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#### Deep-Sea Research I

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## Ventilation time and anthropogenic CO<sub>2</sub> in the South China Sea based on CFC-11 measurements



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#### ARTICLE INFO

# Article history: Received 7 June 2015 Received in revised form 15 August 2016 Accepted 19 August 2016 Available online 23 August 2016

Keywords: Anthropogenic CO<sub>2</sub> CFCs TTD South China Sea Mean age

#### ABSTRACT

The South China Sea (SCS) is the largest semi-enclosed marginal sea in Southeast Asia, and is bounded by the Asian continent, Philippine Archipelago, and Great Sunda Islands. Due to the wide shelves on its northwestern and southern ends as well as the presence of numerous islets, atolls and reefs, the average depth of the SCS is only 1350 m. In this study we used measurements of the transient tracer CFC-11 from the SCS to calculate ventilation time-scales and the concentration of anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) based on the transit time distribution. The CFC-11 concentrations decreased consistently with depth and the deep and bottom water in the SCS had a CFC-11 value close to the detection limit (0.01 pmol  $kg^{-1}$  or 0.5 ppt). The ventilation times (mean ages) for the deep and bottom water column were  $\sim 500-600$  years, and based on the mean age profiles the southern part of the intermediate SCS water was older than the northern part. The ventilation time distribution was in agreement with the existence of mean annual cyclonic circulation in the SCS. The mean column inventory of Cant in the northern SCS was 28.9 mol C m $^{-2}$  (error range (ER): 22.8–35.6 mol C m $^{-2}$ ), while in the southern SCS it was 28.4 mol m $^{-2}$ (ER:  $21.9-35.2 \text{ molCm}^{-2}$ ). The total  $C_{\text{ant}}$  inventory was estimated to be 1 Pg C (ER: 0.8-1.3 Pg C, referenced to the year 2011), suggesting that the SCS stores less Cant then the adjacent seas. The vertical diffusivity was estimated to be  $2-4.6 \times 10^{-4}$  m<sup>2</sup> s<sup>-1</sup> in the SCS based on the "transient steady state". The upwelling was estimated as 13–34 myr<sup>-1</sup>. The high diffusivity was probably due to the strong internal tide, while the strong upwelling was due to the persistent counterclockwise (cyclonic) circulation.

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#### 1. Introduction

The South China Sea (SCS) is the largest semi-enclosed marginal sea in Southeast Asia and is bounded by the Asian continent, Philippine Archipelago, and Great Sunda Islands. The area of the SCS is approximately  $3.5\times 10^6~\rm km^2$  and the maximum depth is 5377 m in the Manila Trench (Morton and Blackmore, 2001). However, as a result of the wide shelves on the northwestern and southern ends as well as the presence of numerous islets, atolls and reefs, the average depth of the SCS is only 1350 m (Chen et al., 2006). The SCS connects with the East China Sea through the shallow Taiwan Strait (50 m deep). The Mindoro Strait ( $\sim$ 420 m) and the Balabac Strait ( $\sim$ 100 m), which are narrow and shallow

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passages located in the north and south of Palawan Island, connect the SCS and the Sulu Sea. The SCS primarily exchanges the most water with the Pacific through Luzon Strait, where the deepest sill is around 2400 m (Qu et al., 2006). Hence, the water exchange between the SCS and the Pacific Ocean through the Luzon Strait plays an important role in the SCS circulation, heat and salt budgets (Qu et al., 2009).

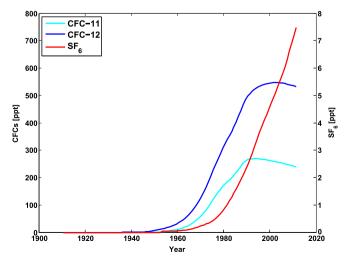
The seasonally reversing monsoon is the key driving force for the upper circulation in the SCS, and the dynamics of flow in the SCS are relatively complicated owing to the variable atmospheric forcing and complex geometry (Chen et al., 2004). The intermediate water ( $\sim 500$  m), lies in the deeper layer and cannot be influenced by the monsoon. The exchange of intermediate water takes place mainly in the Luzon Strait, while some subthermocline waters are exchanged with the Sulu Sea owing to the surrounding topography (Wyrtki, 1961; Broecker et al., 1986; Tessler et al., 2010). Li and Qu (2006) demonstrated that, based on the oxygen data, the intermediate water flows out to the Pacific Ocean.

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Tian et al. (2009)) noted that the SCS intermediate water flows out to the Pacific Ocean based on direct LADCP measurements near the Luzon Strait. Generally, there is a "sandwich" structure of the water flow pattern in the SCS (Qu et al., 2002). The upper and the deep waters flow into the SCS while the intermediate water flows out to the Pacific. However, the depth of the intermediate outflow is still controversial (Chen et al., 2001; Li and Qu, 2006; Tian et al., 2009). For the deep SCS with a depth greater than  $\sim$ 2000 m, there is no direct water exchange pathway to the adjacent oceans (Qu et al., 2006). The properties of the deep water in the SCS (e.g., salinity, temperature) are rather homogeneous and very similar to those of the Pacific at depths around 2400 m (Qu, 2002). This means that the deep SCS water is influenced strongly by the Pacific water, which flows westward around the sill depth of the Luzon Strait and sinks down to the deep SCS (Wyrtki, 1961), with the overflow transport estimated to be 0.7-3.0 Sv (Wang et al., 2011). To compensate for this descending movement, unsurprisingly, upwelling must occur in the interior of the SCS. Consequently, the renewal of the deep water in the SCS is believed to be relatively short, most likely between 30 and 100 years based on hydraulic theory and chemical tracers such as radiocarbon and oxygen (Broecker et al., 1986; Gong et al., 1992; Ou et al., 2006).

To date, the most extensively measured tracers in the SCS are temperature, salinity, oxygen and nutrients (e.g., Gong et al., 1992; Chen et al., 2001; Li and Qu, 2006). However, none of these tracers can be used to determine the age of a water parcel, defined here as the time since last contact with the sea surface. In contrast, transient tracers such as chlorofluorocarbons (CFCs) can be applied to date the age of water (e.g., Fine, 2011). The CFCs have well known input functions (Fig. 1) and are widely used to understand oceanic processes such as ocean circulation (e.g., Weiss et al., 1985; Rhein, 1994; Smethie et al., 2000), the rates and variability of water mass formation (e.g., Orsi et al., 1999; Smethie and Fine, 2001; Rhein et al., 2002; LeBel et al., 2008; Hartin et al., 2011), upper ocean ventilation (e.g., Sonnerup et al., 2008), and estimation of anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) (e.g., Gruber et al., 1996; Sabine et al., 2004; Waugh et al., 2006; Tanhua et al., 2009).

Until recently, most of the transient tracer studies focused on the open ocean and the regions in which strong overturning systems occur, such as the Mediterranean Sea (Schneider et al., 2014), the East (Japan) Sea (Min and Warner, 2005) and the Weddell Sea (Huhn et al., 2013). Comparatively, few studies based on CFCs and/or sulfur hexafluoride (SF<sub>6</sub>) have been carried out in Southeast Asian seas, including the SCS. Fieux et al. (1996) are the only ones who studied Indonesian throughflow based on measured CFC data,



**Fig. 1.** CFC and  $SF_6$  mixing ratios in the northern hemisphere. Data is taken from Bullister (2015).

and Fleischmann et al. (2001) studied the pathway of inflow water from the Western Pacific Ocean to the Banda Sea using CFC-11 data. Observations of CFCs in the SCS is spare, though Sun (2006) studied the CFC distribution and the partial pressure age based roughly on limited data. The CFC-11 measurements presented in this paper thus provide an opportunity to characterize the ventilation and circulation of the SCS.

It is well known that atmospheric CO2 has increased exponentially since the first industrial revolution largely attributable to anthropogenic activities (i.e. burning of fossil fuel, land use changes, etc.). The ocean is the world's largest sink for total CO<sub>2</sub>, absorbing about 30% of the emitted Cant has been absorbed (Sabine et al., 2004). However, the oceanic Cant is only a small fraction of the total carbon reservoir in the ocean (e.g., Khatiwala et al., 2013), which means that Cant cannot be directly separated from the natural background of dissolved inorganic carbon (DIC). While Cant has been estimated in the open ocean such as the North Atlantic Ocean (e.g., Gruber et al., 1996; Gruber 1998; Lee et al., 2003), the North Pacific Ocean (Sabine et al., 2002; Sabine et al., 2004) and the Indian Ocean (e.g., Sabine et al., 1999; Sabine et al., 2004; Hall et al., 2004), comparatively less effort has been made to determine the Cant in marginal seas and coastal areas (e.g., Chen et al., 2006; Park et al., 2006; Chou et al., 2007; Schneider et al., 2010; Wakita et al., 2003). Few data have been obtained in these areas (Lee et al., 2011) recently. To investigate the ventilation time scale and C<sub>ant</sub> in the SCS, we utilized the commonly used transit time distribution (TTD) method, with a CFC data set collected in 2011.

The outline of this paper is as follows: Section 2 outlines the methodology, including analytical procedure of CFCs, TTD and transient steady state; Section 3 describes and discusses the CFC distribution and the derived variables, including mean age, C<sub>ant</sub>, vertical diffusivity and upwelling rate; and the conclusions are presented in Section 4.

#### 2. Methodology

#### 2.1. Sampling and tracer measurements

During 2011, two cruises were undertaken to the SCS: one cruise with the R/V Dongfanghong 2 (Spring973) in April and May and the other with the R/V Xiangyanghong 9 (ROSE3) in October. The stations sampled for CFCs in both cruises are shown in Fig. 2. The samples were collected in glass bottles with a special foil-lined cap, which was similar to that of the USGS (http://water.usgs.gov/lab/chlorofluorocarbons/sampling/bottles/). All the samples were collected with 50 mL glass bottles, sealed and stored at room temperature ( $\sim$ 25 °C), and then taken back for shore-based analysis.

The samples were measured in the laboratory using a set-up similar to the one described by Bullister and Weiss (1988) and detailed in Cai et al. (2013). Briefly, the samples were introduced to a purge and trap system that allowed extraction of the entire CFC content of the sample bottles. After purging the seawater, the CFCs were trapped within a 1/16" stainless steel tube packed with 80–100 mesh Porapak T. After 5 min of trapping, the trap was isolated, and then rapidly heated to 120 °C. The CFCs were analyzed using capillary column electron capture detector (ECD) gas chromatography (GC).

CFC-11 and CFC-12 were measured simultaneously, but we only obtained CFC-11 data owing to a series of procedural and logistical problems when running the GC. The younger tracer SF<sub>6</sub>, though, could not be detected owing to the small volume of the sample (Bullister and Wisegarver, 2008). Because the atmospheric level of CFC-11 has decreased since the mid-1990s, the age of waters formed within the last about 20 years may be hard to constrain

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