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Variations in stable hydrogen and oxygen isotopes in atmospheric water vapor in the marine boundary layer across a wide latitude range

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ABSTRACT

The newly-developed cavity ring-down laser absorption spectroscopy analyzer with special calibration protocols has enabled the direct measurement of atmospheric vapor isotopes at high spatial and temporal resolution. This paper presents real-time hydrogen and oxygen stable isotope data for atmospheric water vapor above the sea surface, over a wide range of latitudes spanning from 38°N to 69°S. Our results showed relatively higher values of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in the subtropical regions than those in the tropical and high latitude regions, and also a notable decreasing trend in the Antarctic coastal region. By combining the hydrogen and oxygen isotope data with meteoric water line and backward trajectory model analysis, we explored the kinetic fractionation caused by subsiding air masses and related saturated vapor pressure in the subtropics, and the evaporation-driven kinetic fractionation in the Antarctic region. Simultaneous observations of meteorological and marine variables were used to interpret the isotopic composition characteristics and influential factors, indicating that *d*-excess is negatively correlated with humidity across a wide range of latitudes and weather conditions worldwide. Coincident with previous studies, *d*-excess is also positively correlated with sea surface temperature and air temperature (T_{air}), with greater sensitivity to T_{air} . Thus, atmospheric vapor isotopes measured with high accuracy and good spatial-temporal resolution could act as informative tracers for exploring the water cycle at different regional scales. Such monitoring efforts should be undertaken over a longer time period and in different regions of the world.

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Introduction

Atmospheric water vapor plays a crucial role in the global water cycle and in moisture transportation. Knowledge of this role has enhanced our understanding of global water and energy circulation and has

revealed the influence of the water cycle on global climate change. Although near-surface water vapor comprises a very small percentage of the total mass of the hydrosphere (0.001%) and has very low vapor concentration (0.4×10^2 – 400×10^2 ppm), it is worth noting that its average retention time is only 10 days (Gat et al., 2001) and

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that it experiences frequent and intensive exchange with other water phases. These processes generate isotopic contrasts between different water exchanges among oceans and continents (e.g. ocean water, vapor, ice and groundwater).

Both the hydrogen ($^2\text{H}/\text{H}$) and oxygen ($^{18}\text{O}/^{16}\text{O}$) stable isotope ratios in atmospheric water vapor are valuable tracers because they can reflect water exchange processes and help us understand the different phases of the global moisture cycle (Noone, 2008; Worden et al., 2007; Yamanaka and Shimizu, 2007). Our understanding of the mechanisms that control natural stable isotopic fractionation has improved greatly over recent years (Craig, 1961; Dansgaard, 1964; Jouzel et al., 1997; Merlivat and Jouzel, 1979; Tian et al., 2007). Stable isotope values are reported using the δ notation, $\delta^{18}\text{O}$ for H_2^{18}O and $\delta^2\text{H}$ for HD^{16}O , and using the derived parameter deuterium excess (d -excess = $\delta^2\text{H} - 8\delta^{18}\text{O}$). The δ values are predominantly determined by the conditions at the water vapor source, the degree of moisture recycling during vapor transport, and the temperature of final condensation; meanwhile, d is mainly controlled by the temperature and relative humidity of the air mass at the moisture source. However, there remain many unknowns regarding moisture cycling and related stable isotope fractionation, in particular due to the scarce observations of atmospheric vapor isotopes. To address these unknowns, recent studies have concentrated on observations of atmospheric water vapor isotopes in different places (Gat, 1996; Kurita, 2011; Noone et al., 2011; Steen-Larsen et al., 2011; Uemura et al., 2008).

Traditional methods of atmospheric vapor isotope measurement, such as collecting vapor samples by the cryogenic method, neither allow measurements in real time nor provide a wide spatial coverage. Fortunately, with recent developments in cavity-enhanced absorption spectroscopy, it is now possible to make continuous measurements of atmospheric water vapor isotopic composition in real time (Kerstel et al., 1999; Lee et al., 2005; Steen-Larsen et al., 2011).

Following instrumental development during the 28th Chinese Antarctic Expedition, a newly developed cavity-enhanced near

infra-red absorption spectroscopy analyzer (PICARRO L-1102i) was installed on the expedition ship to continuously monitor stable isotopes of hydrogen and oxygen in atmospheric vapor in real time. PICARRO L-1102i is a cavity ring-down, near-infrared laser absorption spectroscopy analyzer. The cavity ring-down laser absorption spectroscopy instrument is based on the measurement of the exponential decay time of light resonating in an optical cavity to quantify the molecular absorption characteristics at different optical wavelengths, which makes small dose measurement possible in field applications (Brand et al., 2009; Crosson et al., 2002). Observations were made from 3rd to 29th November 2011, along a route from the Chinese coast southwards to the Antarctic coastal region (38°N to 69°S) (Fig. 1, left panel). Meteorological and oceanic data were also collected along the same route. The spatial and temporal distributions of $\delta^{18}\text{O}$, $\delta^2\text{H}$ and derived d -excess above the ocean were determined to investigate the characteristics of relationships between vapor isotope fractionation and meteorological conditions.

1. Instruments and data calibration

1.1. Instruments and methodology

Owing to progress in theoretical isotopic hydrology since the 1960s, different instruments have been applied for stable water isotope measurement (Dansgaard, 1953; Craig and Gordon, 1965; Merlivat and Coantic, 1975; Gat, 1996; Noone, 2008; Uemura et al., 2008; Steen-Larsen et al., 2013). The mass spectrometric analyzer, which is the earliest and the most reliable method for liquid isotope measurement, has been commonly used in natural physiochemical and environmental water isotope studies. However, mass spectrometers are

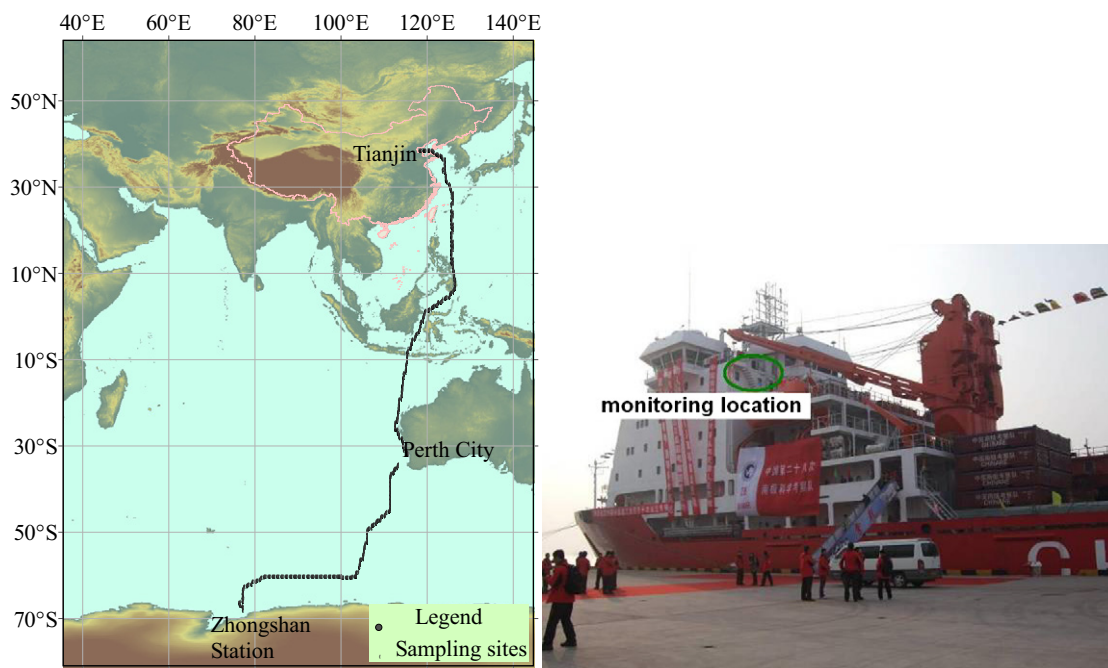


Fig. 1 – Cruise track and data collection sites (left) and location of the water vapor isotope sampling inlet and measurement instrument on the ship (right).

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