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Atmospheric significance of aeolian salts in the sandy deserts of northwestern China

B.-Q. Zhu
Key Laboratory of Water Cycle and Related Land Surface Processes, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

Abstract: Large sandy deserts in the middle latitudes of northwestern China were investigated for soluble salt variations in modern and ancient aeolian sediments, aiming to explore the environmental significance of "aeolian salts". Results revealed that aeolian salt variations have a clear relationship with the changing meridional and zonal gradients of the desert locations and the aeolian differentiation effect, but are weakly linked to local geological conditions. It suggests that the natural system of aeolian salts is hydrologically open and the chemistry of the parent brines are different from that predicted for hydrologically closed systems. Atmospheric depositions of water-soluble chemical species are an important process/source contributing to aeolian salt. Sequential variations of soluble salts in sedimentary profiles interbedded with aeolian and non-aeolian deposits and their palaeoenvironmental implications in the hinterland areas of these deserts were further evaluated, based on the constraints of OSL dating and radiocarbon dating data. The results indicate that the inorganic salts may be a latent geoproxy in revealing regional palaeoclimatic changes in desert areas for the sediments deposited under onefold depositional environment, but the interpretation should be more careful for the sediments deposited under diverse depositional conditions. This study presents the evidence of atmospheric origin of aeolian salt in sandy deserts, with limited climatic significance in paleoenvironmental reconstruction.

Keywords: aeolian salt archive; environmental geoproxy; atmospheric deposition; palaeoclimate change; middle-latitude sandy desert; northwestern China

1. Introduction

Salt deposits (evaporites) are found in locations as diverse as Antarctica to the equatorial latitudes, and in depositional settings ranging from intracontinental to marginal marine (Handford, 1991). Much of what we know about ancient sedimentary facies and their depositional environments was derived from the study of modern sedimentary environments. Formation of soluble/inorganic salts in many types of deposits (e.g., soils, playa and lake sediments) is supposed to be governed mainly by climate factors including annual precipitation inputs, soil moisture changes, evapotranspiration losses, and solar radiation (Borchert and Muir, 1964; Sinha and Raymahashay, 2004; Warren, 2006; Singer, 2007). Thus salt archives in sediments are critical to understanding a number of geochemical processes in the supergene environment of the hydrogeologic and hydroclimatic systems. For instance, the soluble salt mineral assemblages embody significant signals indicating the processes of the solutes origin and transportation after their deposition (Warren, 2006; Zhu and Yang, 2010). Because soluble salts in soil/sediments response sensitively to regional-scale climate and hydrology changes (Borchert and Muir, 1964; Smoot and Lowenstein, 1991), they have been used as a key environmental proxy indicator in sedimentary records of oceans, lakes, sandy deserts and loess-paleosol sediments (Borchert and Muir, 1964; Sinha and Raymahashay, 2004; Hay et al., 2006; Warren, 2006; Sun et al. 2006; Liu et al., 2008; Sun et al. 2008). In these case studies, vertical variations of salt solutes in sediment strata are used as a geoproxy to indicate past salinity conditions and climatic changes in many parts of the world (Wasson et al., 1984; Last, 1990; Dean and Schwalb, 2000; Schutt, 2000; Last and Vance, 2002; Sinha and Raymahashay, 2004).

In arid conditions the desert landscape is globally distributed. For these areas, non-marine salt deposits are common features of modern arid closed basins. Researches on salt deposits under arid climate have been documented over a long time, however, the environmental implications of inorganic salt formation in desert areas are still not clear, to a certain extent because of the diverse compositions of salt solutes with complicated formation mechanisms in different geomorphologic units. Researches about salt formation have been performed worldwide in different arid conditions, with the main purpose to identify the relationship between salt regime and environmental factors, for instance, salt formation in different clay type (Rengasamy et al., 1984), hydrological distribution (Borchert and Muir, 1964; Warren, 2006), fire ashes (Lasanta and Cerda, 2005; Bodi et al., 2012, 2014), soil erosion (Lasanta and Cerda, 2005; Cerda et al., 2013; Agata et al., 2015), hydrological distribution (Borchert and Muir, 1964; Warren, 2006; Bodi et al., 2012), geomorphologic setting and dust source (Wang et al., 2008), volcanism and atmospheric fixation (Oyarzun and Oyarzun, 2007), rainfall patterns (Ahuja, 1990; Dragovich and Dominis, 2008) and mean annual climatic conditions (Dan and Yaalon, 1982; Lavee et al., 1991; Pariente, 2001), but relatively few have been recognized in the aeolian sediments. Until now, few investigations have been involved into the environmental implications of salt deposits from the sandy deserts in northern China (Zhu and Yang, 2010; Zhu et al., 2012), although these deserts comprise of the majority of middle-latitude deserts in the temperate zone of the Northern Hemisphere (NH).

In this work we present a physical and geochemical exploration into inorganic solutes filtrated from aeolian sediments collected from the inland sandy deserts in northwestern China, representing a cross-section of different environments (from westerly to monsoon climate control) in the NH middle latitudes. The objective of the paper is to explore the atmospheric significance of inorganic salts in aeolian sediments and their possible climatic ~~i~~implications.

2. Methodology

The study areas and sampling sites were chosen from famous sandy deserts in China (Fig. 1a), i.e., the Taklamakan Desert in the Tarim Basin (the westerly climate control) and the Badain Jaran~~Badanjilin~~ Desert in the Alex~~ashan~~ Plateau (the monsoon climate control). Both the modern and ancient aeolian sediments were designed to be sampled in the field. For modern aeolian sediment, thirty-three dunes in the Taklamakan Desert and fourteen dunes in the Badain Jaran~~Badanjilin~~ Desert were sampled in the field. The modern sands were sampled from the active dune

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surface. The geographical locations of these sample sites are mainly situated in dune fields from the center and southern edges of the Taklamakan Desert (Fig. 1b) and the southern inland area in the Badain Jaran~~Badanjilin~~ Desert (Fig. 1c-d).

In the Inner part of the Taklamakan and the Badain Jaran~~Badanjilin~~ Deserts, aeolian and lacustrine (or fluvial in Taklamakan) deposits (palaeosediments) are frequently found interbedded (Fig. 2). For instance, lacustrine/fluvial sediment strata widely occur in the hinterland dune fields of the Taklamakan Desert along the N-S oil-transporting highway (Fig. 2a-b). Four sedimentary sequences with clearly stratigraphical layers were selected from hinterland areas of the two deserts in this study (Fig. 1). It is about 4 m in depth for the Arerjilin-I section from the Badain Jaran~~Badanjilin~~ Desert (Fig. 1d). Lacustrine sediments in this section were interbedded by aeolian sand layers (Fig. 3a). For the Tazhong-XIII section in the Taklamakan Desert (Fig. 3b), it is general a lacustrine sequence interlayered with dune sediments. The Yaogan-VIII section is situated at the lower reaches of the Keriya River (Fig. 1b) close to the south margin of the Taklamakan Desert. The top of this section is buried by a 30-m active dune (Fig. 3c). The Tumiya-II section is located at the low reaches of the Tumiya palaeochannel close to the south edge of the Taklamakan Desert (Fig. 1b). This section is general a silty sand or sandy silt sequence interlayered by cemented calcium-carbonate layers (Fig. 3d). In general, lithologies of the lacustrine sediments are greatly different from those of the aeolian sediments in all sedimentary sequences from the Taklamakan Desert. These sequences were chronologically analyzed based on optically stimulated luminescence (OSL) dating and radiocarbon (^{14}C) dating methods.

The analytical methods for soluble salt geochemistry of aeolian sediment samples can be seen from Zhu and Yang (2010), and the OSL and radiocarbon dating methods can be seen from Yang et al. (2006, 2010). The physical and chemical analytical data of the soluble salt compositions of sediments are shown in On-line Supplementary Table 1 and Table 2, respectively. The resulting OSL and radiocarbon dating ages are summarized in On-line Supplementary Table 3 and Table 4, respectively. Note that the partial characteristics of aeolian salts from the Taklamakan and Badain Jaran~~Badanjilin~~ Deserts were previously reported in our early works (Zhu and Yang, 2010; Zhu et al., 2012) with preliminary descriptions of their composition and distribution and related influencing factors, but their complete stratigraphies and chronologic results are presented here along with other new palaeoenvironmental estimations.

3. Results

Salinity values for all samples are mathematically calculated based on TDS values of the filtrates and their water-sediment mass ratios. The pH and the calculated salinity values for the dune sediments from the Taklamakan Desert range between 8.66 and 9.54 (mean 9.22) and 0.27‰ and 1.86‰ (mean 0.78‰), respectively. For the dune sediments in the Badain Jaran~~Badanjilin~~ Desert, the pH and the salinity values lie between 8.82 and 9.42 (mean 9.12) and 0.05 and 0.42‰ (mean 0.15‰) , respectively. The range of pH values between the aeolian sediments is narrow (8.6-9.5). It indicates that the soil conditions and acid buffer capacities for these deserts are similar, generally in alkaline nature. Studies have proved that pH values in soil between 6.4-12.2 are mainly caused by bicarbonate (6.4-10.3) and carbonate (10.3-12.2) (Wetzel and Likens, 2000). It thus means that the alkalinities of aeolian salts in Chinese deserts are mainly controlled by the carbon-bearing salts, particularly bicarbonate.

For the 15 palaeo-aeolian sediment samples buried beneath modern dunes in the Taklamakan Desert with OSL ages ranging between 40-2 ka B.P. (On-line Supplementary Table 4), The salinities and pH values are slightly higher in salinity and lower in pH than those of modern dune sand samples (Fig. 4a). The modern dune samples collected from the identical months in different climatic year (such as the autumns in the arid 2006 and in the wet 2008) in the Badain Jaran~~Badanjilin~~ Desert are quite similar to each other (Fig. 4b).

The major anions of aeolian salts are Cl^- and HCO_3^- in all samples. The SO_4^{2-} concentrations are relatively low (Fig. 4d). Na^+ is the first major cation and Ca^{2+} is the second. The concentrations of K^+ and Mg^{2+} are relatively low (Fig. 4e-f). It should be noted that the carbon-bearing ion concentrations in these sediments are highly correlated with pH (Fig. 4c). Based on charge balances and mass matches between major cations and anions, the potential salt mineralogy of these aeolian sediments can be categorized as: $NaHCO_3$ + NaCl + ($CaCO_3$) in the Badain Jaran~~Badanjilin~~ Desert (Fig. 4g), and NaCl + $NaHCO_3$ in the Taklamakan Desert (Fig. 4h). It indicates that the assemblage of sodium carbonate and chloride is widely distributed in aeolian sediments within the Chinese sandy deserts.

The fingerprint diagrams of the ion distribution patterns for aeolian sediment solutes, local groundwaters and surface waters are shown in Fig. 5 a-b. The distribution patterns for dune surface sediments from the Badain Jaran~~Badanjilin~~ Desert are different from the local groundwaters and the local lake waters (Fig. 5a). The chemical differences between aeolian salts and natural waters are also evident in the Taklamakan Desert (Fig. 5b), particularly in the patterns of magnesium and sulfate. These factors reflect a weaker influence of local waters on aeolian salts both in the Badain Jaran~~Badanjin~~ and Taklamakan Deserts.

For the Taklamakan Desert, the salt salinities of aeolian sediments have a strong positive correlation with the median grain sizes of the corresponding samples (Mz in phi unit, Fig. 5c), indicating an enrichment of soluble salts towards the finer particles of aeolian sediments. A same trend is also observed in the Badain Jaran~~Badanjilin~~ Desert, as the salt concentrations in different gain size fractions increasing when particle fractions become finer (Fig. 5d).

In dune surface sediments from the tow deserts along the longitude (E) and latitude (N) lines of sampling sites, the salt salinities show regularly varied trends, with an increasing salinity together with the decreases of the longitude and latitude degrees, respectively(Fig. 5e-h). Because the mean annual precipitation in northern China increases gradually along the same meridional direction, and the temperature or mean solar radiation decreases along the same latitudinal gradients, this indicates that enrichment of aeolian salt contents have a clearly relationship with the changing meridional and zonal gradients of the sample locations, which correspond to the regional climatic gradient, as well as the aeolian effects of granularity differentiation caused by wind/atmospheric dynamics, but are weakly linked with local hydrological conditions.

For the Arerjilin-I section in the Badain Jaran~~Badanjilin~~ Desert, black carbon from the highest shorelines of Arerjilin Lake (+10 m) were dated to 5628±221 cal yr BP (CNR-185, Fig. 3a and On-line Supplementary Table 3). Calcareous gyttja (+6 m) and black carbon (+5.5 m) from deposits above the lake surface was dated to 4757±315 cal yr BP (CNR-173) and 7144±200 cal yr BP (CNR-186), respectively (Fig. 3a and On-line Supplementary Table 3). Normally, deposits are younger at locations below the highest lake level. However, in this case it is believed that younger sediments were removed by wind erosion. Consequently, the older lacustrine sediments are now exposed to the surface.

The Tazhong-XIII section contains three lacustrine layers interbedded by aeolian sand sediments (Fig. 3b). The OSL age of the bottom aeolian layer is 39,800±2900 a (On-line Supplementary Table 4). Aeolian layers underlying the top two lacustrine layers are dated to 28,000±2300 a and 29,200±2600 a. The age limits of the lower lacustrine layer range between ca 40,000 and 30,000 a ago (Fig. 3b).

Lacustrine strata in the Yaogan-VIII section are intercalated with two aeolian sand layers (Fig. 3c). The aeolian sands underlying the lacustrine section is dated to 14,500±1100 a (On-line Supplementary Table 4), and the aeolian sands overlying the lacustrine section is dated to 2320±180 a. The age constraints of aeolian sand layers indicate that the paleolake was dried between ca 14,000 and 2000 a (Fig. 3c).

In the Tumiya-II section that deposited on the palaeoterrace of the Tumiya River near the south margin of the Taklamakan Desert, The OSL chronology is dated to between 23,700±1800 a and 8700±800 a (On-line Supplementary Table 4). The accumulation of sandy loess at this section indicates that southern margin of the Taklamakan Desert at the time between 23,700±1800 and 8700±800 a was wet.

The above chronological data show that the OSL data hold great uncertainty. It may be necessary to discuss the uncertainty in different locations in this study. As previously reported, textures such as grain size and shape are important parameters influencing the OSL dating of sediment (Aitken, 1985; Fain et al., 1999; Brennan, 2003; Guerin et al., 2012; Duval et al., 2015), since they have a direct impact on several correction factors that are used for evaluating the dose rate. The aeolian sand layers for OSL dating in the Tazhong-XIII section have a mean particle sizes of 3.3-3.5 phi (sieving result), which is much coarser than the interbedded lacustrine deposits (mean particle size 5.8-6.7 phi). The aeolian sand layers in the Yaogan-VIII section have a mean grain size of 3.8 phi, also coarser than the intercalated lacustrine deposits (mean particle size 4.6-6.2 phi). Aeolian silt or sandy loess deposits in the Tumiya-II section have a mean grain size of 3.6-4.0 phi. These data show a large variability in term of size of grains between the samples for OSL dating in the Taklamakan Desert. Because there is a distinction between the sieve aperture size, or sieve opening, and the diameter of a particle, consequently, the main dimension controlling the sorting of the grains for non-spherical particles is the so-called intermediate diameter. For example, for standard mesh of 50, 100, 200 and 300 mm, grains passing through may have an intermediate dimension in the range of from 50 to 71 mm, 100-141 mm, 200-283 mm, 300-424 mm, respectively (Duval et al., 2015). Besides, the grain size distribution of the raw sediment may have also an impact on the dating results, like in the case of a bimodal distribution (Duval et al., 2015). So the principal sources of uncertainty in OSL dating data of this study may potentially be derived from the grain size of aeolian sediments. On the other hand, the difference in ^{14}C ages between the uppermost and lowermost organic carbon layers

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of a sediment profile usually underestimates the time of sediment deposition, and the difference in OSL ages taken from sediment units overlying and underlying a buried soil most likely overestimates it (Miao et al., 2016). For example, the comparison of OSL and ^{14}C ages suggests that the radiocarbon dating technique may significantly underestimate the age of sediments for samples older than 30 cal ka BP (corresponding to ~25 ^{14}CkaBP) (Long et al., 2015). This factor could also lead to a certain uncertainty in the chronological data of this study.

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4. Discussion

4. 1. Primary or secondary salt in origin?

Firstly, it is necessary to understand whether the nature of soluble salt in aeolian sediment is primary or secondary in origin. As defined by Warren (2006), an original salt deposit (evaporite) is hydrologically driven by solar evaporation and is sourced from saturated brine on ground surface or nearsurface. In order to emphasize the highly reactive nature of soluble salts in the sedimentary realm, such salts that is precipitated from surface brine and retaining crystallographic evidence of the depositional process are regarded as primary salt. According to this definition and the related two limiting factors, almost every subsurface salt texture is secondary, because it is diagenetically altered, frequently with fabrics indicating pervasive early recrystallization. Secondary salts can be formed in surface/subsurface settings equivalent to the eogenetic, mesogenetic and telogenetic realms (Choquette and Pray, 1970). Under this definition of a primary versus secondary salt, we can say that, outside of a few Neogene examples (Riding et al., 1998; Valero-Garces et al., 1999, 2001; Pedley et al., 2003), there are few salt deposit with textures that are wholly and completely "primary". Without exception, soluble salts in aeolian sediments should be secondary salt originated in the eogenetic realm.

According to the salt category defined by Warren (2006), salt minerals can be simply subdivided into evaporitic alkaline earth carbonates and evaporite salts. In a view of depositional process, evaporitic alkaline earth carbonates are the firstly-precipitated salt minerals that sourced from concentrating hypersaline water. Compared with the evaporite salts, they are tend to be formed during the early stages of surface brine concentration-crystallization, while the evaporite salts are formed during the more saline stages of concentration-crystallization process (Smoot and Lowenstein, 1991). As analyzed above, $NaHCO_3$ + $CaCO_3$ are the identical components in dune sediments from both the Taklamakan and the Badain Jaran~~Badanjilin~~ Deserts. It indicates that evaporitic alkaline earth carbonates are major component of these aeolian salts.

Although texture of most of the subsurface salts is secondary, the earliest secondary salts are often syndepositional precipitates (Warren, 2006), with formation of cements and replacement even as the primary matrix accumulates around them. For example, nodular anhydrite and aragonite were recognized in Permian mudflats by Kerr and Thomson (1963), they interpreted it as a subaqueous saline pan indicator. Works by Gerdes et al. (1994, 2000) have shown that carbonate grains, such as ooids and peloids, typically thought of as indicators of marine conditions and mechanical agitation, can precipitate in situ in $CaCO_3$-saturated evaporitic settings. For aeolian sediments in arid environment, we image that much of surface salts are deposited in multiple episodes of early diagenetic (syndepositional) cementation. This syndepositional salt may be formed in multiple dissolution-precipitation events in the sand particle surface and are precipitated between successive depositional episodes of salt crust formation.

Besides alkaline earth carbonates ($NaHCO_3$ + $CaCO_3$), NaCl is another major component of aeolian salts in the deserts studied. This salt, dominantly halite, is evaporite salts according to the classification by Warren (2006) that precipitated from concentrated brine after the brine has crystallized the alkaline earth carbonates. Both the evaporitic alkaline earth carbonates and evaporite salts coexist in the aeolian sediments of the deserts, indicating that aeolian salts are complex salt mixture formed by multiple evaporation and precipitation processes in the eogenetic realm.

4. 2. Possible mechanism of aeolian salt formation

All evaporite salts are ionic salts. The definition of a salt deposit (evaporite) proposed by Warren (2006), as mentioned above, has no assumption as to the origin of the parent brine. It may be marine, nonmarine or a hybrid (Smoot and Lowenstein, 1991). By definition, there is a need for aridity and for water loss to exceed inflow (Borchert and Muir, 1964). This means deposition and diagenesis in evaporites is more dependent on climate than in either siliciclastic or carbonate sediments. Therefore, modern continental evaporites typically accumulate in semi-arid to hyperarid deserts. Suites of precipitated salts follow the geochemical make-up of the parent brine (Braitsch, 1971). It means that the mineralogy and order of precipitates in any brine pool are controlled by the ionic make-up of the parent brine. This provides a clue to reveal the possible source and mechanism of salt formation under particular environment.

Generally speaking, eight mechanisms or processes precipitate most salt minerals and crusts worldwide: 1) in-situ pedogenic weathering and eluviation processes of existing salty or volcanogenic (ash) parent materials; 2) eolian or fluvial input of salt or salt-rich sediment; 3) upward movement of salt via capillary flow from a shallow salty groundwater, 4) an oceanic aerosol source, creating in salt-enriched rainfall that evaporatively concentrates within the regolith, 5) intrusion and flooding by seawater, 6) decomposition of vegetation, 7) in-situ oxidation of sulphide minerals, 8) anthropogenic pollutants (Petrov, 1976; Smoot and Lowenstein, 1991; Warren, 2006; Singer, 2007). The first two sources are direct evaporite associations, while the third is typically associated with salt lakes, sabkhas and playas in semi-arid or desert settings within 500-1000 km of the coast. Many salt soil horizons are combinations of both pedogenically precipitated and detrital wind-reworked salt (Warren, 2006; Singer, 2007). Based on the eight views, we can get clues into the possible mechanism of aeolian salt formation in Chinese deserts.

To the weathering mechanism in view (1), our previous study has shown that the degree of weathering of aeolian sand in the Taklamakan is very low (CIA $\approx$ 50) (Zhu and Yang, 2009), which is similar to that of the un-weathered Upper Continental Crust. This observation suggests that salt contributions directly from bedrock weathering are of minor significance. For the eluviations process in view (1), studies have demonstrated that in a region with a mean annual precipitation less than 500 mm, the export of erosible elements by eluviation processes in the soil will be less significant than their preservation by evaporation-crystallization processes (Birkeland, 1999; Kraimer et al., 2005). Since the mean annual precipitation in desert regions of northern China is lower than 400 mm (Fig. 1a), the degree of eluviation exerted on soluble salts in the two sandy seas is lower than that of preservation. The salt concentrations of samples buried in dune subsurface layers in this study are equal to or only slightly higher than those of the surface samples (Fig. 4a), indicating only a little downward movement of these salts is taking place. Case studies on salt movement in desert soil have also shown that only 1.64% of the rainfall-leached salt can reach 1.0 m depths, and only 0.02% of the rainfall-leached salt can penetrate 2.0 m below the ground surface (Marion et al., 2008). This means that salt contributions from eluviation processes of parent salts are also minor.

To the view (3), dunes in both the Taklamakan and Badain Jaran~~Badanjilin~~ Deserts are generally higher than 20~50 m (Zhu et al., 1985), particularly being higher than 200 m in the Badain Jaran~~Badanjilin~~ Desert, so it is impossible for groundwater to contribute salt to aeolian sediments by drawing upward to the dune surface via capillarity.

To the view (4) and (5), both the two sandy deserts studied are geographically far away from any ocean and have been under the control of terrestrial processes since at least the Cenozoic (Zhu et al., 1980). Salt contributions from present or ancient seawater and aerosol can, therefore, be neglected. However, contributions from oceanic-sourced rainfall precipitation (atmospheric wet deposition) will discuss below.

To the view (6), although the decomposition of vegetation may contribute to the salts of some desert areas (Petrov, 1976), but plants in/around the sampling sites of the deserts in this study are scarce, so any influence of vegetation is minimal.

To the view (7), Petrov (1975) suggested that the salt chemistry in the world deserts can be classified into four types: chloride, sulfate, carbonate and mixed. Many desert soils in arid to semi-arid environment are sulphate-rich in areas surrounding the perennial saline lakes, the brine pan depressions and their dune margins (Petrov, 1976; Smoot and Lowenstein, 1991; Warren, 2006). For example, gypsum crusts have been reported from all the continents, including Antarctic (El-Sayed, 1993). Studies have shown that chloride–sulfate salts but not bicarbonate salts are dominated in the Kalahari Desert (Wang et al., 2009). This is also the case for wide-distributed aeolian loess deposits in the Loess Plateau of northern China (Zhu et al., 2012), which are mainly composed of Na_2SO_4+$NaHCO_3$ for loess and of Na_2SO_4+NaCl+$NaHCO_3$ for paleosol (Sun et al., 2006). While, the analysis of this study shows that aeolian salts in the Taklamakan and Badain Jaran~~Badanjilin~~ Deserts are mixture of sodium bicarbonate and chloride. This is different from both the Chinese loess deposits and the global tropical/subtropical deserts. Although sodium sulphate (Na_2SO_4) is an important salt existed in loess-paleosol sequences, but less common in the dune sediments from the sandy deserts in this study. So mechanism of in-situ oxidation of sulphide minerals can be excluded for the aeolian salts in this study.

To the view (8), anthropogenic pollutants, generated by human activities, are usually inorganic ions such as NH_4^+, NO_3^-, $H_2PO_4^-$ and SO_4^{2-} and some organic anions.

The contents of all samples studied, however, have roughly equivalent numbers of cations and anions with low/none above-mentioned ions (On-line Supplementary Table 2), suggesting that human impact on salt concentrations across the two sandy deserts is negligible.

On the basis of all above results, only the view (2) is unexplained. Actually the mechanism in view (2) involves the hydrological and atmospheric processes of salt origin and transportation. We discuss it below.

4. 3. Atmospheric origin?

There are wide similarities but partial differences existing in the salt chemistry and ion distribution patterns of aeolian salts between the Taklamakan and Badain Jaran~~Badanjinlin~~ deserts (Fig. 4g-h). Besides, they are weakly correlated with the chemical compositions of the nearby ground waters under local hydrological setting (Zhu and Yang, 2010) (Fig. 5a-b), but are strongly associated with the regional climatic and wind regime (Zhu et al., 2012) (Fig. 5c-h). All these evidences indicate that the natural system of aeolian salts is hydrologically open and the chemistry of the parent brines and the associated salt deposits may be significantly different from that predicted for hydrologically closed systems. It means that the sources of aeolian salts in the deserts are strongly controlled by earth surface processes at a regional or large scale, but not at a local scale. Seen from this point, only an atmospheric origin but not hydrological origin can be responsible for this pattern.

It has been recognized that atmospheric chemical species is a significant source contributing acidic and eutrophic elements to both the terrestrial and ocean ecosystems (e.g., Sehmel, 1980; Wesely and Hicks, 2000; Inomata et al., 2009). For example, soil salts in many desert areas have been implicated to be originated from the atmospheric sources (Amit and Yaalon, 1996; Bohlke et al., 1997; Oyarzun and Oyarzun, 2007). Generally, there are two forms for atmospheric deposition, wet and dry deposition. Wet deposits contain salts derived from rainwater and snow. Studies have shown that chloride and carbonate are major species in rainfall in central Asia and north China (Fujita et al., 2000; Li et al., 2007; Zhao et al., 2008). It indicates that the chemical compositions between wet depositions and aeolian salts are similar.

Besides rainfall, aerosol and dust, known as atmospheric dry deposition, is another global phenomenon of salt transportation (Goudie and Middleton, 2006). At present there are few data being available for understanding the salt compositions of dust in the study areas. However, wide studies have proved that salt transportation and redeposition via dust are one of the important patterns for biogeochemical cycles in arid areas (Logan, 1974; Yaalon and Ginzboung, 1966; Reheis and Kihl, 1995; Al-Momani et al., 1995; Kiefert, 1997; Blank et al., 1999; Kulshrestha et al., 2003; Zheng et al., 2005; Abuduwaili et al., 2009). It should be a similar case for the hinterlands of northwestern China.

4. 4. Climatic implications

The coexistences of palaeo-lacustrine sediments (playa) and aeolian sediments are commonly seen in desert regions globally. These are clear evidences for past environmental changes in desert conditions (Smith, 1971; Yechieli and Wood, 2002;

Alonso-Zarza, 2003; Yang et al., 2006; Tripaldi et al., 2011; Dong et al., 2012), Therefore, the large number of playa deposits in the inter-dune depressions of these deserts offers an exceptional opportunity to study regional paleohydrological and paleoclimatic change. Over the past decade, there have been significant advances in our understanding of the late Quaternary history of the desert regions of northwestern China. OSL and ^{14}C chronologies of aeolian sediments from various locations in these deserts provide important archives to understand the timing of lacustrine/fluvial and aeolian processes related to environmental and climate changes. However, despite this increased interest, paleolimnological and palaeoaeolian research is limited by a paucity of potential geoproxy and the study sites with relatively long, uninterrupted stratigraphic sequences. Sequential variations of inorganic salts have been tentatively applied to reconstruct the palaeoenvironmental history in desert areas. For instance, taking the soluble salts variations in thick Cenozoic deposits as one of the indicators of paleoenvironmental change, Sun et al. (2008) reconstructed the aridity history of the Tarim Basin and argued that hyperarid climate had been prevailed within the basin since 5.3 Ma ago.

The variations in soluble salts content for the four sections collected from the Taklamakan and Badain Jaran~~Badanjilin~~ deserts are examined in this study, as shown in Figs. 6-9. The OSL and ^{14}C age constraints of these sequences are also shown in these figures. The data indicates that all of these playa deposits had experienced fluctuations in high moisture conditions during the period from late Pleistocene to mid-Holocene.

For the Arerjilin-1 section, there are an increase-decline pattern in salinity variation and a decline-increase-decline pattern in pH variation during the early-middle Holocene (Fig. 6). For the Tazhong-XIII section, there is a clear serrate shape (Fig. 7) in the pattern of salinity variation. It indicates that there are at least three times of humid environmental fluctuations during the past 40 to 28 ka B.P.. In the Yaogan-VIII section, the sequence salinities are relatively high (Fig. 8). It suggests that a saline lacustrine environment has occurred this area and the local hydrological settings have been retained the saline conditions for a long time from the end of the last glacial to late Holocene. In the Tumiya-II section, variations of the sequence salinities were unstable during the LGM (24 ka B.P.). There are two stages of salinity increase during the end of the LGM (about 10-11 ka) and the early Holocene (after 8.7 ka B.P.), respectively (Fig. 9). . Taken the four sections as a whole into consideration, there are evident differences between the aeolian sediments and fluvial sediments in salt content. In general, salinities in aeolian sediments are evidently lower than those of lacustrine sediments. It suggests that a great salt depositional discrepancy exists between the aeolian and the lacustrine sedimentary systems, which could be owing to the differential mechanisms of salt formation and origin in wind and water regimes. The results suggest that the inorganic salt is a latent proxy indicator for single depositional-environment sequence in revealing local hydrological variations and climate changes in desert areas during the late Quaternary.

However, the interpretation based on salt archives should be careful for the sedimentary sequences with dual/multiple depositional end-members. Research~~e~~ers have reported that in a sediment strata formed by an identical geological process like lacustrine or aeolian dynamics, the increase of sequence salinity commonly reflects a stepped-up environmental aridity (Sinha and Taymahashay, 2004; Warren, 2006; Liu et al., 2008). Because whatever under the aeolian and lacustrine depositional settings, the increase in salt content is a reflection of the enhanced evaporation potential. However, in a sediment profile controlled by multiple geological processes like combined aeolian and lacustrine dymanics, the increase in salt content could be not a reflection of the enhanced aridity, just like the case in this study. It is generally higher in salinity for the lacustrine sediments than for the aeolian sediments. In this study, the salinity increases in sections of the Arerjilin-I, Tazhong-XIII and Yaogan-VIII are general a representation of the enhanced wetter conditions (Figs. 6-8), because the higher salinities are always located in the lacustrine/fluvial layers. In general, lacustrine/fluvial strata in aeolian sequences always reflect enhanced humid periods than that of the aeolian period (Yang et al. 2006). However, this is not the similar conditions for the Tumiya-II section, as the salt increases in this sequence should be attributed to enhance aridity (Fig. 9), because the sedimentary structure of this section is uniformly dominated by aeolian process~~.~~. Thus it can be confirmed that once the coexistence of aeolian and fluvial/lacustrine sediments occurs in a sedimentary structure in desert regions, the single use of salt archives should be cautious in reconstructing the regional environmental changes. A combined use of salt archives and sedimentary proxies is recommended under this situation.

Besides, land surface processes such as chemical, physical and mechanical processes can weld younger to older sediment profiles and affect the accumulation, dissolution, and reprecipitation of mineral materials in sediments, as illustrated by Olson and Nettleton (1998). Sediment properties most affected include texture and the content and distribution of soluble salts. Processes such as erosion and deposition can truncate profiles or bury them either rapidly or extremely slowly. Effects of these and other processes on sediment properties must be examined with care in paleosols that have been buried even for short interval. The above-mentioned analogies of fluvial/lacustrine-aeolian profiles in the Taklamakan and Badain Jaran~~Badanjilin~~ deserts should be evaluated carefully in future studies and possible avoided as single indicators in paleoenvironmental reconstruction in the desert environment.

5. Conclusion

Formations of inorganic salt in aeolian sediments in the desert environment are significant to understanding the interrelationships between aoelian dynamics and regional hydrological and climate regimes. Geological information related to the inorganic salt composition and distribution and their environmental significance in aeolian sediments in the middle-latitudes deserts are still rare. Geochemistry of soluble salts in modern and ancient dune sediments from the sandy deserts in northwestern China was surveyed in this work. Variations of the salt composition around these sandy deserts are closely related to regional climatic parameters, such as the precipitation, temperature and wind agent. The aeolian salts are proposed to be sourced from the atmospheric depositions that are strongly associated with the local-scale and regional-scale dry and wet depositions, such as rainfall and dustfall precipitation. Single salt archive in sedimentary deposits interlayered by aeolian and lacustrine facies should be carefully used as geoproxy to reconstruct palaeoenvironmental histories in arid conditions.

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Figure Captions

Figure 1 Locations of the study areas and sampling sites in this study. (a) The distributions of deserts and the mean annual precipitation isohyets in China, (b) the Taklamakan Desert, (c) and (d) the Badain Jaran~~Badanjilin~~ Desert.

Figure 2 Photograph views of Quaternary and modern sediments of aeolian and lacustrine/fluvial facies that consisted of clay and sand/silt sand alternations in the Taklamakan and Badain Jaran~~Badanjilin~~ Deserts. (a) and (b) the Taklamakan Desert, (c) and (d) the Badain Jaran~~Badanjilin~~ Desert.

Figure 3 Sketch maps of the sedimentary profiles studied in this study. (a) Lacustrine sediments interbedded by aeolian sand layers with ^{14}C ages at the Arerjilin-I section in the Badain Jaran~~Badanjilin~~ Desert, (b) interbedding of aeolian and lacustrine deposits and their OSL ages at the Tazhong-XIII section in the Taklamakan Desert, (c) lacustrine deposits buried under a 30 m high dune at the Yaogan-VIII section in southern Taklamakan, and (d) sandy loess deposits with OSL dating results at the Tumiya-II section on southern margin of the Taklamakan Desert.

Figure 4 Salinities *vs.* pH of sediment solutions, (a) dune-surface sediments (modern sediment) and buried-dune sediments (palaeo-sediment) in the Taklamakan Desert, (b) dune-surface sand samples collected in different climatic seasons (dry and wet) in the Badain Jaran~~Badanjilin~~ Desert. (c) Carbon-bearing ions *vs.* pH. Triangular plots of major anions (d) and cations (e and f) of the soluble salts in mEq/l unit. Distribution patterns of the major ions of the soluble salts in the aeolian sediments from (g) the Badain Jaran~~Badanjilin~~ Desert and from (h) the Taklamakan Desert.

Figure 5 Distribution patterns of the major ions of aeolian salts, groundwaters and lake waters in (a) the Badain Jaran~~Badanjilin~~ Desert and (b) the Taklamakan Desert. The data of local lake water and groundwater bodies in the Badain Jaran~~Badanjilin~~ are cited from Yang and Williams (2003), and the river water and groundwater bodies in the Taklamakan from Zhu and Yang (2007). The relationship between salinities and grain size compositions of the aoelian sediments: (c) salinity *vs.* median particle diameter (Mz, in phi unit) of dune-surface sediment from the Taklamakan Desert, and (d) salinity *vs.* grain size compositions of randomly-selected dune sediments from the Badain Jaran~~Badanjilin~~ Desert. Spatial distributions of the salinities of the aeolian sediments along the longitudinal and latitudinal degrees in the Taklamakan Desert (e and g) and in the Badain Jaran~~Badanjilin~~ Desert (f and h), respectively.

Figure 6 Sequential variations in soluble salts contents (salinity and pH) of the Arerjilin-I section in the Badain Jaran~~Badanjilin~~ Desert.

Figure 7 Sequential variations in soluble salts contents (salinity and pH) of the Tazhong-XIII section in the Taklamakan Desert.

Figure 8 Sequential variations in soluble salts contents (salinity and pH) of the Yaogan-VIII section in the Taklamakan Desert.

Figure 9 Sequential variations in soluble salts contents (salinity and pH) of the Tumiya-II section in the Taklamakan Desert.

On-line Supplementary Table Captions

On-line Supplementary Table 1 Physical parameters of the soil-water extraction of the modern and ancient aeolian sediments from the Taklamakan and Badain Jaran~~Badanjilin~~ deserts in northwestern China.

On-line Supplementary Table 2 The analytical data of major ionic compositions for the soil-water extraction of the aeolian sediments from the Taklamakan and Badain Jaran~~Badanjilin~~ deserts

On-line Supplementary Table 3 Conventional radiocarbon ages for section samples from the Badain Jaran~~Badanjilin~~ Desert (cited from Yang et al., 2010)

On-line Supplementary Table 4 Summary data for OSL ages of section samples from the Taklamakan Deserts (cited from Yang et al., 2006).

Atmospheric significance of aeolian salts in the sandy deserts of northwestern China

B.-Q. Zhu
Key Laboratory of Water Cycle and Related Land Surface Processes, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

Abstract: Large sandy deserts in the middle latitudes of northwestern China were investigated for soluble salt variations in modern and ancient aeolian sediments, aiming to explore the environmental significance of "aeolian salts". Results revealed that aeolian salt variations have a clear relationship with the changing meridional and zonal gradients of the desert locations and the aeolian differentiation effect, but are weakly linked to local geological conditions. It suggests that the natural system of aeolian salts is hydrologically open and the chemistry of the parent brines are different from that predicted for hydrologically closed systems. Atmospheric depositions of water-soluble chemical species are an important process/source contributing to aeolian salt. Sequential variations of soluble salts in sedimentary profiles interbedded with aeolian and non-aeolian deposits and their palaeoenvironmental implications in the hinterland areas of these deserts were further evaluated, based on the constraints of OSL dating and radiocarbon dating data. The results indicate that the inorganic salts may be a latent geoproxy in revealing regional palaeoclimatic changes in desert areas for the sediments deposited under onefold depositional environment, but the interpretation should be more careful for the sediments deposited under diverse depositional conditions. This study presents the evidence of atmospheric origin of aeolian salt in sandy deserts, with limited climatic significance in paleoenvironmental reconstruction.

Keywords: aeolian salt archive; environmental geoproxy; atmospheric deposition; palaeoclimate change; middle-latitude sandy desert; northwestern China

1. Introduction

Salt deposits (evaporites) are found in locations as diverse as Antarctica to the equatorial latitudes, and in depositional settings ranging from intracontinental to marginal marine (Handford, 1991). Much of what we know about ancient sedimentary facies and their depositional environments was derived from the study of modern sedimentary environments. Formation of soluble/inorganic salts in many types of deposits (e.g., soils, playa and lake sediments) is supposed to be governed mainly by climate factors including annual precipitation inputs, soil moisture changes, evapotranspiration losses, and solar radiation (Borchert and Muir, 1964; Sinha and Raymahashay, 2004; Warren, 2006; Singer, 2007). Thus salt archives in sediments are critical to understanding a number of geochemical processes in the supergene environment of the hydrogeologic and hydroclimatic systems. For instance, the soluble salt mineral assemblages embody significant signals indicating the processes of the solutes origin and transportation after their deposition (Warren, 2006; Zhu and Yang, 2010). Because soluble salts in soil/sediments response sensitively to regional-scale climate and hydrology changes (Borchert and Muir, 1964; Smoot and Lowenstein, 1991), they have been used as a key environmental proxy indicator in sedimentary records of oceans, lakes, sandy deserts and loess-paleosol sediments (Borchert and Muir, 1964; Sinha and Raymahashay, 2004; Hay et al., 2006; Warren, 2006; Sun et al. 2006; Liu et al., 2008; Sun et al. 2008). In these case studies, vertical variations of salt solutes in sediment strata are used as a geoproxy to indicate past salinity conditions and climatic changes in many parts of the world (Wasson et al., 1984; Last, 1990; Dean and Schwalb, 2000; Schutt, 2000; Last and Vance, 2002; Sinha and Raymahashay, 2004).

In arid conditions the desert landscape is globally distributed. For these areas, non-marine salt deposits are common features of modern arid closed basins. Researches on salt deposits under arid climate have been documented over a long time, however, the environmental implications of inorganic salt formation in desert areas are still not clear, to a certain extent because of the diverse compositions of salt solutes with complicated formation mechanisms in different geomorphologic units. Researches about salt formation have been performed worldwide in different arid conditions, with the main purpose to identify the relationship between salt regime and environmental factors, for instance, salt formation in different clay type (Rengasamy et al., 1984), hydrological distribution (Borchert and Muir, 1964; Warren, 2006), fire ashes (Lasanta and Cerda, 2005; Bodi et al., 2012, 2014), soil erosion (Lasanta and Cerda, 2005; Cerda et al., 2013; Agata et al., 2015), hydrological distribution (Borchert and Muir, 1964; Warren, 2006; Bodi et al., 2012), geomorphologic setting and dust source (Wang et al., 2008), volcanism and atmospheric fixation (Oyarzun and Oyarzun, 2007), rainfall patterns (Ahuja, 1990; Dragovich and Dominis, 2008) and mean annual climatic conditions (Dan and Yaalon, 1982; Lavee et al., 1991; Pariente, 2001), but relatively few have been recognized in the aeolian sediments. Until now, few investigations have been involved into the environmental implications of salt deposits from the sandy deserts in northern China (Zhu and Yang, 2010; Zhu et al., 2012), although these deserts comprise of the majority of middle-latitude deserts in the temperate zone of the Northern Hemisphere (NH).

In this work we present a physical and geochemical exploration into inorganic solutes filtrated from aeolian sediments collected from the inland sandy deserts in northwestern China, representing a cross-section of different environments (from westerly to monsoon climate control) in the NH middle latitudes. The objective of the paper is to explore the atmospheric significance of inorganic salts in aeolian sediments and their possible climatic implications.

2. Methodology

The study areas and sampling sites were chosen from famous sandy deserts in China (Fig. 1a), i.e., the Taklamakan Desert in the Tarim Basin (the westerly climate control) and the Badain Jaran Desert in the Alex Plateau (the monsoon climate control). Both the modern and ancient aeolian sediments were designed to be sampled in the field. For modern aeolian sediment, thirty-three dunes in the Taklamakan Desert and fourteen dunes in the Badain Jaran Desert were sampled in the field. The modern sands were sampled from the active dune surface. The geographical locations of these sample sites are mainly situated in dune fields from the center and southern edges of the Taklamakan Desert (Fig. 1b) and the southern inland area in the Badain Jaran Desert (Fig. 1c-d).

In the Inner part of the Taklamakan and the Badain Jaran Deserts, aeolian and lacustrine (or fluvial in Taklamakan) deposits (palaeosediments) are frequently found interbedded (Fig. 2). For instance, lacustrine/fluvial sediment strata widely occur in the hinterland dune fields of the Taklamakan Desert along the N-S oil-transporting highway (Fig. 2a-b). Four sedimentary sequences with clearly stratigraphical layers were selected from hinterland areas of the two deserts in this study (Fig. 1). It is about 4 m in depth for the Arerjilin-I section from the Badain Jaran Desert (Fig. 1d). Lacustrine sediments in this section were interbedded by aeolian sand layers (Fig. 3a). For the Tazhong-XIII section in the Taklamakan Desert (Fig. 3b), it is general a lacustrine sequence interlayered with dune sediments. The Yaogan-VIII section is situated at the lower reaches of the Keriya River (Fig. 1b) close to the south margin of the Taklamakan Desert. The top of this section is buried by a 30-m active dune (Fig. 3c). The Tumiya-II section is located at the low reaches of the Tumiya palaeochannel close to the south edge of the Taklamakan Desert (Fig. 1b). This section is general a silty sand or sandy silt sequence interlayered by cemented calcium-carbonate layers (Fig. 3d). In general, lithologies of the lacustrine sediments are greatly different from those of the aeolian sediments in all sedimentary sequences from the Taklamakan Desert. These sequences were chronologically analyzed based on optically stimulated luminescence (OSL) dating and radiocarbon (^{14}C) dating methods.

The analytical methods for soluble salt geochemistry of aeolian sediment samples can be seen from Zhu and Yang (2010), and the OSL and radiocarbon dating methods can be seen from Yang et al. (2006, 2010). The physical and chemical analytical data of the soluble salt compositions of sediments are shown in On-line Supplementary Table 1 and Table 2, respectively. The resulting OSL and radiocarbon dating ages are summarized in On-line Supplementary Table 3 and Table 4, respectively. Note that the partial characteristics of aeolian salts from the Taklamakan and Badain Jaran Deserts were previously reported in our early works (Zhu and Yang, 2010; Zhu et al., 2012) with preliminary descriptions of their composition and distribution and related influencing factors, but their complete stratigraphies and chronologic results are presented here along with other new palaeoenvironmental estimations.

3. Results

Salinity values for all samples are mathematically calculated based on TDS values of the filtrates and their water-sediment mass ratios. The pH and the calculated salinity values for the dune sediments from the Taklamakan Desert range between 8.66 and 9.54 (mean 9.22) and 0.27‰ and 1.86‰ (mean 0.78‰), respectively. For the dune sediments in the Badain Jaran Desert, the pH and the salinity values lie between 8.82 and 9.42 (mean 9.12) and 0.05 and 0.42‰ (mean 0.15‰) , respectively. The range of pH values between the aeolian sediments is narrow (8.6-9.5). It indicates that the soil conditions and acid buffer capacities for these deserts are similar, generally in alkaline nature. Studies have proved that pH values in soil between 6.4-12.2 are mainly caused by bicarbonate (6.4-10.3) and carbonate (10.3-12.2) (Wetzel and Likens, 2000). It thus means that the alkalinities of aeolian salts in Chinese deserts are mainly controlled by the carbon-bearing salts, particularly bicarbonate.

For the 15 palaeo-aeolian sediment samples buried beneath modern dunes in the Taklamakan Desert with OSL ages ranging between 40-2 ka B.P. (On-line Supplementary Table 4), The salinities and pH values are slightly higher in salinity and lower in pH than those of modern dune sand samples (Fig. 4a). The modern dune samples collected from the identical months in different climatic year (such as the autumns in the arid 2006 and in the wet 2008) in the Badain Jaran Desert are quite similar to each other (Fig. 4b).

The major anions of aeolian salts are Cl^- and HCO_3^- in all samples. The SO_4^{2-} concentrations are relatively low (Fig. 4d). Na^+ is the first major cation and Ca^{2+} is the second. The concentrations of K^+ and Mg^{2+} are relatively low (Fig. 4e-f). It should be noted that the carbon-bearing ion concentrations in these sediments are highly correlated with pH (Fig. 4c). Based on charge balances and mass matches between major cations and anions, the potential salt mineralogy of these aeolian sediments can be categorized as: $NaHCO_3$ + NaCl + ($CaCO_3$) in the Badain Jaran Desert (Fig. 4g), and NaCl + $NaHCO_3$ in the Taklamakan Desert (Fig. 4h). It indicates that the assemblage of sodium carbonate and chloride is widely distributed in aeolian sediments within the Chinese sandy deserts.

The fingerprint diagrams of the ion distribution patterns for aeolian sediment solutes, local groundwaters and surface waters are shown in Fig. 5 a-b. The distribution patterns for dune surface sediments from the Badain Jaran Desert are different from the local groundwaters and the local lake waters (Fig. 5a). The chemical differences between aeolian salts and natural waters are also evident in the Taklamakan Desert (Fig. 5b), particularly in the patterns of magnesium and sulfate. These factors reflect a weaker influence of local waters on aeolian salts both in the Badain Jaran and Taklamakan Deserts.

For the Taklamakan Desert, the salt salinities of aeolian sediments have a strong positive correlation with the median grain sizes of the corresponding samples (Mz in phi unit, Fig. 5c), indicating an enrichment of soluble salts towards the finer particles of aeolian sediments. A same trend is also observed in the Badain Jaran Desert, as the salt concentrations in different gain size fractions increasing when particle fractions become finer (Fig. 5d).

In dune surface sediments from the tow deserts along the longitude (E) and latitude (N) lines of sampling sites, the salt salinities show regularly varied trends, with an increasing salinity together with the decreases of the longitude and latitude degrees, respectively(Fig. 5e-h). Because the mean annual precipitation in northern China increases gradually along the same meridional direction, and the temperature or mean solar radiation decreases along the same latitudinal gradients, this indicates that enrichment of aeolian salt contents have a clearly relationship with the changing meridional and zonal gradients of the sample locations, which correspond to the regional climatic gradient, as well as the aeolian effects of granularity differentiation caused by wind/atmospheric dynamics, but are weakly linked with local hydrological conditions.

For the Arerjilin-I section in the Badain Jaran Desert, black carbon from the highest shorelines of Arerjilin Lake (+10 m) were dated to 5628±221 cal yr BP

(CNR-185, Fig. 3a and On-line Supplementary Table 3). Calcareous gyttja (+6 m) and black carbon (+5.5 m) from deposits above the lake surface was dated to 4757±315 cal yr BP (CNR-173) and 7144±200 cal yr BP (CNR-186), respectively (Fig. 3a and On-line Supplementary Table 3). Normally, deposits are younger at locations below the highest lake level. However, in this case it is believed that younger sediments were removed by wind erosion. Consequently, the older lacustrine sediments are now exposed to the surface.

The Tazhong-XIII section contains three lacustrine layers interbedded by aeolian sand sediments (Fig. 3b). The OSL age of the bottom aeolian layer is 39,800±2900 a (On-line Supplementary Table 4). Aeolian layers underlying the top two lacustrine layers are dated to 28,000±2300 a and 29,200±2600 a. The age limits of the lower lacustrine layer range between ca 40,000 and 30,000 a ago (Fig. 3b).

Lacustrine strata in the Yaogan-VIII section are intercalated with two aeolian sand layers (Fig. 3c). The aeolian sands underlying the lacustrine section is dated to 14,500±1100 a (On-line Supplementary Table 4), and the aeolian sands overlying the lacustrine section is dated to 2320±180 a. The age constraints of aeolian sand layers indicate that the paleolake was dried between ca 14,000 and 2000 a (Fig. 3c).

In the Tumiya-II section that deposited on the palaeoterrace of the Tumiya River near the south margin of the Taklamakan Desert, The OSL chronology is dated to between 23,700±1800 a and 8700±800 a (On-line Supplementary Table 4). The accumulation of sandy loess at this section indicates that southern margin of the Taklamakan Desert at the time between 23,700±1800 and 8700±800 a was wet.

The above chronological data show that the OSL data hold great uncertainty. It may be necessary to discuss the uncertainty in different locations in this study. As previously reported, textures such as grain size and shape are important parameters influencing the OSL dating of sediment (Aitken, 1985; Fain et al., 1999; Brennan, 2003; Guerin et al., 2012; Duval et al., 2015), since they have a direct impact on several correction factors that are used for evaluating the dose rate. The aeolian sand layers for OSL dating in the Tazhong-XIII section have a mean particle sizes of 3.3-3.5 phi (sieving result), which is much coarser than the interbedded lacustrine deposits (mean particle size 5.8-6.7 phi). The aeolian sand layers in the Yaogan-VIII section have a mean grain size of 3.8 phi, also coarser than the intercalated lacustrine deposits (mean particle size 4.6-6.2 phi). Aeolian silt or sandy loess deposits in the Tumiya-II section have a mean grain size of 3.6-4.0 phi. These data show a large variability in term of size of grains between the samples for OSL dating in the Taklamakan Desert. Because there is a distinction between the sieve aperture size, or sieve opening, and the diameter of a particle, consequently, the main dimension controlling the sorting of the grains for non-spherical particles is the so-called intermediate diameter. For example, for standard mesh of 50, 100, 200 and 300 mm, grains passing through may have an intermediate dimension in the range of from 50 to 71 mm, 100-141 mm, 200-283 mm, 300-424 mm, respectively (Duval et al., 2015). Besides, the grain size distribution of the raw sediment may have also an impact on the dating results, like in the case of a bimodal distribution (Duval et al., 2015). So the principal sources of uncertainty in OSL dating data of this study may potentially be derived from the grain size of aeolian sediments. On the other hand, the difference in ^{14}C ages between the uppermost and lowermost organic carbon layers of a sediment profile usually underestimates the time of sediment deposition, and the difference in OSL ages taken from sediment units overlying and underlying a buried soil most likely overestimates it (Miao et al., 2016). For example, the comparison of OSL and ^{14}C ages suggests that the radiocarbon dating technique may significantly underestimate the age of sediments for samples older than 30 cal ka BP (corresponding to ~25 ^{14}CkaBP) (Long et al., 2015). This factor could also lead to a certain uncertainty in the chronological data of this study.

4. Discussion

4. 1. Primary or secondary salt in origin?

Firstly, it is necessary to understand whether the nature of soluble salt in aeolian sediment is primary or secondary in origin. As defined by Warren (2006), an original salt deposit (evaporite) is hydrologically driven by solar evaporation and is sourced from saturated brine on ground surface or nearsurface. In order to emphasize the highly reactive nature of soluble salts in the sedimentary realm, such salts that is precipitated from surface brine and retaining crystallographic evidence of the depositional process are regarded as primary salt. According to this definition and the related two limiting factors, almost every subsurface salt texture is secondary, because it is diagenetically altered, frequently with fabrics indicating pervasive early recrystallization. Secondary salts can be formed in surface/subsurface settings equivalent to the eogenetic, mesogenetic and telogenetic realms (Choquette and Pray, 1970). Under this definition of a primary versus secondary salt, we can say that, outside of a few Neogene examples (Riding et al., 1998; Valero-Garces et al., 1999, 2001; Pedley et al., 2003), there are few salt deposit with textures that are wholly and completely "primary". Without exception, soluble salts in aeolian sediments should be secondary salt originated in the eogenetic realm.

According to the salt category defined by Warren (2006), salt minerals can be simply subdivided into evaporitic alkaline earth carbonates and evaporite salts. In a view of depositional process, evaporitic alkaline earth carbonates are the firstly-precipitated salt minerals that sourced from concentrating hypersaline water. Compared with the evaporite salts, they are tend to be formed during the early stages of surface brine concentration-crystallization, while the evaporite salts are formed during the more saline stages of concentration-crystallization process (Smoot and Lowenstein, 1991). As analyzed above, $NaHCO_3$ + $CaCO_3$ are the identical components in dune sediments from both the Taklamakan and the Badain Jaran Deserts. It indicates that evaporitic alkaline earth carbonates are major component of these aeolian salts.

Although texture of most of the subsurface salts is secondary, the earliest secondary salts are often syndepositional precipitates (Warren, 2006), with formation of cements and replacement even as the primary matrix accumulates around them. For example, nodular anhydrite and aragonite were recognized in Permian mudflats by Kerr and Thomson (1963), they interpreted it as a subaqueous saline pan indicator. Works by Gerdes et al. (1994, 2000) have shown that carbonate grains, such as ooids and peloids, typically thought of as indicators of marine conditions and mechanical agitation, can precipitate in situ in $CaCO_3$-saturated evaporitic settings. For aeolian sediments in arid environment, we image that much of surface salts are deposited in multiple episodes of early diagenetic (syndepositional) cementation. This syndepositional salt may be formed in multiple dissolution-precipitation events in the sand particle surface and are precipitated between successive depositional episodes of salt crust formation.

Besides alkaline earth carbonates ($NaHCO_3$ + $CaCO_3$), NaCl is another major component of aeolian salts in the deserts studied. This salt, dominantly halite, is evaporite salts according to the classification by Warren (2006) that precipitated from concentrated brine after the brine has crystallized the alkaline earth carbonates. Both the evaporitic alkaline earth carbonates and evaporite salts coexist in the aeolian sediments of the deserts, indicating that aeolian salts are complex salt mixture formed by multiple evaporation and precipitation processes in the eogenetic realm.

4. 2. Possible mechanism of aeolian salt formation

All evaporite salts are ionic salts. The definition of a salt deposit (evaporite) proposed by Warren (2006), as mentioned above, has no assumption as to the origin of the parent brine. It may be marine, nonmarine or a hybrid (Smoot and Lowenstein, 1991). By definition, there is a need for aridity and for water loss to exceed inflow (Borchert and Muir, 1964). This means deposition and diagenesis in evaporites is more dependent on climate than in either siliciclastic or carbonate sediments. Therefore, modern continental evaporites typically accumulate in semi-arid to hyperarid deserts. Suites of precipitated salts follow the geochemical make-up of the parent brine (Braitsch, 1971). It means that the mineralogy and order of precipitates in any brine pool are controlled by the ionic make-up of the parent brine. This provides a clue to reveal the possible source and mechanism of salt formation under particular environment.

Generally speaking, eight mechanisms or processes precipitate most salt minerals and crusts worldwide: 1) in-situ pedogenic weathering and eluviation processes of existing salty or volcanogenic (ash) parent materials; 2) eolian or fluvial input of salt or salt-rich sediment; 3) upward movement of salt via capillary flow from a shallow salty groundwater, 4) an oceanic aerosol source, creating in salt-enriched rainfall that evaporatively concentrates within the regolith, 5) intrusion and flooding by seawater, 6) decomposition of vegetation, 7) in-situ oxidation of sulphide minerals, 8) anthropogenic pollutants (Petrov, 1976; Smoot and Lowenstein, 1991; Warren, 2006; Singer, 2007). The first two sources are direct evaporite associations, while the third is typically associated with salt lakes, sabkhas and playas in semi-arid or desert settings within 500-1000 km of the coast. Many salt soil horizons are combinations of both pedogenically precipitated and detrital wind-reworked salt (Warren, 2006; Singer, 2007). Based on the eight views, we can get clues into the possible mechanism of aeolian salt formation in Chinese deserts.

To the weathering mechanism in view (1), our previous study has shown that the degree of weathering of aeolian sand in the Taklamakan is very low (CIA $\approx$ 50) (Zhu and Yang, 2009), which is similar to that of the un-weathered Upper Continental Crust. This observation suggests that salt contributions directly from bedrock weathering are of minor significance. For the eluviations process in view (1), studies have demonstrated that in a region with a mean annual precipitation less than 500 mm, the export of erosible elements by eluviation processes in the soil will be less significant than their preservation by evaporation-crystallization processes (Birkeland, 1999; Kraimer et al., 2005). Since the mean annual precipitation in desert regions of northern China is lower than 400 mm (Fig. 1a), the degree of eluviation exerted on soluble salts in the two sandy seas is lower than that of preservation. The salt concentrations of samples buried in dune subsurface layers in this study are equal to or only slightly higher than those of the surface samples (Fig. 4a), indicating only a little downward movement of these salts is taking place. Case studies on salt movement in desert soil have also shown that only 1.64% of the rainfall-leached salt can reach 1.0 m depths, and only 0.02% of the rainfall-leached salt can penetrate 2.0 m below the ground surface (Marion et al., 2008). This means that salt contributions from eluviation processes of parent salts are also minor.

To the view (3), dunes in both the Taklamakan and Badain Jaran Deserts are generally higher than 20~50 m (Zhu et al., 1985), particularly being higher than 200 m in the Badain Jaran Desert, so it is impossible for groundwater to contribute salt to aeolian sediments by drawing upward to the dune surface via capillarity.

To the view (4) and (5), both the two sandy deserts studied are geographically far away from any ocean and have been under the control of terrestrial processes since at least the Cenozoic (Zhu et al., 1980). Salt contributions from present or ancient seawater and aerosol can, therefore, be neglected. However, contributions from oceanic-sourced rainfall precipitation (atmospheric wet deposition) will discuss below.

To the view (6), although the decomposition of vegetation may contribute to the salts of some desert areas (Petrov, 1976), but plants in/around the sampling sites of the deserts in this study are scarce, so any influence of vegetation is minimal.

To the view (7), Petrov (1975) suggested that the salt chemistry in the world deserts can be classified into four types: chloride, sulfate, carbonate and mixed. Many desert soils in arid to semi-arid environment are sulphate-rich in areas surrounding the perennial saline lakes, the brine pan depressions and their dune margins (Petrov, 1976; Smoot and Lowenstein, 1991; Warren, 2006). For example, gypsum crusts have been reported from all the continents, including Antarctic (El-Sayed, 1993). Studies have shown that chloride–sulfate salts but not bicarbonate salts are dominated in the Kalahari Desert (Wang et al., 2009). This is also the case for wide-distributed aeolian loess deposits in the Loess Plateau of northern China (Zhu et al., 2012), which are mainly composed of Na_2SO_4+$NaHCO_3$ for loess and of Na_2SO_4+$NaCl$+$NaHCO_3$ for paleosol (Sun et al., 2006). While, the analysis of this study shows that aeolian salts in the Taklamakan and Badain Jaran Deserts are mixture of sodium bicarbonate and chloride. This is different from both the Chinese loess deposits and the global tropical/subtropical deserts. Although sodium sulphate (Na_2SO_4) is an important salt existed in loess-paleosol sequences, but less common in the dune sediments from the sandy deserts in this study. So mechanism of in-situ oxidation of sulphide minerals can be excluded for the aeolian salts in this study.

To the view (8), anthropogenic pollutants, generated by human activities, are usually inorganic ions such as NH_4^+, NO_3^-, $H_2PO_4^-$ and SO_4^{2-} and some organic anions. The contents of all samples studied, however, have roughly equivalent numbers of cations and anions with low/none above-mentioned ions (On-line Supplementary Table 2), suggesting that human impact on salt concentrations across the two sandy deserts is negligible.

On the basis of all above results, only the view (2) is unexplained. Actually the mechanism in view (2) involves the hydrological and atmospheric processes of salt origin and transportation. We discuss it below.

4. 3. Atmospheric origin?

There are wide similarities but partial differences existing in the salt chemistry and ion distribution patterns of aeolian salts between the Taklamakan and Badain Jaran deserts (Fig. 4g-h). Besides, they are weakly correlated with the chemical compositions of the nearby ground waters under local hydrological setting (Zhu and Yang, 2010) (Fig. 5a-b), but are strongly associated with the regional climatic and wind regime (Zhu et al., 2012) (Fig. 5c-h). All these evidences indicate that the natural system of aeolian salts is hydrologically open and the chemistry of the parent brines and the associated salt deposits may be significantly different from that predicted for hydrologically closed systems. It means that the sources of aeolian salts in the deserts are strongly controlled by earth surface processes at a regional or large scale, but not at a local scale. Seen from this point, only an atmospheric origin but not hydrological origin can be responsible for this pattern.

It has been recognized that atmospheric chemical species is a significant source contributing acidic and eutrophic elements to both the terrestrial and ocean ecosystems (e.g., Sehmel, 1980; Wesely and Hicks, 2000; Inomata et al., 2009). For example, soil salts in many desert areas have been implicated to be originated from the atmospheric sources (Amit and Yaalon, 1996; Bohlke et al., 1997; Oyarzun and Oyarzun, 2007). Generally, there are two forms for atmospheric deposition, wet and dry deposition. Wet deposits contain salts derived from rainwater and snow. Studies have shown that chloride and carbonate are major species in rainfall in central Asia and north China (Fujita et al., 2000; Li et al., 2007; Zhao et al., 2008). It indicates that the chemical compositions between wet depositions and aeolian salts are similar.

Besides rainfall, aerosol and dust, known as atmospheric dry deposition, is another global phenomenon of salt transportation (Goudie and Middleton, 2006). At present there are few data being available for understanding the salt compositions of dust in the study areas. However, wide studies have proved that salt transportation and redeposition via dust are one of the important patterns for biogeochemical cycles in arid areas (Logan, 1974; Yaalon and Ginzboung, 1966; Reheis and Kihl, 1995; Al-Momani et al., 1995; Kiefert, 1997; Blank et al., 1999; Kulshrestha et al., 2003; Zheng et al., 2005; Abuduwaili et al., 2009). It should be a similar case for the hinterlands of northwestern China.

4. 4. Climatic implications

The coexistences of palaeo-lacustrine sediments (playa) and aeolian sediments are commonly seen in desert regions globally. These are clear evidences for past environmental changes in desert conditions (Smith, 1971; Yechieli and Wood, 2002; Alonso-Zarza, 2003; Yang et al., 2006; Tripaldi et al., 2011; Dong et al., 2012), Therefore, the large number of playa deposits in the inter-dune depressions of these deserts offers an exceptional opportunity to study regional paleohydrological and paleoclimatic change. Over the past decade, there have been significant advances in our understanding of the late Quaternary history of the desert regions of northwestern China. OSL and ^{14}C chronologies of aeolian sediments from various locations in these deserts provide important archives to understand the timing of lacustrine/fluvial and aeolian processes related to environmental and climate changes. However, despite this increased interest, paleolimnological and palaeoaeolian research is limited by a paucity of potential geoproxy and the study sites with relatively long, uninterrupted stratigraphic sequences. Sequential variations of inorganic salts have been tentatively applied to reconstruct the palaeoenvironmental history in desert areas. For instance, taking the soluble salts variations in thick Cenozoic deposits as one of the indicators of paleoenvironmental change, Sun et al. (2008) reconstructed the aridity history of the Tarim Basin and argued that hyperarid climate had been prevailed within the basin since 5.3 Ma ago.

The variations in soluble salts content for the four sections collected from the Taklamakan and Badain Jaran deserts are examined in this study, as shown in Figs. 6-9. The OSL and ^{14}C age constraints of these sequences are also shown in these figures. The data indicates that all of these playa deposits had experienced fluctuations in high moisture conditions during the period from late Pleistocene to mid-Holocene.

For the Arerjilin-1 section, there are an increase-decline pattern in salinity variation and a decline-increase-decline pattern in pH variation during the early-middle Holocene (Fig. 6). For the Tazhong-XIII section, there is a clear serrate shape (Fig. 7) in the pattern of salinity variation. It indicates that there are at least three times of humid environmental fluctuations during the past 40 to 28 ka B.P.. In the Yaogan-VIII section, the sequence salinities are relatively high (Fig. 8). It suggests that a saline lacustrine environment has occurred this area and the local hydrological settings have been retained the saline conditions for a long time from the end of the last glacial to late Holocene. In the Tumiya-II section, variations of the sequence salinities were unstable during the LGM (24 ka B.P.). There are two stages of salinity increase during the end of the LGM (about 10-11 ka) and the early Holocene (after 8.7 ka B.P.), respectively (Fig. 9). . Taken the four sections as a whole into consideration, there are evident differences between the aeolian sediments and fluvial sediments in salt content. In general, salinities in aeolian sediments are evidently lower than those of lacustrine sediments. It suggests that a great salt depositional discrepancy exists between the aeolian and the lacustrine sedimentary systems, which could be owing to the differential mechanisms of salt formation and origin in wind and water regimes. The results suggest that the inorganic salt is a latent proxy indicator for single depositional-environment sequence in revealing local hydrological variations and climate changes in desert areas during the late Quaternary.

However, the interpretation based on salt archives should be careful for the sedimentary sequences with dual/multiple depositional end-members. Researchers have reported that in a sediment strata formed by an identical geological process like lacustrine or aeolian dynamics, the increase of sequence salinity commonly reflects a stepped-up environmental aridity (Sinha and Taymahashay, 2004; Warren, 2006; Liu et al., 2008). Because whatever under the aeolian and lacustrine depositional settings, the increase in salt content is a reflection of the enhanced evaporation potential. However, in a sediment profile controlled by multiple geological processes like combined aeolian and lacustrine dymanics, the increase in salt content could be not a reflection of the enhanced aridity, just like the case in this study. It is generally higher in salinity for the lacustrine sediments than for the aeolian sediments. In this study, the salinity increases in sections of the Arerjilin-I, Tazhong-XIII and Yaogan-VIII are general a representation of the enhanced wetter conditions (Figs. 6-8), because the higher salinities are always located in the lacustrine/fluvial layers. In general, lacustrine/fluvial strata in aeolian sequences always reflect enhanced humid periods than that of the aeolian period (Yang et al. 2006). However, this is not the similar conditions for the Tumiya-II section, as the salt increases in this sequence should be attributed to enhance aridity (Fig. 9), because the sedimentary structure of this section is uniformly dominated by aeolian process. Thus it can be confirmed that once the coexistence of aeolian and fluvial/lacustrine sediments occurs in a sedimentary structure in desert regions, the single use of salt archives should be cautious in reconstructing the regional environmental changes. A combined use of salt archives and sedimentary proxies is recommended under this situation.

Besides, land surface processes such as chemical, physical and mechanical processes can weld younger to older sediment profiles and affect the accumulation, dissolution, and reprecipitation of mineral materials in sediments, as illustrated by Olson and Nettleton (1998). Sediment properties most affected include texture and the content and distribution of soluble salts. Processes such as erosion and deposition can truncate profiles or bury them either rapidly or extremely slowly. Effects of these and other processes on sediment properties must be examined with care in paleosols that have been buried even for short interval. The above-mentioned analogies of fluvial/lacustrine-aeolian profiles in the Taklamakan and Badain Jaran deserts should be evaluated carefully in future studies and possible avoided as single indicators in paleoenvironmental reconstruction in the desert environment.

5. Conclusion

Formations of inorganic salt in aeolian sediments in the desert environment are significant to understanding the interrelationships between aoelian dynamics and regional hydrological and climate regimes. Geological information related to the inorganic salt composition and distribution and their environmental significance in aeolian sediments in the middle-latitudes deserts are still rare. Geochemistry of soluble salts in modern and ancient dune sediments from the sandy deserts in northwestern China was surveyed in this work. Variations of the salt composition around these sandy deserts are closely related to regional climatic parameters, such as the precipitation, temperature and wind agent. The aeolian salts are proposed to be sourced from the atmospheric depositions that are strongly associated with the local-scale and regional-scale dry and wet depositions, such as rainfall and dustfall precipitation. Single salt archive in sedimentary deposits interlayered by aeolian and lacustrine facies should be carefully used as geoproxy to reconstruct palaeoenvironmental histories in arid conditions.

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Figure Captions

Figure 1 Locations of the study areas and sampling sites in this study. (a) The distributions of deserts and the mean annual precipitation isohyets in China, (b) the Taklamakan Desert, (c) and (d) the Badain Jaran Desert.

Figure 2 Photograph views of Quaternary and modern sediments of aeolian and lacustrine/fluvial facies that consisted of clay and sand/silt sand alternations in the Taklamakan and Badain Jaran Deserts. (a) and (b) the Taklamakan Desert, (c) and (d) the Badain Jaran Desert.

Figure 3 Sketch maps of the sedimentary profiles studied in this study. (a) Lacustrine sediments interbedded by aeolian sand layers with ^{14}C ages at the Arerjilin-I section in the Badain Jaran Desert, (b) interbedding of aeolian and lacustrine deposits and their OSL ages at the Tazhong-XIII section in the Taklamakan Desert, (c) lacustrine deposits buried under a 30 m high dune at the Yaogan-VIII section in southern Taklamakan, and (d) sandy loess deposits with OSL dating results at the Tumiya-II section on southern margin of the Taklamakan Desert.

Figure 4 Salinities *vs.* pH of sediment solutions, (a) dune-surface sediments (modern sediment) and buried-dune sediments (palaeo-sediment) in the Taklamakan Desert, (b) dune-surface sand samples collected in different climatic seasons (dry and wet) in the Badain Jaran Desert. (c) Carbon-bearing ions *vs.* pH. Triangular plots of major anions (d) and cations (e and f) of the soluble salts in mEq/l unit. Distribution patterns of the major ions of the soluble salts in the aeolian sediments from (g) the Badain Jaran Desert and from (h) the Taklamakan Desert.

Figure 5 Distribution patterns of the major ions of aeolian salts, groundwaters and lake waters in (a) the Badain Jaran Desert and (b) the Taklamakan Desert. The data of local lake water and groundwater bodies in the Badain Jaran are cited from Yang and Williams (2003), and the river water and groundwater bodies in the Taklamakan from Zhu and Yang (2007). The relationship between salinities and grain size compositions of the aoelian sediments: (c) salinity *vs.* median particle diameter (Mz, in phi unit) of dune-surface sediment from the Taklamakan Desert, and (d) salinity *vs.* grain size compositions of randomly-selected dune sediments from the Badain Jaran Desert. Spatial distributions of the salinities of the aeolian sediments along the longitudinal and latitudinal degrees in the Taklamakan Desert (e and g) and in the Badain Jaran Desert (f and h), respectively.

Figure 6 Sequential variations in soluble salts contents (salinity and pH) of the Arerjilin-I section in the Badain Jaran Desert.

Figure 7 Sequential variations in soluble salts contents (salinity and pH) of the Tazhong-XIII section in the Taklamakan Desert.

Figure 8 Sequential variations in soluble salts contents (salinity and pH) of the Yaogan-VIII section in the Taklamakan Desert.

Figure 9 Sequential variations in soluble salts contents (salinity and pH) of the Tumiya-II section in the Taklamakan Desert.

On-line Supplementary Table Captions

On-line Supplementary Table 1 Physical parameters of the soil-water extraction of the modern and ancient aeolian sediments from the Taklamakan and Badain Jaran deserts in northwestern China.

On-line Supplementary Table 2 The analytical data of major ionic compositions for the soil-water extraction of the aeolian sediments from the Taklamakan and Badain Jaran deserts

On-line Supplementary Table 3 Conventional radiocarbon ages for section samples from the Badain Jaran Desert (cited from Yang et al., 2010)

On-line Supplementary Table 4 Summary data for OSL ages of section samples from the Taklamakan Deserts (cited from Yang et al., 2006).