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NO_3^- sources and circulation in the shallow vadose zone in the edge of Dunhuang Mingsha sand dunes in an extremely arid area of Northwestern China

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ABSTRACT

The unsaturated zone is a significant feature of the nitrogen cycle in arid areas connecting atmospheric precipitation and surface water to groundwater. To measure NO_3^-, Cl^-, and NH_4^+ concentrations, precipitation samples (39.9592°N, 94.3302°E) were collected at the western edge of the Mingsha sand dunes. Soil profiles were taken along the northern edge of the dunes to determine NO_3^- and Cl^- concentrations and $\delta^{18}O\text{-}NO_3^-$ and $\delta^{15}N\text{-}NO_3^-$ levels in soil water. The mNO_3^-/Cl^-, mNH_4^+/Cl^- and $NH_4^+\text{-}N/NO_3^-\text{-}N$ values in precipitation showed seasonal variations, reflecting the influence of distinct anthropogenic sources. The $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ of the LT profile ranged from 2.38 to 7.84‰ and 14.72 to 57.5‰, respectively, and that of GC profile ranged from −1.36 to 2.35‰ and 5.07 to 21.3‰, respectively. These results suggest that the main sources of vadose zone nitrogen in the profiles are NO_3^- fertilizer and NH_4^+ in fertilizer and precipitation, all of which are influenced by agricultural sources. Obvious nitrification and the mixing of different sources were noted, but the profiles showed no evidence of denitrification. The findings of this study show that migration of NO_3^- and Cl^- in the desert vadose zone is influenced by heavy rainfall events, vegetation, and evaporation. Heavy rainfall events promote leaching of NO_3^- and Cl^- into the deep vadose zone, while evaporation at the surface prevents leaching and vegetation hinders the downward migration of NO_3^- and Cl^-. Migration of NO_3^--N and Cl^- in sand dunes is affected by the solute gradient concentration difference of the soil water, moving from areas of higher concentrations to lower concentration. The NO_3^--N and Cl^- accumulations were 1.02 kg N ha^{-1} and 3.45 kg ha^{-1}, respectively, throughout autumn. These were larger than could be attributed to the input flux of atmospheric precipitation, especially in the near-surface soil layer, likely illustrating the influence of atmospheric dry deposition.

1. Introduction

Changes to the nitrogen cycle have occurred at regional and global scales (Galloway et al., 2008; Li et al., 2014). Beginning in the 20th century, agricultural activities and energy usage greatly increased inputs of reactive nitrogen species (N_2O, NO_x, NO_3^-, NH_3, and NH_4^+) to the ecosystem, changing the nitrogen distribution in the atmosphere and in terrestrial and aquatic systems. Cui et al. (2012) noted that, beginning in 1956 in China, reactive nitrogen resulting from anthropogenic activity surpassed that from natural sources, contributing over 80% of total reactive nitrogen by 2010. This has had significant negative impacts on global water quality, biodiversity, and ecosystem functions, resulting in serious environment problems, including eutrophication of rivers and lakes (Fenn et al., 2003a; Gruber and Galloway, 2008), groundwater pollution, soil nitrogen saturation, soil acidification (Guo et al., 2010; Liu et al., 2011; Matson et al., 2002), promotion of invasive species (Stevens et al., 2004), and the loss of biodiversity (Bobbink et al., 2010; Tilman, 1993; Liu et al., 2013). Cleveland et al. (1999) and Evans and Ehleringer (1993) noted that more systematic information was needed on all aspects of the sources, migration pathways, and storage of nitrogen to accurately understand the negative effects of the altered nitrogen cycle at global and regional scales.

Semi-arid and arid regions cover 30–40% of global land area and these areas are expanding (Dregne, 1991). Deserts and areas undergoing desertification are primary storage areas for terrestrial nitrogen

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(Walvoord et al., 2003). Nitrogen transfer into the atmosphere from terrestrial ecosystems is estimated at 102 Tg N yr^{-1} (Zhu and Xing, 2010); one-third of this occurs from deserts and areas undergoing desertification (Bowden, 1986).

Traditionally, the nitrogen cycle in arid regions has been assumed to occur mainly as a result of biological activity in the rooting zone in the upper vadose (unsaturated) zone. With increasing depth, nitrogen concentrations decrease. Nitrogen compounds that migrate into the deep vadose zone are typically ignored in analyses of the nitrogen cycle (Peterjohn and Schlesinger, 1990). However, recent evidence indicates that high NO_3^--N concentrations occur deep in the unsaturated soil of the world's desert regions (Walvoord et al., 2003), indicating that nitrogen compounds can migrate to the deep vadose zone under natural conditions. This new understanding has made it necessary to expand research on the nitrogen cycle in arid zones to include the entire vadose zone. The vadose zone now appears to be an important component of the nitrogen cycle in arid regions (Menon et al., 2010).

The exchange of heat, water, momentum, and chemical substances between the surface of the Earth and the atmosphere are also influenced by the vadose zone. Hartsough and Biondi (2004), Edmunds and Gaye (1997), Walvoord et al. (2003), and Schwiede et al. (2005) found elevated NO_3^--N in the vadose zones of arid regions at concentrations that impact groundwater quality. This challenges the traditional assumption that nitrogen loss can be overlooked in desert ecosystems, and especially the migration of energy and materials within the vadose zone and its responses to climate (Boring et al., 1988; Peterjohn and Schlesinger, 1990). Additionally, protecting groundwater resources in arid regions from pollution is an increasingly serious challenge. Accordingly, improved understanding of the nitrogen cycle in the vadose zone of arid regions and its responses to past climatic conditions is important in the support of research on global climate change and hydrogeology (Hartsough et al., 2001; McMahon and Böhlke, 2006; Scanlon et al., 2006; Ma et al., 2012).

This paper describes work carried out near the edge of Dunhuang Mingsha sand dunes that combines stable isotopic and ion concentration analysis to identify nitrate sources and to study nitrate transformation relationships between precipitation and soil water. The principal goal was to improve the understanding of nitrogen migration and its accumulation and cycle in the vadose zone of high dunes in an extremely arid area.

2. Study area

The Mingsha sand dunes are located near Dunhuang city, Gansu province in northwest China (Fig. 1). The desert is about 40 km long and 20 km wide and is bounded by the Mogao Caves to the east and the Danghe reservoir to the west. The megadunes are generally about 100 m high, with some reaching 170 m. The megadunes, which have different shapes, consist mainly of fine sand with mean grain sizes between 0.22 and 0.25 mm. The Mingsha sand dunes are formed by wind deposition; the heights and morphology of the megadunes are controlled by the underlying landforms (Dong and Bian, 2004). The basement rock of the Dunhuang basin is of Tertiary age and consists of conglomerate, argillaceous sandstone, and muddy siltstone, all of which are calcareous mud. Unconsolidated Quaternary sediments, mainly sandy conglomerate, pebbly sandstone, and sandy loam, overlie the basement rock. These unconsolidated sediments serve as a reservoir for groundwater resources (Fig. 2) (Ma et al., 2013; He et al., 2015; Sun et al., 2016).

The groundwater of Dunhuang basin is divided into three systems: pore water within unconsolidated Quaternary sediments, interlayer water within Tertiary clastic sediments, and fissure water in pre-Mesozoic metamorphic and igneous rocks. The pore water of unconsolidated Quaternary sediments consists of phreatic water and confined water. The pore phreatic water is distributed mainly in the upper diluvial fan of the Danghe and Daquanhe rivers, the ancient river channel near Dunhuang city, and the alluvial lacustrine plain of the Danghe and Shulehe rivers. The pore confined water is located principally along the middle and lower diluvial plain and at the lower alluvial lacustrine plain. The climate of the region is very arid. Annual precipitation in the study area from 1955 to 2014 averaged 38.87 mm. Meteorological data were obtained from the Dunhuang Station (China Meteorological Administration-National Climate Center), located at the northern edge of the Mingsha Mountains. The mean monthly temperature ranges from −9.3 °C in January to 24.9 °C in July and averages 9.3 °C. The annual temperature difference between day and night reaches 34 °C. The average relative humidity is < 40%, and the annual evaporation can reach 2486 mm (Ma et al., 2013; Sun et al., 2016). A positive correlation is evident between annual precipitation and annual temperature (Fig. 3). Precipitation mainly occurs from April to September (Fig. 3). In general from 1955 to 2014, precipitation varied significantly throughout the year, with the greatest precipitation in summer and the least in winter (Fig. 4). Over the 60-year period, the average precipitation in spring, summer, autumn, and winter was 8.57 mm, 24.33 mm, 4.18 mm, and 2.53 mm, respectively. The maximum summer precipitation was 87 mm in 1979 and the minimum precipitation in summer was 3 mm in 1956, during the period of record, 1979 was the wettest year and 1956 was the driest.

The dunes are generally subject to northeasterly, northwesterly, and southerly winds, with frequent dry, hot winds and sandstorms. Zhou et al. (2015) reported that the megadunes are essentially stable as a result of the prevailing wind directions that are offsetting. Low, regional vegetation cover is dominated by the sparse xeric grasses and small shrubs, such as *Nitraria schoberi L.*, *Calligonum*, *Alhagi*, and *Haloxylon ammodendron*.

3. Sample collection and analysis

Unsaturated zone profiles (LT and GC profiles, elevation 1165 m) were collected at the edge of the Dunhuang Mingsha sand dunes on 10 September 2016 (late summer) using a 50-cm diameter, hollow-stem hand auger with interchangeable 1.5-m aluminum rods. The LT profiles were taken in bare sand and the GC profiles were taken in vegetated dunes. Both profiles were taken in sandy soil, near fruit tree orchards and were separated by approximately 15 m. Two additional profiles, within 5 cm of the original LT and GC profiles, were taken on 22 December 2016 (early winter). The four profiles were taken from the vadose zone of the dunes to a depth of 3 m; samples were collected at 25-cm intervals. To avoid moisture loss, samples were immediately sealed in polyethylene bags. Precipitation samples (39.9592°N, 94.3302°E, elevation 1406 m) were collected at Danghe reservoir in western of the Mingsha sand dunes from August 2014 to July 2016. All precipitation samples were collected in polythene bottles which were sealed immediately, then analyzed for NO_3^-, Cl^-, and NH_4^+ concentrations.

Fig. 1 shows the locations of the Mingsha sand dunes, soil profiles, and precipitation samples.

Moisture contents were determined by oven-drying the samples for 12 h at 110 °C. Ionic species were obtained from 50 g of each sand sample by elutriation with 50 ml of deionized water. After filtration through a 0.45-μm filter, the Cl^- and NO_3^- concentrations in the supernatant water were analyzed by ion chromatography (ICS-2500, Dionex, Sunnyvale, CA, USA, analytical precision reaching ng g^{-1} level). The chemical analyses were all performed at the Key Laboratory of Western China's Environmental Systems at Lanzhou University. The $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ of the sand samples were measured at the Laboratory of Applied Physics and Chemistry Analysis, Ghent University (Analytical precision of $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ were ± 0.2‰ and ± 0.5‰, respectively).

Wet deposition fluxes of NH_4^+-N, NO_3^--N, or Cl^- were calculated by the following two equations (Liu et al., 2006):