



Tracing the sources of sulfur in Beijing soils with stable sulfur isotopes



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ABSTRACT

Stable sulfur isotope analyses were applied in order to trace local sulfur sources and to identify sulfur turnover rates in Beijing soil. Anthropogenic sources and atmospheric deposition are the main processes responsible for the occurrence of sulfide, organic and sulfate sulfur in three soil profiles located in the vicinity of a steel company in Beijing, China. This input results in an atypical invariable sulfur isotopic composition along the vertical profiles. The $\delta^{34}\text{S}$ values of soil sulfate from the industrial area revealed that both the topsoil and deep soil have been affected by industrial activities. Three soil profiles from non-industrial areas displayed vertical patterns in both sulfur abundance and sulfur isotopic composition, indicative for (close to) natural soils.

Eight soil profiles collected in Beijing and their vertical distribution of different sulfur species, concentrations and sulfur isotopic compositions reveal both anthropogenic and natural contributions.

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1. Introduction

The fast economic development of Beijing in the past few decades, and the subsequent industrial, communal and agricultural contributions have affected the environment in this region. Soil is a prime recipient for industrial, communal and agricultural input. The release of sulfur dioxide and particles by coal combustion especially in industrial areas, constitutes a serious perturbation to the pre-existent environmental geochemical sulfur cycle, and contributes to atmospheric pollution and acid rain deposition. Usually, there are six main inputs of sulfur (S) into soil: (i) atmospheric deposition (Legge and Krouse, 1992), (ii) weathering of parent material (Knöller et al., 2004; Migazewski et al., 2013, 2015), (iii) application of S-containing fertilizers, manure and sewage (Otero and Soler, 2002; Szykiewicz et al., 2011), (iv) plant and animal residues (Legge and Krouse, 1992), (v) CS (carbon, sulfur)-mineralization in humic-mixed soils and debris covers under aerated conditions (e.g. Novák et al., 2001, 2004; Knöller et al., 2005; Shanley et al., 2005), and (vi) anaerobic bacterial (dissimilatory) reduction (BSR) of sulfates derived from different sources (e.g. Mayer et al., 1995, 2004; Galuszka, 2005). Despite an established understanding of these inputs, the development/application of effective methods for tracing the sources of sulfur in soil are still posing a challenge.

The application of stable isotope analyses on S compounds in different environments for tracing the source of sulfur is a growing field (Legge and Krouse, 1992; Thompson and Bottrell, 1998; Chu, 2000; Xiao and Liu, 2002; Novák et al., 2001, 2004; Knöller et al., 2004; Liu

et al., 2008; Migazewski et al., 2013, 2015). The $\delta^{34}\text{S}$ variations among different S compounds can be attributed to isotopic selectivity in biochemical transformations or to different S sources.

From a number of previous studies, it was concluded that besides the concentration and duration of exposure to the source (Krouse et al., 1984, 1991; Legge and Krouse, 1992), the amount of plant cover (Krouse, 1989; Winner et al., 1981), soil type and texture (Krouse and Tabatabai, 1986; Krouse et al., 1991), S amounts in the soil (Mayer, 1993), and net drainage (Krouse (1989)) are the major factors which determine the depth of penetration of anthropogenic S into the soil (Legge and Krouse, 1992).

There are different sulfur species existing in soil samples, including (1) soluble S which is represented by monosulfides and soluble sulfate; (2) chromium reducible S (CRS), consisting primarily of pyrite sulfur; and (3) organically bound sulfur (OBS). A large number of detailed studies have shown that SO_4^{2-} strongly interacts with soils, and participates in a complex organic–inorganic soil S cycle (Van Stempvoort et al., 1992; Zhang, 2009). Through analyzing the sulfur isotope composition of SO_4^{2-} in soil, the sources of sulfur can be traced.

A few articles (Legge and Krouse, 1992; Thompson and Bottrell, 1998) have focused on the usefulness of different sulfur isotope techniques applied on distinct sulfur species from the four compartments of the global environment: atmosphere, biosphere, hydrosphere, and lithosphere.

In this study, the stable isotope investigations of different sulfur compounds in soil and water will be used for tracing the sources of sulfur in Beijing soils and compare soil profiles from non-industrial and industrial areas and water samples from Beijing in order to identify natural and anthropogenic processes, which influence the soil sulfur cycle.

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2. Study area, sampling, and analytical methods

2.1. Study area

Beijing (39°20′–40°90′N, 115°20′–117°20′E) is located in North China (Fig. 1). The mountains in the west and north of Beijing consist of Lower Paleozoic rocks (Xie, 1990). The city was built on the northern tip of the North China Plain, at an elevation between 40 and 60 m above sea level. The climate is humid continental, driven by the monsoon, and characterized by hot and humid summers as well as generally cold, dry and windy winters. The average temperatures are 1.6 °C in January and 30.8 °C in July.

2.2. Sampling

In total, 112 soil samples were collected from eight soil profiles at eight different sites in Beijing (Fig. 1; Table S1) (Guo et al., 2013). Soil sampling was performed using a hand-held soil sampler.

Soil profiles A, B and C (DLC) are located in the Beijing Steel Company (Guo et al., 2013).

The FCK profile (D) and the 469 profile (E) are located outside of the main area of the Beijing Steel Company (Guo et al., 2013).

The Tongzhou profile (F), the Yongledian profile (G), and the Guadi profile (H) are situated in the Beijing countryside (Guo et al., 2013).

Four coal samples were collected from the Beijing Steel Company (Table S2).

Eleven lake water samples were separately collected in January and May 2010 from six lakes (Jishuitan lake (JST), Zizhuyuan lake (ZZY), Yiheyuan lake (YHY), Chuandongyuan lake (CDY), Weiminghu lake (WMH), Yuanmingyuan lake (YMY)) in the Beijing urban area (Table S2).

Six rain water samples were separately collected in August and October 2010 from the top of the main building of the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing (Table S2).

2.3. Analytical methods

The soil samples were analyzed for their C and S abundance and the isotopic composition of different S-bearing phases.

Soil and coal samples were dried at room temperature and grinded. The content of soil organic carbon (SOC) ($\text{SOC} = \text{TC} - \text{TIC}$) was determined as the difference between total carbon (TC) and total inorganic carbon (TIC). These were measured using IR spectroscopy following

the liberation of carbon dioxide from soil samples via combustion at 1350 °C (TC) or acid-digestion (TIC). Analyses were performed using a CS Mat 5500 at the Institut für Geologie und Paläontologie, University of Münster, Germany, standard deviation for the sulfur abundance was ± 0.03 wt.%.

Concentration measurements of water samples on anions were analyzed by gas chromatography using an AMS Smartchem 300® in the laboratories of the Institute of Geographic Sciences and Natural Resources Research, Beijing, China. Analytical precision was better than $\pm 5\%$.

Lake and rain water samples were first filtered using 0.45 μm cellulose nitrate filters, and sulfate was subsequently extracted from the solution via precipitation as barium sulfate (BaSO_4) after adding BaCl_2 using standard procedures (e.g. Dogramaci et al., 2001).

For sulfur isotope analyses of the soil samples, three sulfur species were extracted from the soil by sequential extraction: (1) soluble S represented by monosulfides and soluble sulfate, (2) chromium reducible S (CRS) consisting primarily of pyrite sulfur, and (3) organically bound sulfur (OBS). First, the CRS was liberated through a HCl solution and a hot 1.0 mol/L CrCl_2 solution. Resulting H_2S from both steps was precipitated as ZnS and thereafter transformed to Ag_2S with 0.1 mol/L AgNO_3 solution (Canfield et al., 1986). In the second stage, the sample residue was filtered, soluble sulfate was extracted from the solution and precipitated as barium sulfate (BaSO_4) after adding BaCl_2 using standard procedures. Finally, the solid residue was thoroughly mixed with Eschka powder (ASTM, 1977) and combusted at 800 °C in order to oxidize any residual sulfur. This final fraction represents organic bound sulfur (OBS), which was precipitated as BaSO_4 , using a BaCl_2 solution. Sulfate was measured as a trace constituent in soils.

The sulfur isotopic composition from BaSO_4 and Ag_2S was precipitates measured with a Finnigan Delta V Plus mass spectrometer equipped with an elemental analyzer at the Institut für Geologie und Paläontologie, University of Münster, Germany. Results are reported in the standard delta notation against the Canyon Diablo troilite standard (VCDT). Reproducibility was better than $\pm 0.3\%$.

3. Results

Analytical results are presented in Tables S1 and S2 and plotted in Figs. 2–5. The total sulfur content of the collected soil samples was less than 1 wt.-%. The $\delta^{34}\text{S}$ values of organic sulfur are rather homogeneous: $\delta^{34}\text{S}_{\text{OBS}}$ values vary from 2.8‰ to 6.5‰ ($5.1 \pm 1.5\%$, $n = 5$, VCDT).

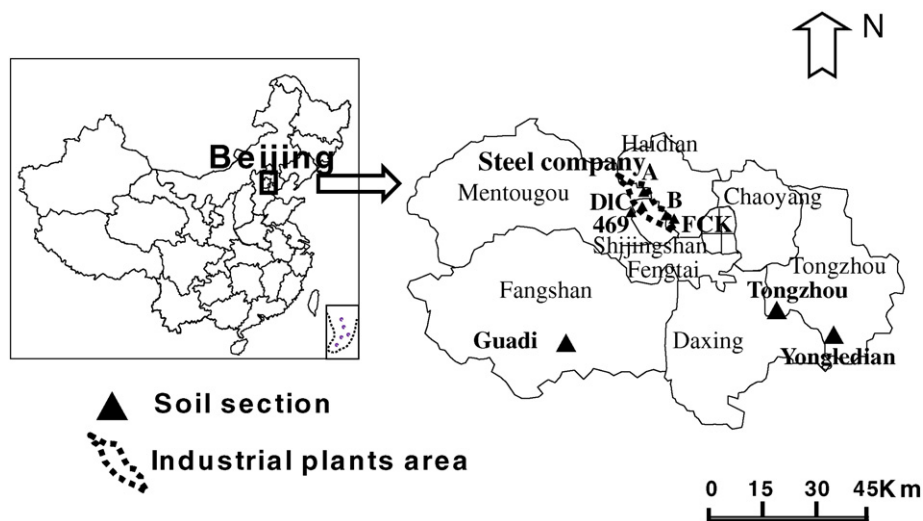


Fig. 1. Location of soil profiles (1) A, sintering plant section, (2) B, rolling plant section, (3) DLC section, (4) FCK section, (5) 469 section, (6) Tongzhou section, (7) Yongledian section, (8) Guadi section, Beijing city, China.

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