{12}\text{C})_{\text{samm}}$ is the $^{14}\text{C}:^{12}\text{C}$ ratio in the sampled material, normalised to a $\delta^{13}\text{C}$ value of 25‰ .

Although the precision and reproducibility of the Accelerator Mass Spectrometer measurements was better than 3‰ (Liu et al., 2007), our repeated laboratory tests for the above extraction and measurement procedures guaranteed better precision. Table 1 lists the results of reproducibility tests for ^{14}C before the treatment of the samples obtained along the *Xuelong* cruise. In the tests, the measured targets were from three steel cylinders (labelled as #2, #4, and #6 in Table 1), which were filled with compressed air with different concentrations of CO_2 provided by the Chinese Academy of Meteorological Sciences. The airflow from the cylinders was directed simultaneously into two 4-L vacuumed glass bottles (labelled as 1 or 2 after the cylinder number in the first column of Table 1) for parallel extraction and measurement. The results for cylinder #4 $\Delta^{14}\text{CO}_2$ measured during two different periods clearly demonstrate that the standard deviations of our measurement system were $<2\text{‰}$, while the $\Delta^{14}\text{CO}_2$ differences for cylinders #2 and #6 were $<2.5\text{‰}$.

Table 2. In situ records, levels of atmospheric $\delta^{13}\text{CO}_2$ and $\Delta^{14}\text{CO}_2$, and statistics of vertical trajectories of the samples.

Sample	Date Yr./Mo./Day	Lat. (°N)	Lon. (°E)	Wind direction	Wind speed (m s ⁻¹)	$\delta^{13}\text{CO}_2$ (‰)	$\Delta^{14}\text{CO}_2$ (‰)	Vertical trajectories (m)
No.							(mean $\pm$ 2 σ)	(Maximum/mean)
1	2011/1/06	-69.37	76.37	—	—	-8.5	51.9 $\pm$ 2.4	3487/2536
2	2011/1/18	-63.99	91	—	—	-8.6	43.1 $\pm$ 2.4	3321/1344
3	2011/1/19	-64	100.6	—	—	—	44.7 $\pm$ 2.4	3038/1086
4	2011/1/20	-66.27	110.47	—	—	-8.6	44.4 $\pm$ 2.4	3031/1306
5	2011/2/26	-69.18	76.31	55°	10.5	-8.5	44.4 $\pm$ 2.4	2246/730
6	2011/2/27	-64.73	74	60–120°	10	-8.8	33.8 $\pm$ 2.4	50/17
7	2011/2/28	-62	74.45	204°	11.7	-8.6	35.7 $\pm$ 2.3	50/9
8	2011/3/1	-57.72	74.95	116°	12	-8.3	45.0 $\pm$ 2.6	90/10
9	2011/3/2	-54	80.52	280°	10	—	35.9 $\pm$ 2.4	155/146
10	2011/3/2	-51.7	83.68	240–120°	10	-8.0	35.8 $\pm$ 2.3	206/64
11	2011/3/3	-48.1	89.4	240°	9.5	-8.7	41.1 $\pm$ 2.4	602/374
12	2011/3/4	-44.9	95.1	203°	11.2	-8.4	48.6 $\pm$ 2.6	1016/518
13	2011/3/5	-40.67	97.1	220°	6.7	-8.6	45.7 $\pm$ 2.4	1228/410
14	2011/3/6	-35.61	101.85	175°	8.8	-8.9	18.4 $\pm$ 2.8	554/318
15	2011/3/17	-31.59	115	260°	6.7	—	5.3 $\pm$ 2.5	3003/1766
16	2011/3/18	-26.83	112.53	202°	5.6	-9.3	41.2 $\pm$ 2.4	2431/1538
17	2011/3/19	-21.23	113.23	187°	6.3	-8.5	36.9 $\pm$ 2.6	2225/1027
18	2011/3/20	-17.1	114.1	190°	5.8	-8.2	42.6 $\pm$ 2.5	233/114
19	2011/3/20	-12.9	114.9	221°	5.1	-9.6	5.3 $\pm$ 2.6	452/125
20	2011/3/21	-7.85	116.04	68°	3.4	-8.9	37.5 $\pm$ 2.7	129/24
21	2011/3/22	-2.4	118.71	61°	6.7	-8.7	34.4 $\pm$ 2.5	112/27
22	2011/3/23	2.36	121.52	211°	3	-9.0	18.0 $\pm$ 3.3	661/84
23	2011/3/24	6.23	126.29	58°	12.9	-8.9	39.6 $\pm$ 5.4	205/28
24	2011/3/25	10.6	127.16	359°	16.4	-8.7	29.7 $\pm$ 2.5	50/21
25	2011/3/26	15.12	127.16	53°	19.5	-9.0	40.7 $\pm$ 2.5	1914/293
26	2011/3/27	19.8	127.02	75°	22.5	—	-28.3 $\pm$ 2.7	3259/1388
27	2011/3/28	25.3	125.99	95°	18.4	—	26.6 $\pm$ 2.6	4057/1698
28	2011/3/28	28.04	124.8	65°	15.9	—	32.5 $\pm$ 2.4	2680/1457
29	2011/3/29	31.36	121.65	50°	12.9	-9.1	52.00 $\pm$ 2.5	509/82

The atmospheric $\delta^{13}\text{CO}_2$ level was measured using a Finnigan Model-251 mass spectrometer operated at the Xi'an Institute of Earth Environment, Chinese Academy of Sciences (Zhou et al., 2014), and the precision of the data is within 0.5‰. Ultimately, six samples failed in the $\delta^{13}\text{CO}_2$ measurements because the fraction volume of purified CO_2 was too small.

3. Results and discussion

3.1. Preliminary results and data evaluation

Levels of atmospheric $\Delta^{14}\text{CO}_2$ and $\delta^{13}\text{CO}_2$, together with sampling information that includes the maximum and mean altitudes of the 96-h backward trajectories for the sampled air masses are listed in Table 2. The sampling time was usually set at local 14:00 to allow collection of well-mixed ambient air. The $\Delta^{14}\text{CO}_2$ levels of the 29 samples listed in Table 2 range from -28.3‰ to 52.0‰ with an average of $33.1 \pm 17\%$ (1 σ). This large variability is

also illustrated in Fig. 1, in which the $\Delta^{14}\text{CO}_2$ levels (excluding the extremely low value of sample #26) vary as a possible function of latitude, i.e. a general trend of decrease of $\Delta^{14}\text{CO}_2$ level as a function of northward latitude is apparent. This trend is most evident between 40°S and 28°N (if $\Delta^{14}\text{CO}_2$ levels of <20‰ are ignored). The trend is generally consistent with reported hemispheric-scale $\Delta^{14}\text{CO}_2$ gradients observed at continental sites (Levin et al., 2010; Graven et al., 2012a).

The variation of the $\Delta^{14}\text{CO}_2$ levels reveals that the sampled air parcels contain not only a certain well-mixed atmospheric level of $\Delta^{14}\text{CO}_2$ but also additional significant inputs that temporally affect the $\Delta^{14}\text{CO}_2$ levels with respect to sampling location. The term ‘well-mixed atmospheric level’ refers to the average status of atmospheric $\Delta^{14}\text{CO}_2$ homogeneously distributed at the large (e.g. hemispheric) scale. This status must be determined prior to evaluation of the magnitude of the extra $\Delta^{14}\text{CO}_2$ input.

Measurement data regarded as representative of the average atmospheric $\Delta^{14}\text{CO}_2$ level in 2011 are scarce.

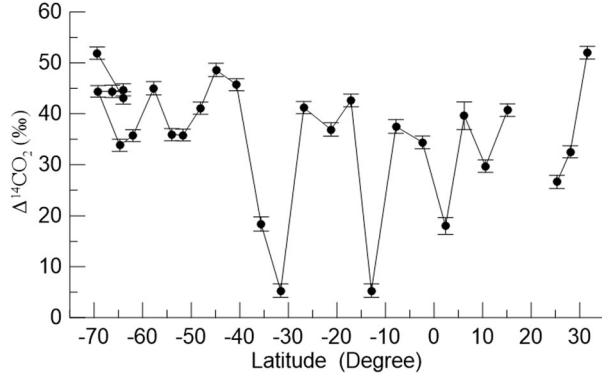


Fig. 1. Variation of atmospheric $\Delta^{14}\text{CO}_2$ level (within 1σ) as a function of northward increasing latitude.

There are only two public sources of such data: one is Jungfraujoch (46.83°N , 7.86°E , 3450 m a.s.l.) in Switzerland (Levin et al., 2013) and the other is Baring Head in New Zealand (Turnbull et al., 2017). Both sites are far from the direct influence of fossil fuel emissions, and their mean atmospheric $\Delta^{14}\text{CO}_2$ level was 39.4‰ (39.3‰ – 39.5‰ , $n=2$) and 42.3‰ (41.1‰ – 43.4‰ , $n=2$), respectively, during the cruise period. In addition, reconstructed average atmospheric $\Delta^{14}\text{CO}_2$ levels of $40 \pm 3\text{‰}$ (1σ) in the Northern Hemisphere in the middle of 2010 and $46 \pm 2\text{‰}$ (1σ) in the Southern Hemisphere in 2011 were presented by Hua et al. (2013). Both values are slightly higher than observed in their respective hemispheres.

Unfortunately, there are no contemporary Antarctic atmospheric $\Delta^{14}\text{CO}_2$ data available for comparison. To make a comparison, the rate of decrease of the level of atmospheric $\Delta^{14}\text{CO}_2$ with time in the Antarctic region was simply assumed constant during 1999–2011. Then, an average level was extrapolated based on data measured at the South Pole station during 1999–2007 (Graven et al., 2012a). The extrapolated level of $\sim 45\text{‰}$ is similar to the level reconstructed by Hua et al. (2013) for the Southern Hemisphere.

Atmospheric $\Delta^{14}\text{CO}_2$ is reasonably sensitive to emissions of ^{14}C -free associated with fossil fuel combustion. Theoretically, those samples with $\Delta^{14}\text{CO}_2$ levels lower than the abovementioned averages could be preliminarily regarded as contaminated by fossil fuel, except for those samples collected over high-latitude oceans where ocean–atmosphere exchange could have negative impact on the atmospheric $\Delta^{14}\text{CO}_2$ level. Conversely, $\Delta^{14}\text{CO}_2$ levels higher than the averages could be attributed to extra $\Delta^{14}\text{CO}_2$ flux from STT, NPP release, or the biosphere. This study found three cases in which the $\Delta^{14}\text{CO}_2$ level was higher than the averages: sample #1 ($51.9 \pm 1.2\text{‰}$ (1σ)) at Zhongshan, sample #12 ($48.6 \pm 1.3\text{‰}$ (1σ)) in the Southern

Ocean around 45°S , and sample #29 ($52.0 \pm 1.3\text{‰}$ (1σ)) in Shanghai.

The influences of local and regional transport on atmospheric $\Delta^{14}\text{CO}_2$ can be investigated using local wind vectors and 3-dimensional backward trajectories, respectively (Draxler and Rolph, 2003). Details of the local wind speed and direction at the time of sampling are presented in Table 2. The 96-h trajectory distributions for all the samples, shown in Fig. 2(a) and (b), are based on the National Centers for Environmental Prediction (NCEP) reanalysis meteorological data (Kalnay et al., 1996). Based on sampling latitude and the distributions of the backward trajectories, the entire cruise track was divided into five segments (regions): East Antarctica coastline, remote Southern Ocean zone, Western Australia coastline, Southeast Asia intertropical convergence zone (ITCZ), and subtropical western Pacific.

East Antarctica coastline (samples #1–#5). The backward trajectories demonstrate that the transport paths of the sampled air masses started from the remote Antarctic continent and that the highest altitude of the trajectories could be traced to over 3 km. The average $\Delta^{14}\text{CO}_2$ level of this sample group of $45.7 \pm 3.5\text{‰}$ (1σ) is in agreement with the extrapolated average for the South Pole station mentioned above. Except for sample #1, the $\Delta^{14}\text{CO}_2$ levels of the samples in this group are reasonably close to 44‰ , i.e. slightly higher than the contemporary average (42.3‰) at Baring Head (Turnbull et al., 2017). Geographically, the positions of sampling sites #1 and #5 are reasonably close; however, sample #1 was taken at Zhongshan, a continental site, while sample #5 was obtained at the R/V *Xuelong*. The cause of the highest $\Delta^{14}\text{CO}_2$ level (51.9‰) in sample #1 is associated with STT. The 96-h-average altitude of the source air parcel was 2536 m (maximum: $\sim 3500\text{ m}$) for sample #1, whereas the corresponding altitude for sample #5 was only 730 m. The contribution of STT to the enhancement of $\Delta^{14}\text{CO}_2$ in sample #1 is investigated further in section 3.2.3.

Remote Southern Ocean zone (samples #6–#14). The backward trajectories show that all the transport paths of the air masses sampled in this group started from the remote southern Indian Ocean. However, the MBL $\Delta^{14}\text{CO}_2$ levels of these samples are not distributed homogeneously, i.e. sample #14 displays an obviously lower level (18.4‰), far below the abovementioned average for the Southern Hemisphere. A possible reason for this much lower level is the influence of local fossil fuel emission. At the time sample #14 was taken, the record of local wind vectors (Table 2) suggests that conditions were favourable for contamination by local transport of the exhaust plume of the R/V *Xuelong*. Ignoring this obviously contaminated sample, the average $\Delta^{14}\text{CO}_2$ level of this sample group of $40.2 \pm 5.6\text{‰}$ (1σ) is comparable to

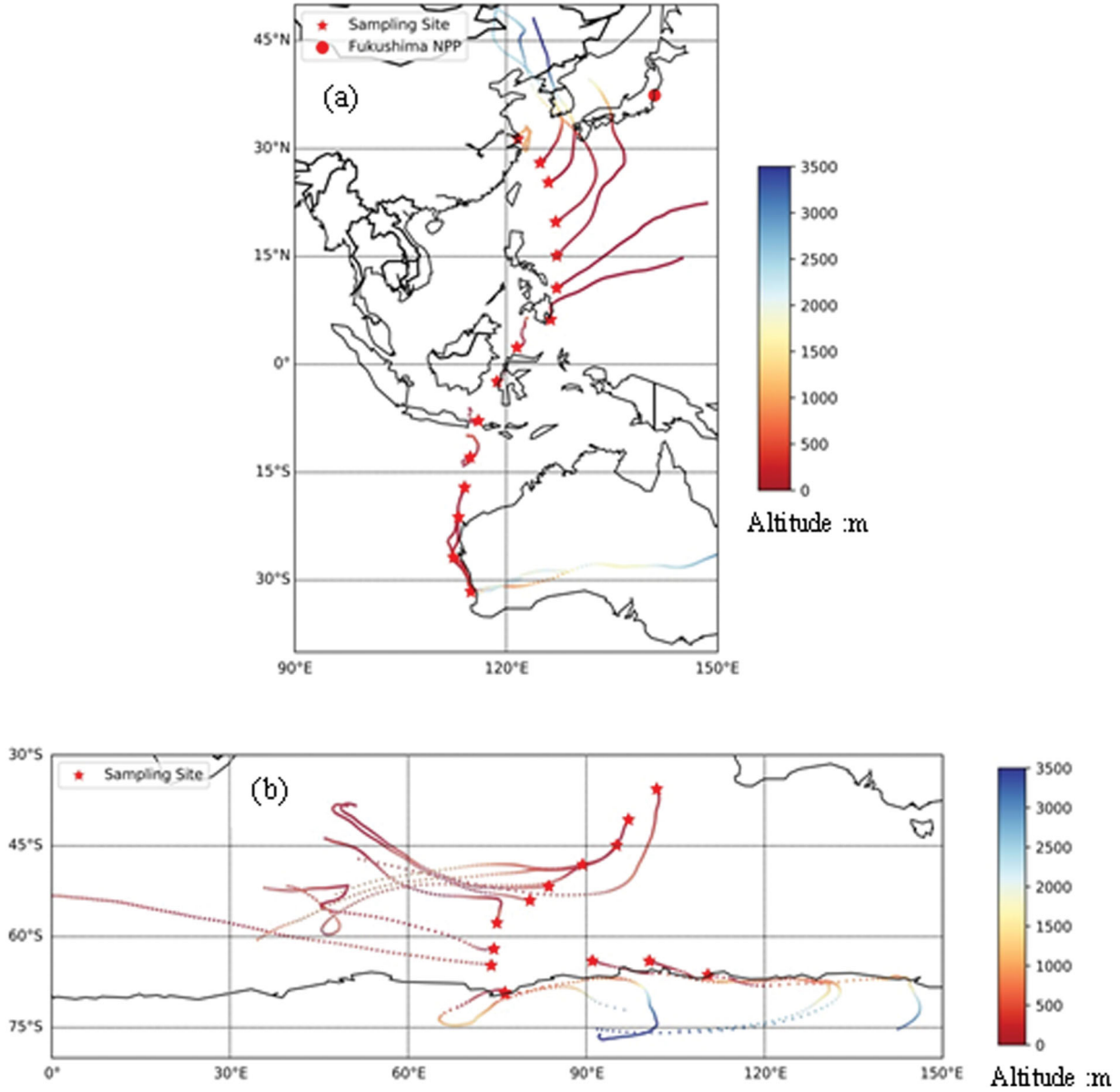


Fig. 2. Three-dimensional 96-h backward trajectories of the sampled air parcels: (a) samples #15–#29 covering the Western Australia coastline, southeast Asia ITCZ, and subtropical western Pacific, (b) samples #1–#14 covering the Southern Ocean and the coastal region of East Antarctica.

but slightly lower than the average value (42.3‰) at Baring Head (Turnbull et al., 2017).

The samples in this group with the next lowest levels of $\Delta^{14}\text{CO}_2$ ($\sim 33\text{‰}$ – 35‰) are #6, #7, #9, and #10 with values $\sim 5\text{‰}$ lower than the group average and $\sim 10\text{‰}$ lower than the average of the coastal region of East Antarctica; the level in sample #11 is $\sim 4\text{‰}$ lower. The cause of such low levels can be attributed to CDW upwelling, the mechanism of which is elucidated in section 3.2.2. Similar to the case of sample #1, the higher altitudes of the backward trajectories for samples #12

and #13 is coincident with their higher $\Delta^{14}\text{CO}_2$ levels. This indicates the possibility of STT contribution, especially for sample #12, in which the $\Delta^{14}\text{CO}_2$ level is $\sim 3\text{‰}$ higher than the average of the coastal region of East Antarctica.

Western Australia coastline (samples #15–#18). The backward trajectories suggest that most of the air parcels sampled (except #18) in this group travelled westward across Australia before turning northward prior to the sampling. Atmospheric $\Delta^{14}\text{CO}_2$ levels of this group display high variability: the levels of samples #16 (41.2‰)

and #18 (42.6‰) are reasonably close to the value measured at Baring Head (Turnbull et al., 2017), while the level in sample #17 is $\sim 5\%$ lower than the Southern Hemisphere average. The $\Delta^{14}\text{CO}_2$ level of sample #15 is the lowest (5.3‰) in this group, and it was obtained at Freemantle, a port near Perth in Western Australia. This low level of $\Delta^{14}\text{CO}_2$ is consistent with an urban ambient environment, which also indirectly supports that all the procedures such as the in situ sampling, laboratory extraction, and measurement for atmospheric $\Delta^{14}\text{CO}_2$ in this study are valid. It is noted that the atmospheric $\Delta^{14}\text{CO}_2$ levels of this group are generally not sensitive to variation of the vertical altitudes of the trajectories. There is no qualitative correspondence between the average height of the vertical backward trajectories and the levels of atmospheric $\Delta^{14}\text{CO}_2$.

Southeast Asia ITCZ (samples #19–#22). Samples #19–#22 are grouped into the ITCZ, while samples #23 and #24 are excluded because they were collected in areas influenced primarily by the East Asian winter northwesterly monsoon, as shown by the backward trajectories. The principal features of the trajectories in this region are that their horizontal coverage is very short and that their vertical transport path generally occurs no higher than 50 m above sea level. These characteristics indicate considerable potential for exposure of the sampled air parcels to fuel combustion emissions from other moving vessels or from the nearby continent. The $\Delta^{14}\text{CO}_2$ levels in samples #19 (5.3‰) and #22 (18.0‰) are much lower than the abovementioned average, and even though the levels of samples #20 (37.5‰) and #21 (34.4‰) are higher, they are also lower than the abovementioned average. Therefore, it can be concluded that the sampled air parcels in this group were highly contaminated by fossil fuel combustion emissions.

Subtropical western Pacific (samples #23–#29). The feature of air mass transport in this region is that monsoonal northeasterly winds were dominant in the backward trajectories except for sample #29. Moreover, a tropical cyclone with high wind speeds developed during 25–28 March (Table 2). Only samples #23 and #25 in this group have $\Delta^{14}\text{CO}_2$ levels close to the stated average. Although the air mass of sample #24 originated in the remote Pacific region, the $\Delta^{14}\text{CO}_2$ level is still $\sim 10\%$ lower than the above average.

The largest variability of MBL $\Delta^{14}\text{CO}_2$ level was found in this group. Sample #26 has the lowest level (-28.3%) among all the samples, and it was obtained under strong wind conditions. Although the increase of ocean–atmosphere exchange coefficients with the rise of wind speed might strengthen CO_2 flux exchange between the ocean and the atmosphere, the lowest $\Delta^{14}\text{CO}_2$ level here cannot be attributed to oceanic release because near-equilibrium

conditions of $\Delta^{14}\text{CO}_2$ between the ocean surface and the troposphere in subtropical areas have been established since 1995 (Key et al., 2004). The coral $\Delta^{14}\text{C}$ level is $\sim 40\%$ in this ocean region (Glynn et al., 2013). It is considered that fossil fuel combustion is the main reason for the low $\Delta^{14}\text{CO}_2$ levels here, particularly the extremely low level in sample #26. Similar extreme cases have also been reported for atmospheric $\Delta^{14}\text{CO}_2$ samples obtained at other continental sites (Turnbull et al., 2011; Niu et al., 2016).

The average atmospheric $\Delta^{14}\text{CO}_2$ level of the group (excluding sample #26) is $34.2 \pm 10.2\%$ (1σ) ($n=8$). There is no general qualitative correlation between the measured $\Delta^{14}\text{CO}_2$ levels and the altitudes of the backward trajectories, even though influence from the maximum seasonal STT in spring is expected. Additionally, the radioactive plume of the Fukushima NPP accident, which occurred on 12 March 2011, was found to have reached the sampling area by 26 March (Hsu et al., 2012). However, no enhancement of the $\Delta^{14}\text{CO}_2$ level was detected except in sample #29. The influence of the Fukushima NPP accident on sample #29 is investigated in section 3.3.4.

3.2. Mechanisms affecting the variations of atmospheric $\Delta^{14}\text{CO}_2$

3.2.1. Fossil fuel combustion. Approximately one third of the samples collected in this study were contaminated by fossil fuel combustion emissions, and the level of contamination was particularly obvious in sample #14 and at subsequent sites. Local fossil fuel contamination is likely to occur when wind conditions are favourable for conveying the engine exhaust gases of the vessel to the sampling site, as in the case of sample #14, when the local wind direction (175°) was consistent with the navigation direction of the ship. However, consistency in the directions of the local wind vector and ship navigation is not the only prerequisite for determining local contamination of $\Delta^{14}\text{CO}_2$ data, e.g. as in the case of samples #16–#18. The detection of levels of MBL $\Delta^{14}\text{CO}_2$ lower than the hemispheric average is the ultimate evidence of contamination attributable to fossil fuel combustion. This is because ocean–atmosphere exchange in the subtropical oceans (unlike the high-latitude oceans) has positively influenced atmospheric $\Delta^{14}\text{CO}_2$ during the post-bomb period (Key et al., 2004; Graven et al., 2012b).

Regional fossil fuel contamination is also highly possible for those samples collected in the Southeast Asia ITCZ and the subtropical western Pacific regions for the following two reasons illustrated by the three-dimensional trajectories: (i) regional-scale atmospheric contamination should be widespread in the area of the ITCZ because

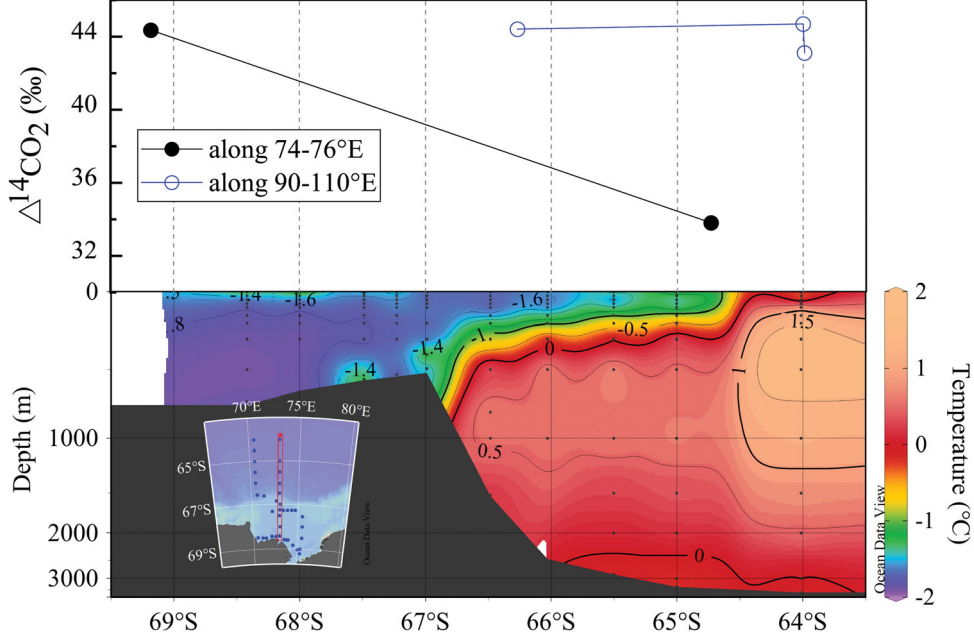


Fig. 3. (Upper) MBL $\Delta^{14}\text{CO}_2$ level from 68.0°S to 64.0°S and (lower) the vertical cross section of in situ measured ocean temperature isotherms along 74.0°E. The red rectangle in the inset of the lower panel; represents the R/V *Xuelong* track positions, along which the vertical section of water temperature contours is plotted.

emissions tend to be transported within the boundary layer, and (ii) all the sampling sites in the western Pacific region, where the regional background levels of atmospheric $\Delta^{14}\text{CO}_2$ are obviously lower than the hemispheric average, were downwind of the winter monsoonal north-easterly winds (Park et al., 2015).

Sample contamination by fossil fuel combustion emissions can also be inferred by comparison of the $\delta^{13}\text{CO}_2$ levels in Table 2. For example, the $\delta^{13}\text{CO}_2$ level of sample #14 is the lowest (-8.9‰) in its group, which is consistent with the $\Delta^{14}\text{CO}_2$ level. Similar cases are evident in samples #19 and #22. The lowest $\delta^{13}\text{CO}_2$ level (-9.1‰) of sample #29 is consistent with the actual urban environment; however, it does not match the highest atmospheric $\Delta^{14}\text{CO}_2$ level, which is analysed in section 3.3.4.

3.2.2. Lower $\Delta^{14}\text{CO}_2$ associated with CDW upwelling in the Southern Ocean. Ignoring samples #6, #7, and #9–#11, the average level of atmospheric $\Delta^{14}\text{CO}_2$ of the other eight samples acquired at locations south of 35.0°S is $46.0 \pm 2.9\text{‰}$ (1σ). The $\Delta^{14}\text{CO}_2$ level in samples #6, #7, #9, and 10 is $\sim 10\text{‰}$ and that in sample #11 is $\sim 5\text{‰}$ lower than the average. The factors causing these lower MBL $\Delta^{14}\text{CO}_2$ levels cannot be attributed to fossil fuel emissions for the following reasons. (i) The backward trajectories indicate that the source of the air parcels is the pristine region of the Southern Ocean, and local wind vectors do not support the transport of local engine

exhaust gases to the sampling site. (ii) The magnitude of the reduction of $\Delta^{14}\text{CO}_2$ in samples #6, #7, #9, and #10 is $\sim 10\text{‰}$, which is clearly different from that of samples obviously contaminated by fossil fuel combustion emissions (i.e. $>20\text{‰}$). (iii) The respective spatial distributions of samples #6–#7, and #9–#11 are reasonably continuous in the mesoscale. (iv) The corresponding $\delta^{13}\text{CO}_2$ levels of the samples do not support contamination by fossil fuel emissions as clearly as shown by sample #14; instead, the highest $\delta^{13}\text{CO}_2$ level (-8.0‰) is detected in sample #10.

Following speculations on the observed lower level of atmospheric $\Delta^{14}\text{CO}_2$ (Rozanski et al., 1995; Levin et al., 2010), we propose that the above $\sim 10\text{‰}$ lower level of MBL $\Delta^{14}\text{CO}_2$ in the Southern Ocean is associated with CDW upwelling. Upwelling of CDW is related to the global ocean overturning circulation in which Antarctic bottom water (AABW) and the southward-penetrating North Atlantic deep water (NADW) in the Antarctic circumpolar current (ACC) are involved. In simple terms, as the deepest, densest water of the world ocean, AABW, together with less-dense bottom water from the North Pacific and Indian Ocean, is mixed upward into the NADW, which is least dense but warmest and most saline, forming a new water type (i.e. the CDW) in the ACC (Reid and Lynn, 1971). The Southern Ocean westerly winds and the resultant net Ekman drift intensify the northward transport of warmer upper-ocean waters. This

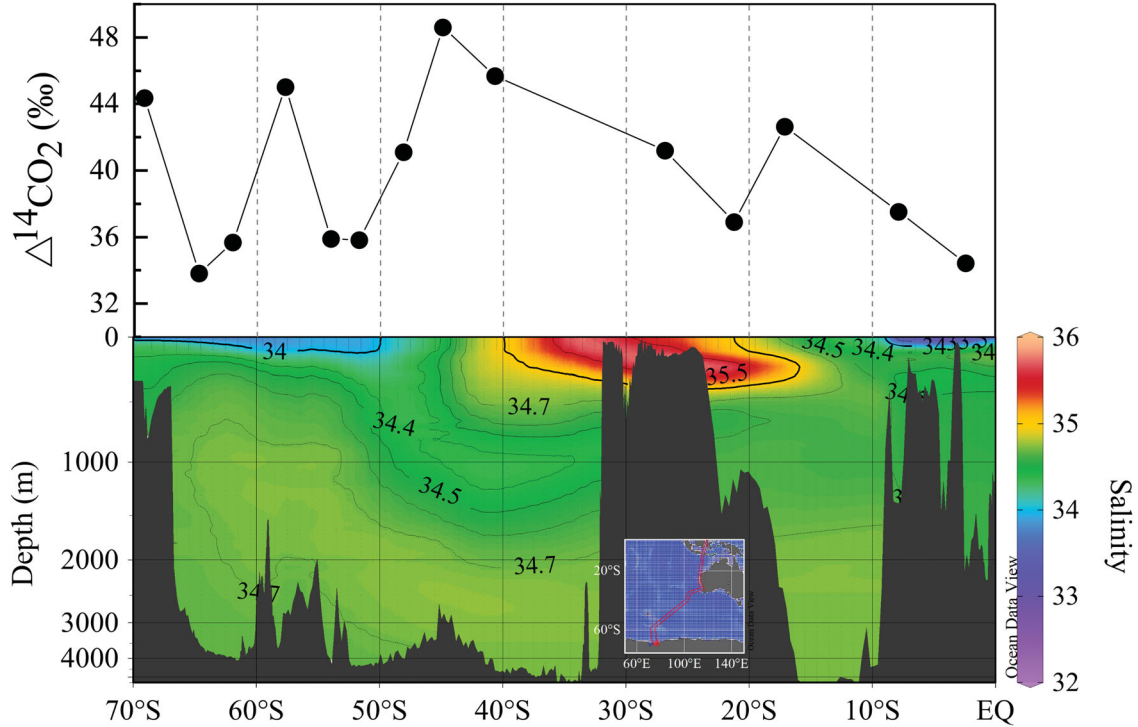


Fig. 4. (Upper) Spatial variation of MBL $\Delta^{14}\text{CO}_2$ from $\sim 70.0^\circ\text{S}$ to the equator along the *Xuelong* cruise track during the 27th CHINARE and (lower) the corresponding vertical cross section of ocean water salinity contours. The red lines in the inset of the lower panel represent the R/V *Xuelong* track, along which the vertical section of the salinity contours is plotted.

wind-driven surface convergence and divergence, together with ocean internal waves and turbulence generated by the interplay between the currents and bottom topography, are favourable for vertical mixing in the ocean interior. Vertically, the CDW in the ACC lies above the denser bottom water and below the less-saline Antarctic surface water, and it is generally at depths of several hundred to thousands of metres below the ocean surface (Mantyla and Reid, 1983). Principally, the CDW exhibits warm (temperature: $0^\circ\text{--}2.0^\circ\text{C}$) and saline (salinity: 34.50–34.75 psu) characteristics. The strong circumpolar westerly winds are favourable for enhancing MBL $\Delta^{14}\text{CO}_2$ dilution through acceleration of the flux of ocean–atmosphere exchange.

There is a large fraction of CDW in the Southern Ocean and it is estimated to account for $\sim 55\%$ in the Indian Ocean (Pu et al., 2002). Such a high fraction of CDW is assumed to bring depleted ^{14}C up to the upper-ocean waters. Approximately, the level of ^{14}C of dissolved inorganic carbon in the Southern Ocean is -165% and it varies around -100% in the upper-ocean waters (Graven et al., 2012b). Consequently, the depleted ^{14}C tends to be emitted into the atmosphere under favourable conditions of ocean–atmosphere exchange.

The reduced $\Delta^{14}\text{CO}_2$ level in samples #6 and #7 is investigated first. Upwelling of CDW is noted on the section of the cruise track along 74.0°E from sampling site #6 (64.73°S) to #7 (62.0°S). Figure 3 (lower) shows the vertical profile of ocean water temperature measured in situ along $\sim 74.0^\circ\text{E}$ between 64.0°S and 68.0°S . The warm CDW (i.e. 1.5°C) is evident 200 m beneath the ocean surface.

Identification of CDW upwelling here is supported by in situ ocean survey data of N_2O . Jixia (2012) reported high concentrations of N_2O at depths within 1000 m of the ocean surface at 65.5°S and 64.0°S in February 2011; the levels were close to that of the deep water (below 2000 m) in the southern Indian Ocean. Thus, the enhanced levels of oceanic N_2O are evidence of CDW.

The reduction of $\Delta^{14}\text{CO}_2$ levels in samples #6 and #7 is also consistent with the elevation of CO_2 partial pressures ($p\text{CO}_2$) in the upper-ocean water near $62.0^\circ\text{--}65.5^\circ\text{S}$ (Yuanhui et al., 1997; Gao, 2002). Chen et al. (2014) proposed a link between the elevated $p\text{CO}_2$ and CDW upwelling found in this region. Our lower MBL $\Delta^{14}\text{CO}_2$ levels independently support this inferred connection.

It appears that the area to the northeast of Prydz Bay, where samples #2–#4 were collected, is free of the

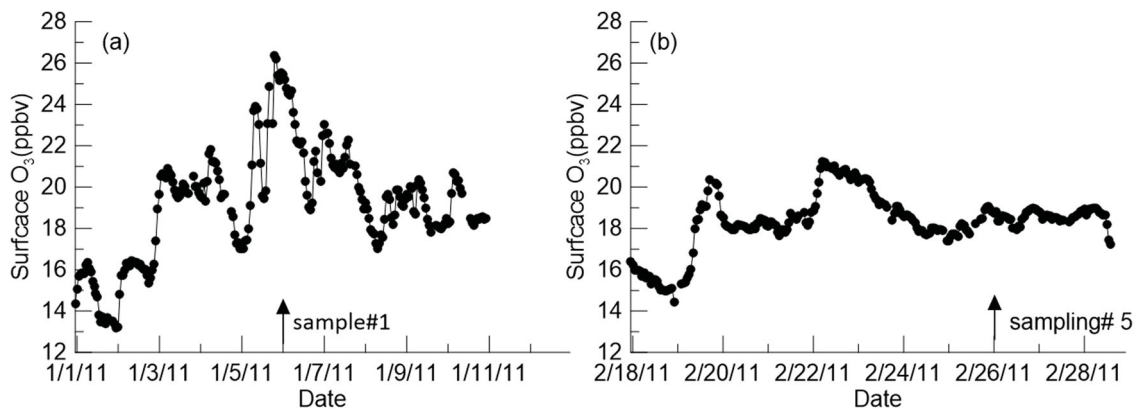


Fig. 5. Hourly averages of surface ozone at Zhongshan corresponding to date of atmospheric $\Delta^{14}\text{CO}_2$ sampling: (a) sample #1 on 6 January 2011 and (b) sample #5 on 26 February 2011.

influence of CDW upwelling, even though samples #2 and #3 were collected near 64.0°S . A hydrographic survey found a transition zone between the warm waters outside the Antarctic continental shelf and the cold waters on the continental shelf near 64.0°S to the east of 83.0°E , and a zonal thermohaline front maintains the relatively stable water parcels (Minjian et al., 1995). The waters in this region comprise mainly cold water from the West Ice Shelf and the Shackleton Ice Shelf, and they are not obviously affected by CDW upwelling.

The reduced $\Delta^{14}\text{CO}_2$ level in samples #9 and #10 and its association with CDW upwelling is also investigated. Figure 4 presents a vertical profile of World Ocean Atlas salinity along the R/V *Xuelong* cruise track, i.e. from Zhongshan to the equator via the coast of Western Australian with an overview of the distribution of CDW upwelling in the region 50.0° – 65.0°S . It should be noted that the spatial resolution of the World Ocean Atlas data is much lower than that of the in situ survey shown in Fig. 3. Large-scale upwelling of deep water is evident at latitudes south of 50.0°S , characterised by the 34.5 psu salinity contour that protrudes up to $\sim 200\text{ m}$ below the ocean surface. Previous studies have measured high levels of $p\text{CO}_2$ that correspond to this regional CDW upwelling (Poisson et al., 1993; Metzl et al., 1999; Zhang et al., 2017). Moreover, the horizontal gradients of those high levels of $p\text{CO}_2$ display mesoscale features attributable to the nearby Kerguelen Plateau (Poisson et al., 1993). Thus, the reduction of the $\Delta^{14}\text{CO}_2$ level in samples #9 and #10 is considered linked to CDW upwelling. Although weakened, this influence remains identifiable in sample #11 at 48.0°S .

3.2.3. Stratosphere–troposphere transport. The vertical evolution of the backward trajectories suggests that sample #1 contains more fractions of higher-altitude air parcels that are more easily influenced by STT. The

influence of air masses from the stratosphere can be characterised by a rapid increase of ozone concentration in the ambient air that is almost free of local photochemical production.

The STT contribution to the high $\Delta^{14}\text{CO}_2$ level in sample #1 is evidenced by continuous measurements of surface ozone at Zhongshan (Wang et al., 2011). Figure 5 shows a comparison of hourly average surface ozone between the two periods covering the dates of collection of samples #1 and #5. It is clear that the high level of ozone ($\sim 26\text{ ppbv}$) on 6 January is related to a rapid transport process that started on 5 January, rather than to local photochemical production. Conversely, the surface ozone level corresponding to sample #5 is very stable, i.e. the concentration varies only slightly from a level of 18 ppbv . The exact coincidence between the high level of atmospheric $\Delta^{14}\text{CO}_2$ in sample #1 and the high level of surface ozone at Zhongshan indicates the occurrence of STT, which is reflected by the vertical evolution of the backward trajectory. For stations on the Antarctic coast, e.g. Neumayer, stratospheric air mass intrusions in late summer/early autumn have been proven by long-term measurements of $^{10}\text{Be}/^7\text{Be}$ in surface aerosol particles (Elsässer et al., 2011). The degree of consistency between elevated levels of surface ozone, $\Delta^{14}\text{CO}_2$, and vertical backward trajectories indicates that summer STT is also common at Zhongshan.

The influence of STT input is also discriminative in terms of the variation of $\Delta^{14}\text{CO}_2$ level in samples #12 and #13, which is also in accord with the climatology of the mid-latitude quasi-permanent STT. The vertical evolutions of the backward trajectories indicate that there could have been possible influence from STT flux in the samples collected along the Western Australia coastline and in the subtropical western Pacific; however, such flux would be very weak because of the lower latitudes of the sampling sites.

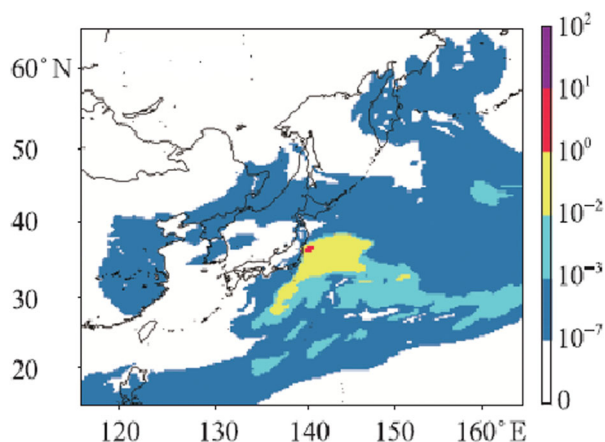


Fig. 6. Distribution of depositional radioactive particles (A25J) from the Fukushima NPP accident at 09:00 (UTC) on 29 March 2011 simulated by Model-3/CMAQ (Xiangwang et al., 2012).

3.2.4. Fukushima NPP accident. The atmospheric $\Delta^{14}\text{CO}_2$ level of the final sample (#29 at Shanghai) is 52‰, which appears atypical for an urban site where the sampled air mass should be exposed to more fossil fuel emissions in comparison with the MBL. Based on the 96-h backward trajectories, this high level cannot be attributed to positive input by STT.

We considered the possibility of influence on sample #29 by the release of $\Delta^{14}\text{CO}_2$ from the Qinshan NPP (30.44°N, 120.96°E), which is located ~ 100 km south of the sampling site in Shanghai. Earlier research has revealed that the $^{14}\text{CO}_2$ emission factor of the Qinshan NPP is low (Graven and Gruber, 2011). Moreover, the three pressurised water reactors at the Qinshan site release ^{14}C mainly in the form of $^{14}\text{CH}_4$. It has been found that the ^{14}C specific activity reaches background levels within 6.5 km of the Qinshan NPP (Wang et al., 2012). Furthermore, the regional meteorological conditions prevailing one day before and on the actual sampling day did not support the possibility of a plume from the Qinshan NPP travelling northward towards Shanghai. In fact, the 24-h forward trajectory of the atmosphere initialised at Qinshan confirmed southward movement of the plume from the morning of 28 March and this transport pattern persisted for more than 30 hours. Therefore, it is difficult to infer a connection between the elevated atmospheric $\Delta^{14}\text{CO}_2$ level in sample #29 and the Qinshan NPP.

We speculate that it is highly possible that the enhancement was associated with the $\Delta^{14}\text{CO}_2$ release associated with the Fukushima NPP accident. The ^{14}C released from the Fukushima NPP was mainly in the form of $\Delta^{14}\text{CO}_2$ (Xu et al., 2016). Measurements of short-lived aerosol particle-borne radionuclides (e.g. ^{137}Cs and ^{131}I) confirmed that the radioactive plume

experienced global circulation and reached Guiyang (Southwest China) on 24–31 March 2011 (Wan et al., 2014) and Taiwan Island on 25–27 March 2011 (Hsu et al., 2012). Furthermore, the transport and dispersion characteristics of the radioactive plume that originated from the Fukushima NPP accident were simulated through A25J, a tracer of the accumulation mode of fine aerosol particles in the third-generation air quality modelling system/community multiscale air quality (Model-3/CMAQ) (Xiangwang et al., 2012). In the simulation, A25J was released only during 12–30 March at the site of the Fukushima NPP accident, and the transport dispersion process was nudged by the NCEP reanalysis meteorological data (Kalnay et al., 1996). The simulated relative distribution of A25J (unit: %) over East Asia at 09:00 (UTC) on 29 March 2011 is shown in Fig. 6 (Xiangwang et al., 2012). It can be seen that A25J from the Fukushima NPP was transported to southeastern parts of China, including Shanghai, during 29–30 March 2011. The shaded area indicating higher levels of A25J in the simulation corresponds exactly to the high level of atmospheric $\Delta^{14}\text{CO}_2$ sampled at Shanghai, indicating a link between the enhanced level of atmospheric $\Delta^{14}\text{CO}_2$ in sample #29 and the regional dispersion transport of $\Delta^{14}\text{CO}_2$ released by the Fukushima NPP accident. The simulation also indicates that the released $\Delta^{14}\text{CO}_2$ crossed the Korean Peninsula, as proposed by Park et al. (2015).

4. Concluding remarks

This study analysed the levels of atmospheric $\Delta^{14}\text{CO}_2$ measured along the cruise track of the R/V *Xuelong* from near Zhongshan Station (Antarctica) to Shanghai (China) in 2011, and the mechanisms influencing the variability of $\Delta^{14}\text{CO}_2$ were investigated. Results showed extensive contamination associated with fossil fuel combustion emissions, especially in samples collected in the Southeast Asia ITCZ and subtropical western Pacific regions, where levels of atmospheric $\Delta^{14}\text{CO}_2$ were generally lower than contemporary hemispheric averages. However, in the southernmost Indian Ocean north to 40.67°S, MBL $\Delta^{14}\text{CO}_2$ was $41.5 \pm 4.9\text{‰}$ (1σ) ($n=12$), i.e. similar to that measured at Baring Head (41.4°S, 174.9°E) in New Zealand. The average level of atmospheric $\Delta^{14}\text{CO}_2$ was $45.7 \pm 3.5\text{‰}$ (1σ) ($n=5$) in the coastal region of East Antarctica, which is consistent with the reconstructed value $46 \pm 2\text{‰}$ (1σ) for the Southern Hemisphere (Hua et al., 2013). In addition, following specific findings were derived.

In comparison with the level of atmospheric $\Delta^{14}\text{CO}_2$ in the coastal region of East Antarctica, the MBL $\Delta^{14}\text{CO}_2$ level in the southern Indian Ocean was approximately -10‰ lower owing to the impact of CDW upwelling.

This influence was highlighted by two cross sections: one at 62.0°–64.0°S along 74.0°E, just outside of Prydz Bay, Antarctica, and the other at 51.7°–54.0°S along 80.5°–84.0°E, near the Kerguelen Plateau.

STT input of atmospheric $\Delta^{14}\text{CO}_2$ was evidenced by the temporal elevation of surface ozone in the case of the sampling at Zhongshan. The STT contribution was estimated to enhance the level of $\Delta^{14}\text{CO}_2$ by $\sim 7\text{‰}$ when compared with the $\Delta^{14}\text{CO}_2$ level in the coastal region of East Antarctica. Similar enhancement of $\sim 3\text{‰}$ was detected in another case around 45.0°S.

The Fukushima NPP accident in Japan on 12 March 2011 was linked to the individual enhancement of atmospheric $\Delta^{14}\text{CO}_2$ ($\sim 52\text{‰}$) in the sample obtained at Shanghai. This link was supported by a Model-3/CMAQ simulation, in which the regional dispersion transport of the released aerosol particle-borne radionuclide tracer from the Fukushima NPP was shown to have spread over East Asia including the Shanghai region.

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Disclosure statement

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