Massive atmospheric nitrate accumulation in a continental interior desert, northwestern China

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ABSTRACT

Large-scale nitrate deposits are rare on Earth's surface due to the high solubility of nitrate minerals. Exceptions are found in extremely old (to 14 m.y.) and hyperarid deserts such as the Atacama Desert in northern Chile, or the McMurdo Dry Valleys in Antarctica. The nitrate in both of these regions has been determined to originate from atmospheric oxidation of NO_x. Here we report a new type of massive atmospheric nitrate deposit, with resources equivalent to the Atacama deposits, in the Turpan-Hami area, northwestern China. This deposit is characterized by (1) a location in the center of a large continent; (2) young age (Pleistocene); (3) general enrichment near the surface rather than at depth; and (4) high spatial variability in the nitrogen and triple oxygen isotope composition within this arid region, the $\delta^{15}N$ ranging from 0.7‰ to 27.6‰, $\delta^{18}O$ from 30.2‰ to 46.7‰, and $\Delta^{17}O$ from 5.9‰ to 20.7‰. The Turpan-Hami nitrate deposit nitrogen and triple oxygen isotope composition is closely between those of the Mojave Desert (southwestern United States) and the Atacama Desert, suggesting that (1) Earth's low-middle latitudes (i.e., non-polar sites) have been receiving atmospheric nitrate deposition of similar nitrogen and triple oxygen isotope composition; and (2) local or regional isotope differences in nitrate deposits can be attributed to postdepositional processes, namely the difference in aridity and associated microbial activities.

INTRODUCTION

Nitrate deposits were important raw materials for gunpowder and fertilizer in recent human history (Barnum, 2003). Despite the fact that industrial production of nitrate using the Haber-Ostwald process has reduced the importance of natural nitrate resources, nitrate ores, especially their associated potassium or iodate deposits, are still in great demand in fertilizer and health industries (Manning, 2010; http://www.atacama.com/i/pdf/Atacama_FactSheet.pdf). A century ago, except for a few local cave deposits, natural nitrate was mostly sourced from the Atacama Desert, northern Chile, where nitrate deposits were found mantling much of the hyperarid landscape (Ericksen, 1983).

The source of the large quantity of nitrate deposits in the hyperarid Atacama Desert had been a subject of debate (Claridge and Campbell, 1968; Ericksen, 1981; Pueyo et al., 1998). Recent measurement of the triple oxygen isotope composition of nitrate, particularly the anomalous ^{17}O enrichment, however, places the origin of much of the desert nitrate deposits clearly in the domain of atmospheric chemistry (Böhlke et al., 1997; Michalski et al., 2004a). Another hyperarid yet frigid desert environment, the McMurdo Dry Valleys in Antarctica, also hosts unusually high concentrations of nitrate in surface soil. Triple oxygen isotope data indicate that the source of nitrate is exclusively atmospheric for this polar site (Michalski et al., 2005). Small-scale nitrate deposits in the arid Mojave Desert in the southwestern Unites States also have a substantial nitrate source from atmospheric deposition (Michalski et al., 2004a).

Atmospheric nitrate is formed by the oxidation of NO_2, the precursor of which is often NO_x (NO + NO_2) (Finlayson-Pitts and Pitts, 1999). Natural sources of NO_x include lightning, biomass burning, and soil emissions (Jaeglé et al., 2005; Logan, 1983). These sources and the responsible oxidants, O_3, hydroxyl radical, proxy radicals, and halogen oxides, are ubiquitous in Earth's atmosphere, and thus atmospheric nitrate should be formed in a wide geographical area and be transported thousands of kilometers. The main reason that atmospheric nitrate only accumulates in massive quantities at a few specific localities is not because of lack of sources but because of lack of preservation; the formation of massive surface nitrate deposits is facilitated by the age and aridity of a surface. The high solubility of nitrate minerals renders large quantities of nitrate deposits unlikely in wet and/or active surfaces. In fact, the major natural sources of NO_x (e.g., biomass burning, soil microbial emission, and lightning) occur globally in coastal sites, continental interiors, or open oceans. Therefore, we should predict that as long as a surface has been arid for a prolonged period, atmospheric nitrate will accumulate to a significant quantity.

Both the Atacama Desert and the McMurdo Dry Valleys are coastal sites. The Mojave Desert, where sporadic nitrate deposits are found, is also <300 km from the nearest coast. These known cases of desert nitrate deposits may have imparted a notion that atmospheric nitrate ores only exist near the coast. Following up on a preliminary finding of anomalous ^{17}O enrichment in nitrate from the Kumutag Basin, a subbasin in the Turpan-Hami area (Xinjiang, northwestern China; Li et al., 2010), we systematically sampled and measured the stable N and triple O isotope composition of nitrate deposits of diverse mineral phases and occurrences in the Turpan-Hami and adjacent basins in the northern edges of the Tarim Basin (Fig. 1). We demonstrate that

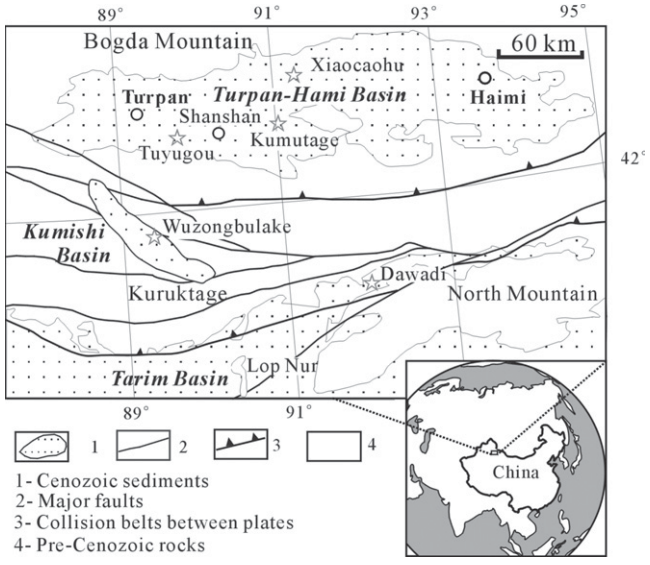

Figure 1. Locations and tectonic backgrounds of basins and mountains in Turpan-Hami area, Xinjiang, northwestern China.

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initial nitrate of atmospheric origin can be concentrated to massive ore deposits in continental interior basins.

GEOLOGICAL BACKGROUND AND NITRATE OCCURRENCES

Dotted with many inland basins, the Turpan-Hami area is almost in the very center of the Eurasian continent, at least 2500 km from any coastlines (Fig. 1). The intermountain basin is part of the north Tianshan fold system and is in a temperate continental arid climate zone. Moisture to the area comes from the west and the north or northwest, with a seasonal variation in source, and is obstructed by the high mountains of Tianshan to the north and west (Dai et al., 2007). Low elevations (from –154 m to 880 m) contribute to its extremely hot weather during the summer months. With an average annual rainfall of 15 mm and intense evaporation, often 200 times the rate of rainfall (Sun and Deng, 1987), it is one of the driest areas in the world. The basin also experiences strong northwest wind throughout the year, strongest during spring and summer months (Li, 1991), and has little surface vegetation.

Petrologic and geochemical data indicate that the Turpan-Hami area evolved from a backarc basin to a foreland basin over the Permian to Mesozoic and to the large intermountain basins of the Tianshan in the Cenozoic, during which time the dominant source of sediments has been the Bogda Mountains to the north (Greene et al., 2005; Shao et al., 1998). Late Pleistocene climate in the area appears to have alternated between a cold-humid glacial period and a warm-arid interglacial period (Luo et al., 2009). Palynology data also indicate that this area underwent further drying from the late Pleistocene to Holocene (Pan, 1993).

Nitrate deposits are widely distributed in the Turpan-Hami area (Zhang et al., 2000). The estimated total nitrate resource is 2.6×10^9 t (Li et al., 2010), equivalent to that of the Atacama Desert, where commercial-grade nitrate ores were estimated as 200×10^6 t, and several times more than that for lower grade ores (Ericksen, 1983). These nitrate deposits were discovered and mined sporadically in the late 1950s; after some preliminary scientific investigation in the 1990s, extensive survey and exploitation started in 2006. Today, potassium and nitrate are the two main mineral resources being mined; yearly production of $NaNO_3$ is 3×10^6 t (Qiu et al., 2009). The most common nitrate deposits in the Pleistocene conglomerates usually are concentrated in the upper 50 cm of a vertical profile, and are found toward basin centers at least 15 km away from mountain foothills.

A study based on ^{14}C and optically stimulated luminescence dating, sediment packages, and their estimated sedimentation rates shows that gypsum was first deposited at 260 ka, and abundant glauberite [$Na_2Ca(SO_4)_2$] precipitated after 160 ka in the Lop Nur area (Wang et al., 2001). This suggests that nitrate ores in the Turpan-Hami area are younger than at least 260 ka, much younger than nitrate deposits in the Atacama Desert and the McMurdo Dry Valleys, which have been accumulating since probably 15 m.y. ago (Nishiizumi et al., 2005) or even longer (Dunai et al., 2005).

GEOCHEMICAL AND STABLE ISOTOPE RESULTS

Field surveys of nitrate deposits, salt mineralogy, and vertical distribution reveal that nitrate is preferentially concentrated near the top (Table DR1 in the GSA Data Repository[1]). This pattern is especially consistent in the Turpan area (Yang, 1999) and could be less so if the entire arid region is considered. For example, two of the four profiles show this pattern in our measured profiles (Fig. DR1 in the Data Repository).

For analytical methods used to determine stable nitrogen and triple oxygen isotope composition of nitrate, see the Data Repository. We calibrated our triple oxygen isotope ratio measurement using NBS-28 (silica sand) assuming $\Delta^{17}O = 0.00‰$ when a slope value of 0.52 is used. Our measurement of USGS-35 yielded a $\Delta^{17}O$ of 21.8‰ ± 0.5‰ (1σ) (n = 4), in comparison to the recommended value of 21.1‰ ± 0.2‰ (Böhlke et al., 2003). Nitrate deposits in the Turpan-Hami area have a wide range of $\delta^{15}N$ values, from 0.7‰ to 27.6‰; most are 2‰–6‰. Sodium nitrate measurements at Xiaocaohu, Kumutag, Tuyugou, and Shaer had lower $\delta^{15}N$ values, ranging from 0.7‰ to 6.0‰, whereas niter values at Wuzongbulake are higher, ranging from 15.0‰ to 27.6‰. The $\delta^{18}O$ of nitrates ranges from 30.2‰ to 46.7‰, with the majority at 40‰–46‰. The $\Delta^{17}O$ ranges from 5.9‰ to 20.7‰, showing pronounced positive values (Fig. 2). Mineralogically, the $\delta^{18}O$ and $\Delta^{17}O$ of sodium nitrate deposits were higher than those of niter deposits; sodium nitrate has $\delta^{18}O$ and $\Delta^{17}O$ values ranging from 38.2‰ to 46.7‰ (average 41.8‰) and from 12.2‰ to 20.7‰ (average 16.2‰), respectively, whereas the Wuzongbulake niter has the lowest $\delta^{18}O$ and $\Delta^{17}O$ values in the Turpan-Hami area, ranging from 30.2‰ to 36.3‰ (average 31.9‰) and from 5.9‰ to 12.4‰ (average 7.4‰), respectively (Fig. 2). Overall, sodium nitrate has well-clustered isotope composition, while niter exhibits larger ranges (Fig. 2).

[1]GSA Data Repository item 2012182, analytical methods, Table DR1 (vertical sedimentological and mineralogical characteristics of soils), Table DR2 (a composite ion and isotope data), and Figure DR1 (nitrate sample sites, and four SO_4^{2-}, NO_3^-, and Cl^- concentration profiles), is available online at www.geosociety.org/pubs/ft2012.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

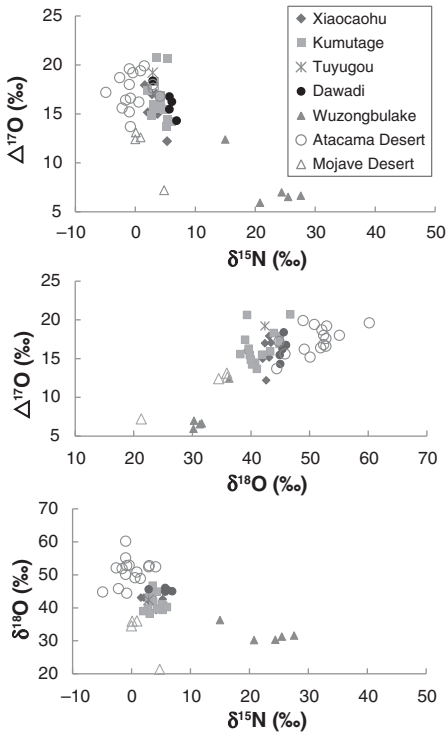

Figure 2. Site-specific nitrogen and triple oxygen isotope composition of nitrate ($\Delta^{17}O$, $\delta^{18}O$, and $\delta^{15}N$) from Turpan-Hami area, in comparison with those from Atacama and Mojave Deserts (data from Michalski, 2004a). Error bars are smaller than symbols.

The nitrogen and oxygen isotope composition in the Turpan-Hami area had no obvious correlation with latitude, longitude, or altitude, and no apparent trends in a vertical profile. However, lower $\Delta^{17}O$, lower $\delta^{18}O$, and higher $\delta^{15}N$ values are seen in a geographically narrower basin (e.g., the Kumishi) than in a broader basin (e.g., the Turpan-Hami or the Tarim) (Fig. 1; Table DR2).

DISCUSSION

Atmospheric Origin of Turpan-Hami Nitrate

The pronounced ^{17}O enrichment indicates unequivocally that the Turpan-Hami nitrate is predominantly of atmospheric origin, because among all original sources of nitrate only those generated from NO_x oxidation in the atmosphere have positive ^{17}O anomalies (Böhlke et al., 1997; Costa et al., 2011; Michalski et al., 2005, 2004a, 2004b; Savarino et al., 2007). This conclusion is supported by the associated high $\delta^{18}O$ values (30.2‰–46.7‰), which are in line with those obtained from the Atacama and the Mojave Deserts, 44‰–60‰ and 21‰–36‰, respectively (Böhlke et al., 1997; Michalski et al., 2004a). Excluding the niter at Wuzongbulake, the $\delta^{15}N$ of the Turpan-Hami, the Atacama, and the Mojave are similar, ranging from 0.7‰ to 6.9‰, –4.9‰ to 4.1‰, and 0‰ to 4.8‰, respectively (Fig. 2).

The $\delta^{15}N$ of atmospheric nitrate reveals the source of nitrogen or NO_x (Morin et al., 2009). The source of atmospheric nitrate, however, is apparently distinct for the McMurdo Dry Valleys because the $\delta^{15}N$ is extremely negative for this polar site, while close to the origin, the air N_2 value, for the Atacama and the Mojave. The origin of the extremely negative nitrate $\delta^{15}N$ in soils of the McMurdo Dry Valleys is debated. Michalski et al. (2005) advocated that the main source of the ^{15}N-depleted nitrate is stratospheric input, a view not echoed by Morin et al. (2009), who instead favored a mechanism of snowpack emission of NO_x at polar sites. Whatever the cause, the ^{15}N-depleted nitrate appears to be restricted to polar sites. This implies that atmospheric nitrate in low-middle latitudes should have somewhat similar $\delta^{15}N$ values, a point being confirmed by the Turpan-Hami nitrate deposits.

The triple oxygen isotope composition of nitrate indicates primarily the pathways involved in NO_x to NO_3^- oxidation (Michalski et al., 2003). Slightly different but relatively constant $\Delta^{17}O$ for atmospheric nitrate was reported for the Southern Hemisphere (26‰ ± 2‰) and the Northern Hemisphere (29‰ ± 2‰) in low-middle latitudes (Morin et al., 2009). The Turpan-Hami nitrate data suggest that the $\Delta^{17}O$ of initial atmospheric nitrate is higher than 20‰, similar to those found in other low-middle latitude sites in the world.

Heterogeneity of the Turpan-Hami Nitrate Isotope Composition

For geological nitrate deposits, additional processes such as mixing of air masses in the troposphere and millions of years of deposition, accumulation, and physical mixing at the surface environment are expected to further homogenize temporal and spatial differences that may have initially existed in the $\delta^{15}N$, $\delta^{18}O$, and $\Delta^{17}O$ of atmospheric nitrate. This implies that any difference seen in the isotope composition of surface nitrate deposits in low-middle latitudes is likely to be the result of postdepositional processes.

The observed variability of the nitrogen and triple oxygen isotope composition within the Turpan-Hami area suggests two possibilities. (1) Initially, all nitrate originated from atmospheric deposition; microbial processes have altered the isotope composition of nitrate to different degrees. (2) There are a few nonatmospheric sources of nitrate in the basin that have mixed into a surface nitrate pool that is overwhelmingly atmospheric, followed by different degrees of microbial processing in the nitrate pool. The role of microbes is evident from the fact that a decrease in $\Delta^{17}O$ value is correlated with an increase in $\delta^{15}N$ and a decrease in $\delta^{18}O$ values (Fig. 2). Microbial denitrification would increase the $\delta^{15}N$ and $\delta^{18}O$. However, nitrification would decrease the $\delta^{18}O$ of initial atmospheric nitrate by incorporating one oxygen atom from the ambient water to nitrite that may or may not have exchanged oxygen with water extensively (Buchwald and Casciotti, 2010); the process also decreases or even erases the ^{17}O anomaly (Tsunogai et al., 2011). The observation that Wuzongbulake niter from the center of the Kumishi Basin has the lowest $\Delta^{17}O$ and $\delta^{18}O$ values and the highest $\delta^{15}N$ suggests that the Kumishi Basin has had more microbial processing, probably linked to more frequent standing-water occurrences in geological history than the other more open and wider basins in the region; this inference is consistent with the deposition of niter, a more soluble nitrate mineral phase than sodium nitrate. Specifically, that higher $\delta^{15}N$ values are associated with lower $\Delta^{17}O$ values suggests an overall higher flux of microbial denitrification than that of nitrification in these arid and semiarid surface and subsurface hydrological systems, an inference consistent with the observed net loss of nitrogen relative to the rainfall N/Cl ratio in another arid site in northwestern China (Gates et al., 2008).

The main nonatmospheric nitrate comes from fertilizer application or point sources often associated with piles of animal waste, none of which has an anomalous ^{17}O signal. Neither of these two nitrate sources, however, should have occurred in the geological past in this arid region. It is probable that older atmospheric nitrate deposits have been redissolved and reconcentrated in different parts of a basin. There is, however, no reason to suspect that the older atmospheric nitrate initially accumulated in the same area had to have a very different isotope composition in the past. Therefore, the heterogeneity in nitrate isotope composition in the Turpan-Hami area is the result of different magnitudes of microbial nitrogen cycling associated with spatial and temporal heterogeneity in surface-water and groundwater availability in the area.

Not considering Wuzongbulake niter, the Turpan-Hami nitrate is higher in $\delta^{15}N$, lower in $\delta^{18}O$, and similar in $\Delta^{17}O$ values compared to the Atacama Desert nitrate, but is higher in all $\delta^{15}N$, $\delta^{18}O$, and $\Delta^{17}O$ values than the Mojave Desert nitrate, indicating an overall level of aridity between that of the Atacama and the Mojave Deserts for the Turpan-Hami area. The Wuzongbulake niter is collected from an often waterlogged salt flat and its isotope composition is apparently close to the Mojave nitrate. However, the limited data show much lower $\delta^{15}N$ values for the Mojave nitrate, suggesting that the $\delta^{15}N$ may be more complicated than the $\delta^{18}O$ and $\Delta^{17}O$ in relation to water availability. An important future task is to understand microbial N cycling and its dynamics in the niter-rich saline water bodies in this area.

Basinal Context and Nitrate Transport-Deposition Model

Although not universally true for the sites we sampled, the general vertical zonation of nitrate, chloride, and sulfate with increasing depth, especially the fact that nitrate is often concentrated on the surface rather than at depth, is the opposite of the Atacama salt profile, where nitrate is generally found below sulfate and chloride and is concentrated at depth (Ericksen, 1983). Capillary force accompanying evaporation can result in more soluble mineral phases migrating from depth and precipitating closer to the surface. A simple explanation is that in the Turpan-Hami area, heavy rain events in mountains raised the groundwater table in adjacent low-lying basins. The high water table dissolved early precipitated salts, and these salts were transported toward the center of a basin and redistributed vertically by evaporation and capillary force (Fig. 3). This model also explains why significant nitrate deposits are only found at least ~15 km away from foothills. Water transportation and evaporative concentration are required for the enrichment of nitrate content in the Gobi conglomerates in the Turpan-Hami area. In the Central Depression of the Atacama Desert, however, there were very few cases in which groundwater tables were elevated high enough to redissolve and redistribute salts upon evaporation. In the Atacama, the more soluble salt minerals are leached deeper in soil profiles, a manifestation of a much more arid climate than in the Turpan-Hami area. This model also explains why the massive nitrate deposits in the Turpan-Hami area are probably younger than 260 ka, a fraction of the time required for the

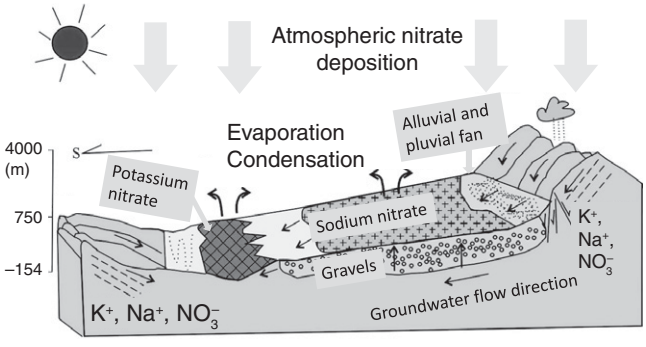

Figure 3. Conceptual model for nitrate deposits and their corresponding geographic factors in Turpan-Hami area.

Atacama deposits to form. Apparently, a major portion of the Turpan-Hami nitrate was deposited before 260 ka in the arid surfaces; 260 ka is just an episode when many of the older nitrate deposits were mobilized, transported, and reconcentrated in alluvial-pluvial deposits.

CONCLUSION

The discovery of nitrate deposits in what is probably the most inland site of any continent supports the notion that the critical condition required for atmospheric nitrate to accumulate to massive quantities is not the sources of nitrate, but the arid condition and stability of a surface over time. Periodic groundwater recharges from surrounding mountains and surface evaporation have both helped the transport and concentration of nitrate in the low-lying areas of many basins in the Turpan-Hami area. This nitrate accumulation mechanism is different from that for most of the Atacama Desert nitrate deposits, where groundwater and capillary processes have played minimal roles in much of the Central Depression. The $\delta^{15}N$, $\delta^{18}O$, and $\Delta^{17}O$ values of Turpan-Hami nitrate are similar to those in other low-middle latitude sites, supporting a comparable atmospheric chemical origin for low-middle latitude atmospheric nitrate. The large spatial variability in the Turpan-Hami nitrate isotope compositions reflects different degrees of postdepositional reworking of the nitrate within an arid area. Of immediate scientific interest are the saline-lake microbial metabolic processes that are responsible for the observed decrease in the $\delta^{18}O$ and $\Delta^{17}O$ with a corresponding increase in the $\delta^{15}N$ for nitrate.

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