



Temporal variations in the distribution and sea-to-air flux of marine isoprene in the East China Sea

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ABSTRACT

Marine isoprene concentrations in the East China Sea were measured during two oceanographic cruises: one from October 10, 2013 to November 21, 2013 and another from May 16, 2014 to June 13, 2014. Mean (range) isoprene concentrations in the surface water in the study area were 32.85 (16.31–108.2) pmol L⁻¹ in late spring and 26.83 (11.22–42.13) pmol L⁻¹ in autumn. The isoprene concentrations showed seasonal variation and were higher in late spring, which could be attributable to terrigenous diluted water and seasonal variations of phytoplankton structure. Isoprene and chlorophyll *a* (Chl-*a*) concentrations were significantly correlated in late spring and autumn even under complex hydrographic conditions. Isoprene and Chl-*a* concentrations also exhibited similar variations during a 25 h period and remained high during daytime. Analysis of depth profiles indicated that isoprene concentrations in the water column peaked at the surface layer and gradually decreased with increasing depth. The atmospheric concentration of isoprene (average: 53.4 ppt; range: 14.8–141 ppt) was high in the onshore area but low in the offshore area in late spring. The mean sea-to-air fluxes of isoprene in the East China Sea were 36.12 (late spring) and 48.34 (autumn) nmol m⁻² d⁻¹. Results revealed the substantial isoprene emission from surface seawater, which was also a net source of atmospheric isoprene during the study period.

1. Introduction

The oceans are a source of non-methane hydrocarbons (NMHCs) in the atmosphere (Baker et al., 2000). Once transported across the sea-to-air interface, NMHCs participate in various atmospheric chemical processes (Pszenny et al., 1999; Liakakou et al., 2009; Sahu et al., 2011). Thus, marine-derived NMHCs are assumed to play significant roles in atmospheric chemistry and climate change (Matsunaga et al., 2002; Arnold et al., 2009). Isoprene (2-methyl-1, 3-butadiene, C₅H₈) is not only the important component of NMHCs but also the most commonly emitted biogenic volatile organic compound (BVOC) in the atmosphere. As far as the global isoprene flux, it might comprise approximately 44% of total VOC flux (Guenther et al., 1995). As much as 90% of atmospheric isoprene is derived from terrestrial plant emissions globally (400–600 Tg C a⁻¹; Guenther et al., 2006; Arneth et al., 2008). Terrestrial vegetation comprises the largest isoprene emissions (Liakakou et al., 2007; Sharkey et al., 2008); however, productive areas of remote oceanic regions, coastal upwelling regions, and wetlands have been

shown to emit isoprene (Bonsang et al., 1992; Broadgate et al., 1997; Ekberg et al., 2009). Owing to the short lifetime of atmospheric isoprene, which ranges from several minutes to several hours, marine-derived isoprene can also potentially influence the oxidation capacity of the atmosphere over remote oceanic and coastal regions (Palmer and Shaw, 2005; Kameyama et al., 2014). Isoprene rapidly reacts with nitrate (NO₃), hydroxyl radicals (OH), and ozone (O₃) (Atkinson and Arey, 2003; Lelieveld et al., 2008). Isoprene oxidation plays an important role in modulating tropospheric O₃ and methane (CH₄) concentrations (Sahu and Lal, 2006; Pacifico et al., 2009; Zindler et al., 2014). In addition, isoprene has been shown to be a significant precursor to the formation of atmospheric secondary organic aerosol (SOA) (Claeys et al., 2004; Kroll and Seinfeld, 2008), which can strongly affect the radiation balance of the atmosphere, modify cloud microphysics, and participate in chemical transformations (Pacifico et al., 2009).

Isoprene dissolved in seawater is mainly produced by phytoplankton (Shaw et al., 2003; Tran et al., 2013; Booge et al., 2016), seaweed (Broadgate et al., 2004), and bacteria (Kuzma et al., 1995;

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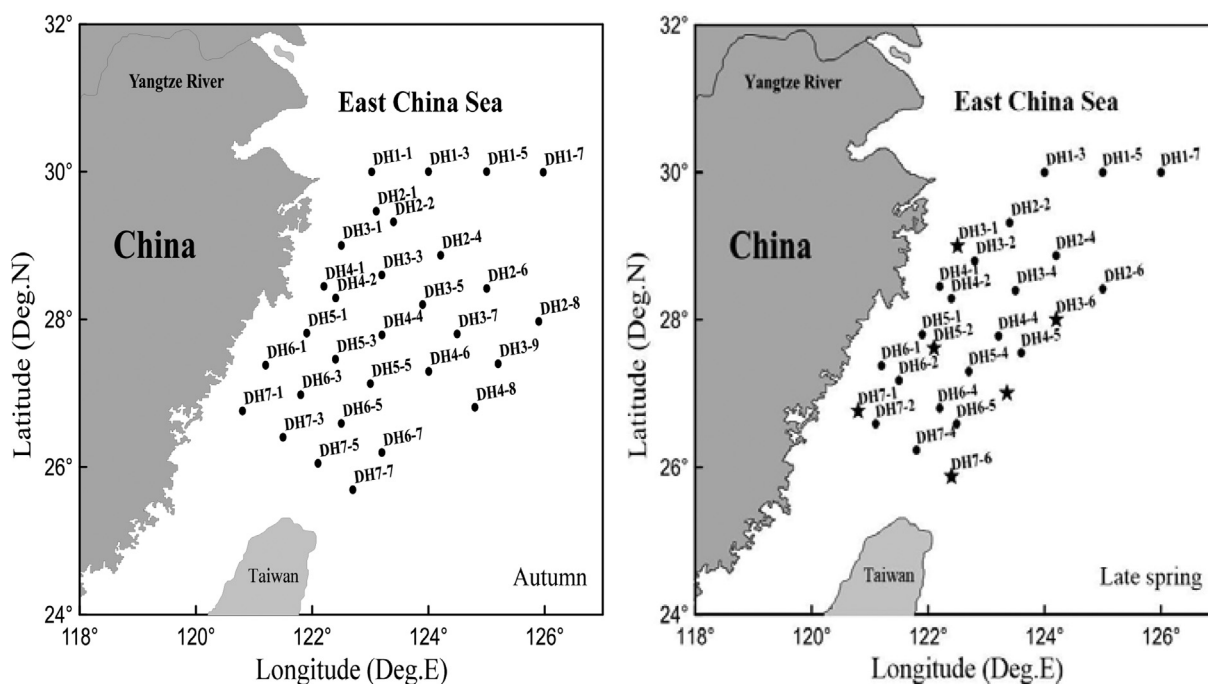


Fig. 1. Locations of sampling stations in the ECS (represent the atmospheric sampling stations in late spring 2014).

Alvarez et al., 2009). Therefore, the oceans play an important role in the biogeochemical cycle of isoprene and the distribution of isoprene in seawater influenced by various environmental factors. Previous studies indicated that isoprene concentration is positively related to the concentration of chlorophyll-*a*, an indicator of phytoplankton abundance (Bonsang et al., 1992; Broadgate et al., 1997; Shaw et al., 2003). However, phytoplankton biomass alone cannot account for the distribution of isoprene in surface seawater because the relationship is not always linear (Kameyama et al., 2014). Previous studies estimated the production rates of isoprene for each phytoplankton group by performing experiments with cultured isolates (Bonsang et al., 2010; Shaw et al., 2010; Booge et al., 2016). The results indicated that different phytoplankton functional types (PFTs) have different production rates of isoprene (Yassaa et al., 2008). Various PFTs also exhibit different producing isoprene capacities, and the isoprene emission rates seem to be mainly influenced by environmental conditions, such as light, ocean temperature, and salinity (Exton et al., 2013; Bonsang et al., 2010; Booge et al., 2018). In addition, the experimental results from Ciuraru et al. (2015) suggested that the photochemical degradation of dissolved organic matter in the sea-surface microlayer can be an abiotic source of isoprene.

Emission to the atmosphere through sea-to-air exchange has been inferred as the primary sink of oceanic isoprene (Matsunaga et al., 2002; Booge et al., 2018). Kim et al. (2017) showed that the sea-to-air flux of isoprene determined using the eddy covariance method was estimated to be 0.57 Tg C a^{-1} from the surface ocean. In addition, the combination of *in-situ* observation and remote sensing data indicates that the isoprene emission of from the ocean to the atmosphere is $0.1\text{--}1.9 \text{ Tg C a}^{-1}$ (Palmer and Shaw, 2005; Arnold et al., 2009; Gantt et al., 2009). However, limited information on the chemical and microbial losses of marine isoprene is reported. Shaw et al. (2003) assumed the biological loss by bacterial degradation to be considerably small. Oceanic emission acts as the main loss of isoprene in seawater to possess significantly spatial and temporal impacts, particularly in the productive and coastal upwelling regions (Zindler et al., 2014; Li et al., 2017). Coastal regions are high in nutrients and organic matter from terrestrial sources to provide suitable growth conditions for marine organisms. As such, coastal regions influenced by human activities have drawn increased research interest in isoprene emission and distribution

of Milne et al. (1995), Broadgate et al. (2004), Ooki et al. (2015).

About 70% of the East China Sea (ECS), one of the largest shelf regions worldwide, is located on the continental shelf (Wong et al., 2000). The ECS exhibits complex hydrographic characteristics, including not only terrigenous diluted waters (e.g., the Yangtze River and the Qiantang River) but also various water masses such as the Kuroshio Current Water, Taiwan Warm Current Water, the Continental Coastal Water and ECS Surface Water (Su, 1998; Lee et al., 2000; Zhang et al., 2008; Qi et al., 2014). Such a unique oceanographic environment renders the ECS a significant area for studying the role of the marginal seas of the Northwest Pacific Ocean in global biogeochemistry. Despite several reported on isoprene emissions in different oceanic regions (e.g., Kurihara et al., 2012; Kameyama et al., 2014; Li et al., 2017; Booge et al., 2018; Zhai et al., 2018), large uncertainties in estimating the global marine isoprene emissions arise because of the small size of measurement dataset on spatial-temporal resolution. Therefore, the need for *in-situ* measurements of isoprene emissions from biologically active regions, such as coastal and upwelling regions, is to elucidate the marine isoprene cycle and improve estimates of global marine isoprene emissions. In the present study, seawater concentrations and spatial-temporal distributions of isoprene in the ECS were measured in the autumn of 2013 and late spring of 2014. The possible sources of isoprene were also discussed based on related environmental and hydrographic parameters. The sea-to-air fluxes of isoprene were also estimated during the two study periods.

2. Methods

2.1. Sampling collection

Seawater samples were collected during two cruises in the ECS (1) aboard the R/V “*Science 3*” from October 10, 2013 to November 21, 2013 and aboard the R/V “*Science 1*” from May 16, 2014 to June 13, 2014. The aim was to study the seasonal and horizontal distributions of isoprene in autumn and late spring. The sampling stations are shown in Fig. 1. The vertical distributions of isoprene were investigated along the DH1 transect in autumn. Variations in isoprene were also evaluated during a 25 h period. In addition, a continuous investigation was conducted at the station of DH1-1 (123.02°E, 30.00°N) in autumn,

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