

# The flux of methyl chloride along an elevational gradient of a coastal salt marsh, Eastern China

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## Abstract

Salt marshes have been suggested to be a large potential source for methyl chloride ( $\text{CH}_3\text{Cl}$ ) that is the major natural carrier of chlorine to the stratosphere. However, the global budget of this trace gas is uncertain, and the empirical field data are still lacking. In this study,  $\text{CH}_3\text{Cl}$  fluxes were measured seasonally using static flux chambers from April 2004 to January 2005, along an elevational gradient of a coastal salt marsh in eastern China. To estimate the contribution of higher plants to the flux, plant aboveground biomass was experimentally removed and the flux difference between the treatment and the intact was examined. In addition, the flux was analyzed in relation to soil and weather conditions.

Along the elevational gradient, the salt marsh generally functioned as a net sink in the growing season (from April to October 2004). The flux of  $\text{CH}_3\text{Cl}$  ranged between  $-1.27$  and  $-29.33 \mu\text{mol m}^{-2} \text{d}^{-1}$  (positive for emission and negative for consumption), and the maximum negative rates occurred at the mudflat and the cordgrass (*Spartina alterniflora*) marsh. However, the measurements made during inundation indicated that the mudflat was a net source of the gas. In the non-growing season (from November to March), the vegetated flat, when frozen, was a minor source of methyl chloride, with an emission rate ranging from  $0.27$  to  $9.13 \mu\text{mol m}^{-2} \text{d}^{-1}$ . However, the measurements made during non-frozen periods indicate that the mudflat was a minor sink of methyl chloride. Overall, the study marsh was a large net sink for the gas because the magnitude of the consumption rates was larger than that of emission, and because the duration of the growing season was longer than that of the non-growing season. Plant aboveground biomass had a great effect on the flux. Comparative analyses showed that higher plants were present as an important source of  $\text{CH}_3\text{Cl}$ , and it could balance 17.26–67.66% of the soil consumption. The net  $\text{CH}_3\text{Cl}$  consumption rate was negatively correlated to soil dissolved salt content and light intensity, but it was positively correlated to soil temperature, soil organic matter content, and ambient atmospheric concentration of  $\text{CH}_3\text{Cl}$ . This suggests that the overall net consumption of  $\text{CH}_3\text{Cl}$  observed in the study marsh may result from the high ambient atmospheric concentration that ranged between 1.3 and 58.5 ppb, and enriched soil organic matter that feed the soil microorganisms using  $\text{CH}_3\text{Cl}$  as a sole source for both carbon and energy.

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**Keywords:** Cordgrass; Methyl chloride; Plant biomass; Soil uptake; Salt marsh; Seasonal variation

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## 1. Introduction

Methyl chloride ( $\text{CH}_3\text{Cl}$ ) is the most abundant chlorinated organic in the atmosphere and the major natural carrier of chlorine to the stratosphere. Important sources of  $\text{CH}_3\text{Cl}$  identified include biomass burning, oceanic emissions, wood-rotting fungi, and higher plants (Moore et al., 1996; Watling and Harper, 1998; Khalil et al., 1999; Lobert et al., 1999; Harper, 2000; Rhew et al., 2000; Yokouchi et al., 2002). In addition, Keppler et al. (2000) have proposed a novel abiotic route to  $\text{CH}_3\text{Cl}$  by a reaction of organic matter in soil with  $\text{Fe}^{3+}$  and chloride ions under aerobic conditions. However, the global budget for the trace gas is uncertain and further studies of its sources and sinks are needed to reduce the uncertainties (Kurylo and Rodriguez, 1999).

Coastal marshes are potentially a significant source for  $\text{CH}_3\text{Cl}$  because they usually have abundant organic matter and free halides. This has been confirmed by several studies (Cox et al., 2004; Dimmer et al., 2001; Rhew, 2001; Rhew et al., 2000, 2001, 2002). Rhew et al. (2000) even suggested that coastal salt marshes might be a globally significant source of the trace gas. Meanwhile, coastal marshes can be a sink of  $\text{CH}_3\text{Cl}$ .  $\text{CH}_3\text{Cl}$  in salt marsh sediments can be converted to  $\text{CH}_3\text{SH}$  in reactions mediated by anaerobic and aerobic bacteria (Oremland et al., 1994), which subsequently is converted to dimethyl sulfide (DMS) through chemical and bacterial reactions. Because the functions of coastal marshes as sources or sinks of  $\text{CH}_3\text{Cl}$  involve different mechanisms and ecological controls, substantial variations have been observed in previous studies, depending on vegetation and environmental conditions (Rhew et al., 2002; Cox et al., 2004). Consequently, more empirical data are needed to assess the importance of coastal marshes in the global  $\text{CH}_3\text{Cl}$  budget. In addition,  $\text{CH}_3\text{Cl}$  emission from coastal marshes can be attributed to sediment fungi, benthic algae, belowground rhizomes and roots, and aboveground green biomass of higher plants, all of which has been demonstrated to be possible  $\text{CH}_3\text{Cl}$  sources (Moore et al., 1996; Watling and Harper, 1998; Khalil et al., 1999; Lobert et al., 1999; Harper, 2000). Some laboratory and field experiments have suggested that leaves of high plants are a major source for  $\text{CH}_3\text{Cl}$  in coastal marshes (e.g., Rhew et al., 2001). It is believed that higher plants may provide the missing 2 million tons of  $\text{CH}_3\text{Cl}$

necessary to balance the gas budget (Harper, 2000). However, there are few studies estimating the relative contribution of plant aboveground biomass and sediments in the  $\text{CH}_3\text{Cl}$  flux of coastal marshes.

China has a long coast line extending from North China to South China surrounded by the Yellow Sea, the East China Sea, and the South China Sea. The total area of tidal flats is about 2 million hectares (Song, 1983). China also is one of the most densely populated countries in the world. More than 60% of its population currently live in rural areas. They traditionally burn plant die-off biomass as fuels or fertilizers. A previous comparative study has shown that the atmospheric  $\text{CH}_3\text{Cl}$  concentration was much higher in both urban and rural areas of China, compared to the western coastal area of USA (Rasmussen et al., 1982). It is logically expected that China is potentially a very important source in the global  $\text{CH}_3\text{Cl}$  budget. However, to our knowledge, no study has addressed the issue of the  $\text{CH}_3\text{Cl}$  flux so far in China. This study was conducted in Yancheng Natural Reserve of Eastern China, one of the most extensive salt marshes in China. We examined the flux of  $\text{CH}_3\text{Cl}$  along an elevational gradient of the salt marsh over an annual cycle. The objectives of this study are: (1) to characterize the soil condition along the elevational gradient of the marsh; (2) to examine the spatial pattern of  $\text{CH}_3\text{Cl}$  flux along the gradient; (3) to determine the seasonal pattern of the  $\text{CH}_3\text{Cl}$  flux; and (4) to estimate the relative importance of plant aboveground biomass in the  $\text{CH}_3\text{Cl}$  flux.

## 2. Materials and methods

### 2.1. Site description

The study salt marsh is located at the core zone of Yancheng Natural Reserve, Jiangsu Province of China ( $\text{E}33^\circ36'$ ,  $\text{N}120^\circ36'$ ) (Fig. 1). It belongs to the East Asia Monsoon Climate Zone between subtropical and warm temperature zones. The salt marsh is inundated by two tides every day. The marsh is currently accreting with the mean high water level advancing seaward at a rate of about 200 m per year and the sediment accreting vertically at a rate of  $2.5\text{--}10\text{ m year}^{-1}$  on average. The mean annual temperature is  $18.7^\circ\text{C}$ , with the highest being  $26.2\text{--}26.6^\circ\text{C}$  in the warmest month of July or August, and the lowest being  $0.5^\circ\text{C}$  in the coldest month, January. The mean annual precipitation

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