

Regional prediction of carbon isotopes in soil carbonates for Asian dust source tracer



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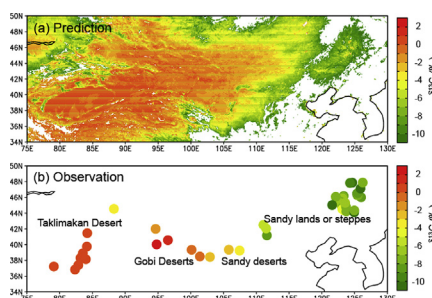
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HIGHLIGHTS

- The $\delta^{13}\text{C}$ of soil carbonates were investigated in northern China.
- $\delta^{13}\text{C}$ is correlated with Normalized Difference Vegetation Index.
- $\delta^{13}\text{C}$ in deserts and semi-arid lands were predicted with satellite NDVI.
- Asian dust source areas were distinguished by the ^{13}C mapping.
- The isotope tracer monitors changes of anthropogenic dust budget.

GRAPHICAL ABSTRACT



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ABSTRACT

Dust particles emitted from deserts and semi-arid lands in northern China cause particulate pollution that increases the burden of disease particularly for urban population in East Asia. The stable carbon isotopes ($\delta^{13}\text{C}$) of carbonates in soils and dust aerosols in northern China were investigated. We found that the $\delta^{13}\text{C}$ of carbonates in surface soils in northern China showed clearly the negative correlation ($R^2 = 0.73$) with Normalized Difference Vegetation Index (NDVI). Using Moderate Resolution Imaging Spectroradiometer (MODIS) satellite-derived NDVI, we predicted the regional distribution of $\delta^{13}\text{C}$ of soil carbonates in deserts, sandy lands, and steppe areas. The predictions show the mean $\delta^{13}\text{C}$ of $-0.4 \pm 0.7\text{‰}$ in soil carbonates in Taklimakan Desert and Gobi Deserts, and the isotope values decrease to $-3.3 \pm 1.1\text{‰}$ in sandy lands. The increase in vegetation coverage depletes ^{13}C in soil carbonates, thus the steppe areas are predicted by the lowest $\delta^{13}\text{C}$ levels ($-8.1 \pm 1.7\text{‰}$). The measurements of atmospheric dust samples at eight sites showed that the Asian dust sources were well assigned by the ^{13}C mapping in surface soils. Predicting ^{13}C in large geographical areas with fine resolution offers a cost-effective tracer to monitor dust emissions from sandy lands and steppe areas which show an increasing role in Asian dust loading driven by climate change and human activities.

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1. Introduction

Atmospheric dust from Taklimakan Desert, Gobi Deserts, sandy lands, and associated drylands in northern China can be transported to cities in East Asia (Lee and Sohn, 2011; Li et al., 2009),

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while across the Pacific dust aerosols even reach North America (Uno et al., 2011; Wang et al., 2011; Zhao et al., 2015). Dust aerosols mixing with urban air pollutants are considered to be particular toxic that may increase the burden of pulmonary tuberculosis (Wang et al., 2016), cerebrovascular disease, even increase mortality in cities in East Asia (Kashima et al., 2016; Pan et al., 2015; Wang and Lin, 2015). The long-range transport of Asian dust aerosols modify cloud formation and alter precipitation efficiency in regional and global scales (Ault et al., 2011; Creamean et al., 2013; Huang et al., 2010).

It is proposed that the stable carbon isotope ($\delta^{13}\text{C}$) of carbonates may serve as an effective tracer to distinguish Asian dust sources (Cao et al., 2005; Chen et al., 2015). Early studies had investigated $\delta^{13}\text{C}$ in soil carbonates in some sampling sites in northern China to establish the isotope tracer (Cao et al., 2008; Wang et al., 2005). However, the sampling campaigns in field and laboratory measurements of carbon isotopes are labor intensive and costly, and such traditional field investigation has limitation in spatial coverage. It should be useful to explore a method to predict the high-resolution map of $\delta^{13}\text{C}$ in soil carbonates in regional scales.

It is known that $\delta^{13}\text{C}$ of pedogenic carbonate is controlled by the proportion of C3 to C4 plants and vegetation coverage (Cerling et al., 1989; Johnston et al., 2012; Quade et al., 1989). Normalized Difference Vegetation Index (NDVI) is satellite-derived vegetation indices to measure vegetation greenness, based on reflectance in the red (RED) and near-infrared (NIR) regions of the electromagnetic spectrum. NDVI is correlated with several variables that including net primary productivity, plant biomass, and leaf area index (Pettorelli et al., 2011). The remote sensed NDVI has global coverage, thus the NDVI referring of vegetation may be a potentially powerful tool to predicting the $\delta^{13}\text{C}$ of soil carbonates at high resolution and large spatial extents.

Here, surface soil samples in deserts, sandy lands, and steppe areas in northern China were investigated for carbonate contents and isotope ratios. The correlation between $\delta^{13}\text{C}$ and NDVI for surface soil was analyzed. Using remote sensed NDVI, the $\delta^{13}\text{C}$ of soil carbonates were predicted with fine resolution in northern China. We discussed on the prediction of $\delta^{13}\text{C}$ as an isotope tracer to distinguish dust sources between deserts (natural sources), and semi-arid lands (anthropogenic sources). The NDVI predicting of ^{13}C map offers several advantages over traditional investigation method, including fine spatial and regional coverage, typically low (or no) data acquisition cost, and perhaps most significantly, no field sampling and high-cost-instrumental measurements.

2. Materials and methods

2.1. Samples

Intense sampling campaigns were made for the collection of surface soils ($n = 65$) in bare lands in deserts, sandy lands, and steppe areas in northern China (Fig. 1). During Asian dust events, atmospheric dust particles were sampled at eight sites along the deserts (Aksu, Dunhuang), sandy lands (Yulin, Xi'an, Yangling, and Beijing), and steppe areas (Rural-Steppe, and Changchun). The dust events were collected by 34 samples of total suspended particles (TSP), 10 samples of deposition dust (D), and one fine particulate matter ($\text{PM}_{2.5}$). The TSP samples, that having total size range, covered 76% of dataset for dust samples.

To avoid the impact of human activity on the samples, the sampling sites of Asku, Dunhuang, and Yulin were in deserts or desert margin areas, and the Rural-Steppe was a remote uninhabited site. The dust samples at urban areas (Beijing and Changchun) were collected at 25–39 m high above ground level (AGL). Our early study suggested that local human activities such as urban road dust

and construction should cause only minor impact on the dust storm samples for analyzing of isotopes in carbonate particles, because dust storm events significantly increased the carbonate concentrations (Chen et al., 2015). Agriculture activity in the surrounding of Beijing and Changchun may contribute to dust particles for the samples. However, the carbonate contents of soils in the surrounding of Beijing and Changchun were 0% ($n = 3$) and 0.1% ($n = 1$), respectively, significant lower than the carbonate contents of dust samples at Beijing (5%) and Changchun (3.7%). Thus the agriculture dust from the surrounding of the urban areas had minimum contributions for the carbonate aerosols. Taken together, the human activities from local urban areas and the surrounding had minor impact on carbonate and isotope signatures for dust tracer.

2.2. Measurements of carbonate contents and isotope ratios

The carbonate contents and isotope ratios were determined for the samples. The samples were allowed to mix with purified phosphoric acid in vacuum reactors, and carbonates were reacted to release CO_2 which was cryogenically purified using vacuum lines. The carbonate contents in sample were thus calculated from the CO_2 determination. The uncertainties were less than 0.01 mg of carbonates, approximately yielding a relative error of 5% for the carbonate content determination of soil and dust samples. The CO_2 production from carbonate-free soil samples was lower than the detection limit, thus the samples were not pre-treated to remove organic matters.

Finnigan MAT isotope ratio mass spectrometer (MAT 251 or MAT 252) was employed to measure the $^{13}\text{C}/^{12}\text{C}$ ratios in the purified CO_2 . The reference material NBS-19 (limestone) was calibration standard for the mass spectrometer. In addition, a laboratory standard (calcium carbonate) was provided to allow routine checking of the overall quality of measurements performed by isotope ratio mass spectrometer, and used as part of a quality control program. This laboratory standard provided a sample of known isotope composition with $^{13}\text{C}/^{12}\text{C}$ and $^{18}\text{O}/^{16}\text{O}$ isotope ratios from Vienna Pee Dee Belemnite (V-PDB) standard. The $^{13}\text{C}/^{12}\text{C}$ and $^{18}\text{O}/^{16}\text{O}$ isotope ratios of the laboratory standard were determined using NBS-19 as the calibration material. Moreover, the Cellulose standard was applied for the confirmation of ^{13}C -depleted samples. The $\delta^{13}\text{C}$ values were reported as per mil deviation relative to the V-PDB standard. The triplicate measurements of samples showed a relative error of $\sim 0.15\%$ for $\delta^{13}\text{C}$.

2.3. The Normalized Difference Vegetation Index

The Normalized Difference Vegetation Index (NDVI) is based on measurements taken by the Moderate Resolution Imaging Spectroradiometer (MODIS) on NASA's Terra satellite (Fig. 1a). It is well established that NDVI is highly correlated with fractional vegetation cover (Carlson and Ripley, 1997). A zero of NDVI means no vegetation and close to +1 indicates the highest possible density of green leaves; forests typically have the NDVI of 0.8–0.9. The MODIS NDVI was suggested to be applied in desert and semi-arid environments (Crech et al., 2016; Sesnie et al., 2012). We retrieved 5 year mean data (2000–2005), and the comparison of 5 year mean with 10 year data (2000–2010) showed almost identical NDVI ($R^2 = 0.999$). The spatial horizontal resolution of NDVI used in this study is 5.6 km. In general, the NDVI was lower than 0.4 in Asian dust source areas, suggesting a low abundance of vegetation coverage.

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