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A new hydrological climatic proxy in arid lake sediments: Iodine-uranium concentrations

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ABSTRACT

Hydrological variation in the past ~2 kyr is important for understanding the impact and driving forces of climate changes in the present and future, especially in arid areas. Sediment is one of the most useful archives in such a time period, and selection and validation of qualified hydrological proxies is the key point for reconstruction of hydrology in the past centuries. However, such research still faces a challenge due to limited knowledge of physiochemical implications of the traditional proxies in the specific lake, and difficulties to discriminate dry-wet from warm-cold signals. Here, a sediment core collected from Keluke Lake, Qaidam Basin, Northwest China was analyzed for iodine and uranium concentrations, to explore their potential as a new hydrological proxy in the arid regions for reconstructing the dry-wet changes during the past ~2200 years. A significant correlation between iodine and uranium concentrations ($r = 0.62$, $P < 0.01$) was observed in the sediment core, and four peak events of synchronously varied iodine and uranium concentrations occurred in the studied sediment core. This indicates similar physiochemical behaviors of the two elements from the catchment to the sediments via lake water. During drier periods, as vegetation degradation and soil organic substance content decreased, water-soluble iodine and uranium were intensively leached, and fine particle associated iodine and uranium were seriously eroded from catchment soils. Meanwhile, the suspended matter concentrations and organic substance content in lake water increased as well, which facilitated more iodine and uranium to be deposited and recorded in the sediment as peak events. The four dry events coincide with the reported climatic changes in other lake sediments, ice core and aeolian sediments, and also correspond with the society evolution in this area, including the establishment of Tuyuhun Kingdom (313–663 CE), Tubo Empire (618–842 CE), Mongolian tribes (1495–1591 CE, and the Han population immigration driven by a severe drought disaster in 1876–1879.

1. Introduction

The climate record in the past ~2 kyr is of the highest significance for understanding the impact and driving forces of climate changes in the present and future, especially in arid areas (Li and Chang, 2014). The present instrumental climate records (since the ~19th century) and paleoclimate records have shown that temperature changes often happen on a large spatial scale, such as the increasing warming period during the 20th century in the whole Northern Hemisphere (IPCC, 2021), the Medieval Climate Anomaly from the 10th century to 13th century and the Little Ice Age from the 15th century to the 19th century that widely recorded in the whole Eurasia (Hao et al., 2018). While the hydrological variations are of great disparity at regional scales, this has significant impacts on the regional environmental change and the

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human society evolutions (Dong et al., 2021), especially in the arid areas.

Due to convenient sampling and long-term record compared to ice core and tree rings, lake sediment is one of the most common used media for extracting information of climatic changes in the past ~2 kyr (Leng and Marshall, 2004). Chemical and physical composition of lake water is heavily influenced by labile lithologies in the drainage basin through leaching and erosion to the lake, this process would be regulated by the regional climate during a long-term evolution (Yang et al., 2015). The input substances from the drainage basin were transported and preserved in the sediment, which might reserve the dry-wet signal, and could be used as dry-wet proxies in the arid areas. Physical parameters such as grain size distribution of sediments varies in sediment cores that collected from different places of the same lake due to different distances to the lake shore. Chemical parameters are usually highly comparable in the whole lake (You et al., 2015), and therefore has high potential in dry-wet climate reconstruction. However, the related physiochemical processes of chemical proxies are not well understood sometimes, this hampers the reconstruction of past dry-wet changes in a specific arid region. The Qaidam Basin with the Qaidam desert (area of 34,900 km^2) is dominated by a continental climate, and is deemed as a typical arid area in northwest China, and also one of the most important dust source areas in East Asia. The dry-wet changes during the past ~2 kyr directly influences the global dust cycling (Niu et al., 2010). However, contradictory information on hydrological change in this region was often obtained due to lack of deep understanding of physiochemical behaviors of the proxies. For example, wet periods indicated by high carbonate in lake sediment disagree with low precipitation intervals recorded by tree-ring width in surrounding mountains (Zhao et al., 2010; Shao et al., 2006); the alkenone-based lake water salinity record is in contrast with that of redness record from the same lake (presented for dry/wet changes) (Liu et al., 2006; Ji et al., 2005; Zhao et al., 2009). In addition, it is also difficult to discriminate specific dry-wet from warm-cold signals (Niu et al., 2010; Wang et al., 2008).

As a biophilic, high-water soluble, and redox-sensitive element, iodine could be a potential proxy in paleoclimatology research. Based on the oceanic origin of iodine in terrestrial environment and dominant wet deposition way from atmosphere, distributions of the in-situ deposited iodine in surface soil and loess samples have been used to reconstruct the path ways of oceanic monsoon and changes of paleo precipitation (Yi et al., 2019; Fan et al., 2021b). While iodine in lake systems mainly originates from runoffs which transport the eroded/leached iodine from soil in the drainage basin because of its larger area compared to surface area of the lake (Hou et al., 2009). As a particulate active and redox-sensitive element, uranium has been widely used as a proxy for paleo-environment changes in marine system (Piper and Isaacs, 1995). Uranium accumulates and deposits in the sediment differently in freshwater compared to that in marine systems, where most uranium is deposited to the marine sediments by diffusion across the sediment-water interface (Chappaz et al., 2010). While a considerable portion of uranium in lake sediments was exogenously originated from weathering and erosion of drainage basin soil and transported by runoffs (Yang et al., 2015; Chappaz et al., 2010). Previous investigations mainly focused on accumulation and migration behaviors of uranium in lake sediment (Lefebvre et al., 2021; Chappaz et al., 2010), and only a few works on application of uranium as a paleo-redox proxy (Yang et al., 2015; Lefebvre et al., 2021; Lawrence et al., 2016). Since erosion/leaching process is highly regulated by the regional dry-wet changes in arid area, both iodine and uranium in lake sediment, as erosion/leaching-sourced elements, have great potential to record the wet-dry changes in the catchment area. Additionally, as suspended matter (including detrital minerals and Fe/Mn oxides and hydroxides), organic matter contents, E_h, pH, temperature and chemical composition of lake water changing with regional climate, removal of uranium and iodine from waterbody to sediment in the lake and the consequent concentrations of uranium and iodine in the sediment are regulated by climate change. Although both iodine and uranium were reported to easily associate with suspended matter and organic matter, the chemical behaviors of the two elements are different, including the response to the change of redox condition and pH of lake water and complexation with carbonate (Andersen et al., 2014; Zhao et al., 2021; Gilfedder et al., 2010; Chappaz et al., 2010; Luo et al., 2013; Qiao et al., 2012). Therefore, analysis of iodine and uranium as pair elements could better clarify their specific physiochemical processes happened in lake water-sediment system and responses to regional dry-wet changes, and avoid misinterpretation of impact of the climate change.

This work aims to explore the potential of iodine-uranium recorded in lake sediment as hydrological proxies in arid regions by investigating the variation of iodine and uranium concentrations in Keluke lake sediment core collected from the Qaidam Basin.

2. Materials and methods

2.1. Study area

A sediment core collected in the Keluke Lake in the northeastern Qaidam Basin (Fig. 1) was investigated in this work. The Qaidam Basin with Quaternary lacustrine and alluvial deposits is surrounded by the Kunlun Mountains to the south, the Altun Mountains to the west and the Qilian Mountains to the northeast (Zhao et al., 2010). The average annual air temperature in the last ~30 years is ~4 °C, and the annual precipitation is ~200 mm, while the potential evapotranspiration is ~2000 mm in this region from Dulan meteorological station where is ~150 km away from the lake.

Keluke Lake with an area of 56.7 km^2, a maximum depth of 9.6 m, a total dissolved solid of 0.98 g/L, and an oxic hypolimnion, is fed by freshwater from Bayin River (a mean discharge of 1.9×10^8 m^3/a), Balegen River (a mean discharge of 0.1×10^8 m^3/a), and groundwater from the mountains, which were all originated from the meltwater of the glacier terminus (Zhao et al., 2009).

2.2. Materials and core chronology

The sediment core named as KLK (37.33° N, 96.87° E, ca. 2817 m), was sampled from Keluke Lake (with an average water depth of 5.7 m) in July 2009 (Fig. S1). A plastic tube sampler (6 cm in diameter) fitted with a piston was used for sampling. The total length of the sediment core was measured to be 55 cm, and the core was sliced into contiguous 1-cm intervals for the whole profile immediately after the collection, then the samples were stored in plastic bags. Five surface soil samples (0–5 cm) were also collected in the nearby drainage basin as supplementary material (Table S1). After transported to the laboratory, the sliced sediment samples were freeze-dried, ground and sieved through a 200-mesh sieve. The dried samples were weighed and sealed into a plastic counting container, which were stored for 3 weeks until the radioactive equilibrium established (^{226}Ra-^{222}Rn-^{218}Po-^{214}Pb-^{241}Bi-^{214}Po-^{210}Pb). ^{210}Pb (22.23 yr) was measured using its gamma ray at 46.5 keV, and ^{226}Ra through its decay daughter ^{214}Pb with ϒ-rays of 295 and 352 keV by a gamma spectrometer (Canberra Be3830, consists of a HPGe detector, multichannel analyzer and Gennie, 2000 software) with 35% relative counting efficiency and 1.8 keV energy resolution (at 1332 keV). The excessive ^{210}Pb ($^{210}Pb_{ex}$) was obtained by subtracting the activity of supported ^{210}Pb (i.e., ^{210}Pb decay from and equilibrium with ^{226}Ra) from the total activity of ^{210}Pb. The efficiency calibration of the detector system was conducted using both the Laboratory Sourceless Calibration Software (LabSOCS) and sediment standard prepared by diluting (GBW04127 standard was mixed with natural marine sediment).

The $^{210}Pb_{ex}$ activities show an exponentially decrease trend from the top to 16 cm depth of the sediment, and became under detection limit in the segment below. Based on the CFCS model (Constant Flux and Constant Sedimentation), the average accumulation rate was estimated to be 0.069 cm/yr in the top 16 cm of KLK core. Although the ^{137}Cs activity

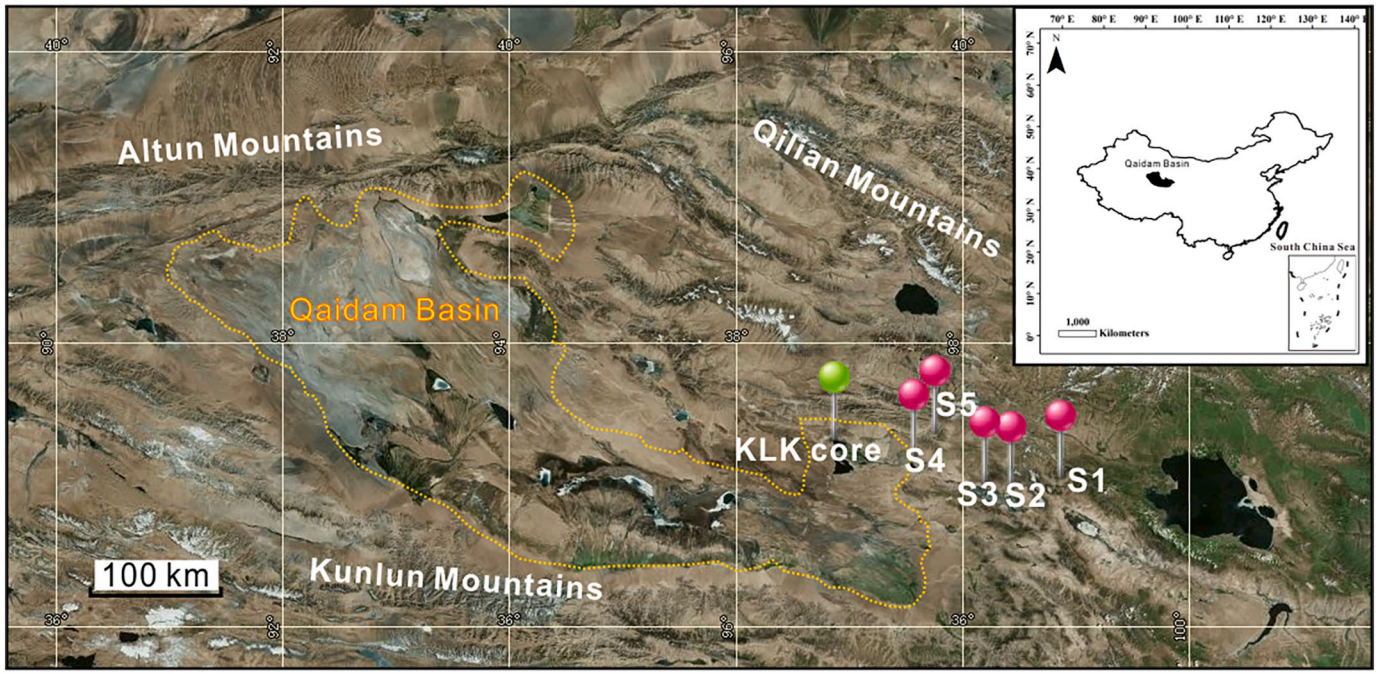

Fig. 1. Map of Qaidam Basin (yellow dotted line) with the location of KLK core (green sphere), five surface soil samples (red sphere), and surrounding mountains, including Altun Mountains, Kunlun Mountains, and Qilian Mountains. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

was measured only in the top 4 samples (0–4 cm) without characteristic unimodal, the first appearance time of ^{137}Cs occurred in 4 cm (^{210}Pb$_{ex}$ age of 1958), which corresponds with the first high yield nuclear weapons test conducted in 1951 and deposition in 1952 with the consideration of dating uncertainties (UNSCEAR, 2000). The non-unimodal might be due to the sheet melting of glacial water that supplies to Keluke Lake, which could pour ^{137}Cs that deposited on the glacier in different years into the lake at the same time. Similar observations have been reported before (Zhang et al., 2012).

The total 8 sediment samples at the depth of 4 cm, 12 cm, 14 cm, 15 cm, 22 cm, 31 cm, 41 cm, and 47 cm were analyzed for ^{14}C, and the raw data is listed in Table S3. TOC (Total organic carbon) samples were separated by treatment of sediment sample with 1 M HCl followed by rinsed with deionized water until neutral to remove carbonate. After dried at 60 °C, the sample with remained TOC (~200 mg) was placed in a quartz combustion tube containing CuO, and the combustion system was evacuated using a high vacuum system. The TOC in the sample was then converted to CO_2 by combustion at 800 °C, all CO_2 was then collected and converted to graphite with an Fe catalyst (Slota et al., 1987; Jull, 2007), ^{14}C in the prepared graphite was measured using the accelerator mass spectrometry (AMS) at the Xi'an Accelerator Mass Spectrometry Center for dating of the sediment (Zhou et al., 2006). It was observed that the dating results of ^{14}C and ^{210}Pb$_{ex}$ are different at the same depth in the studied sediment core (Table S2, S3).

Based on the almost constant difference between the paired ^{14}C and ^{210}Pb$_{ex}$ dates at the depth of 4 cm (2458), 12 cm (2540), 14 cm (2733), and 15 cm (2793), the "old carbon" effect was calculated to be 2631 ^{14}C yr based on the average of these four values, which was used to correct the ^{14}C dates in deeper part of this sediment core. A similar value of 2758 yr for "old carbon" was also reported in this lake before (Zhao et al., 2010). Uncertainties of the date were estimated using two standard deviations for the scattering of individual dating points around the age model curve. The age sequence of the sediment core was established with the BACON programs (Table S4) (Blaauw and Christen, 2011).The

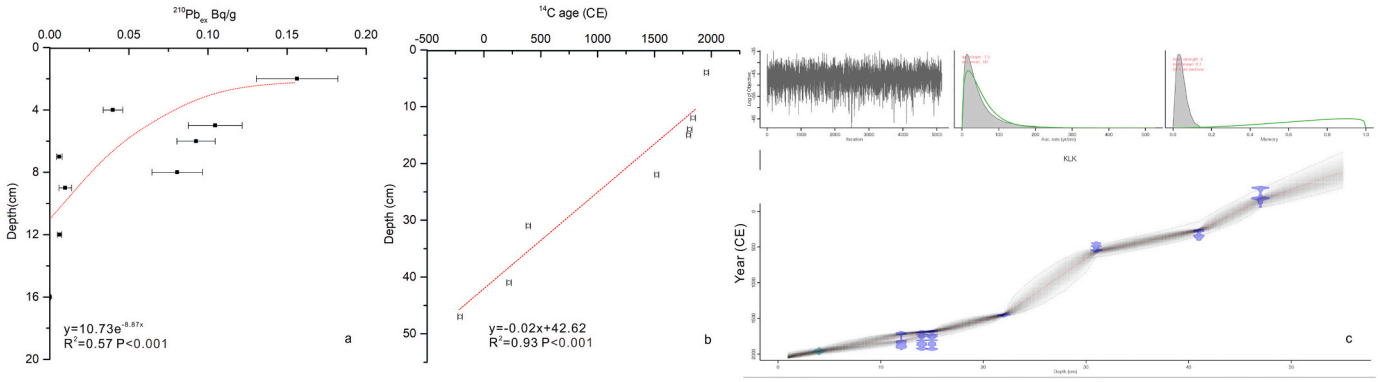

Fig. 2. Exponential decreased of ^{210}Pb$_{ex}$ with depth in KLK core (a); ^{14}C dating results with an "old carbon" correction of 2631 yr (b); Bayesian age-depth model that calculated using BACON program. Transparent blue dots show the calibrated ^{14}C dates, and the darker grey indicate more likely calendar ages (grey stippled lines show 95% confidence intervals; red curve shows single 'best' model based on the weighted mean age for each depth). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

chronology of whole KLK core was established based on the $^{210}Pb_{ex}$ (0–16 cm) and ^{14}C age (17–55 cm) over the last ~2200 yr (Fig. 2, Table S2, S3, S4). The ages at each depth (1 cm intervals) are shown in Table S2 and S4, different accumulation rates occurred in the depth segments of 0–16 cm, 16–22 cm, 22-31 cm, 31–41 cm, and 41-47 cm.

2.3. Analytical method for iodine and uranium concentrations in the sediment

All 55 sediment samples were analyzed for iodine, and only 44 samples with sufficient amount of material were analyzed for uranium. Five surface soil samples were analyzed only for uranium but not for iodine due to the sufficient published iodine data in surface soil from this area (Zhang, 2018).

An analytical method based on oxidation combustion for separation of iodine from solid samples and ICP-MS for measurement of iodine was developed in our laboratory and used in this work for determination of iodine in sediment samples. The detailed method has been published elsewhere (Hou and Zhang, 2018), it is briefly described here. During the combustion procedure, an aliquot of 0.9–3.6 g samples was weighted to a quartz boat,^{125}I solution (about 600 Bq) was spiked as tracer for monitoring the chemical yield of iodine, the sample was combusted at 800 °C for 3 h in a tube furnace (Raddec, Ltd., Southampton, UK) with oxygen flowing. Iodine in different species released from the sample during combustion was trapped in 30 mL of 0.5 M NaOH-0.02 M $NaHSO_3$ solution. An aliquot of 3.0 mL of trap solution was taken to a tube to measure ^{125}I using a NaI detector, the chemical yields of iodine during combustion were estimated by the measured ^{125}I in the trap solution and the total spiked ^{125}I to the sample to be 82–94%. An aliquot of 1.0 mL of the trap solution was taken and diluted 20 times using 1% NH_3, iodine in the diluted solution was measured using ICP-MS (Agilent 8800 ICP-MS) at the Xi'an AMS Center, China. Cs^+ (CsCl) was spiked to samples to a concentration of 2 ng/mL in CsCl as internal standard for calibration and normalization of the measurement data of ICP-MS. Procedure blanks were also prepared with the samples and measured for iodine. The detection limit was estimated using the procedure blank to be 0.012 ng/mL, which is more than two orders of magnitude lower than the level of iodine in the diluted sample solutions. The concentration of iodine in the sediment was corrected in consideration of the chemical yield of iodine measured using ^{125}I by comparison of the ^{125}I remained in the trap solution and the amount spiked to the sample before separation. The loss on ignition (LOI, %) was calculated based on the mass differences before and after combustion to roughly estimate the organic matter content in the sediment.

For determination of uranium in sediment and soil samples, an aliquot of 0.5 g samples was weighted into a beaker and ashed in a muffle furnace at 450 °C for 12 h to decompose of organic matter, which will not loss any uranium and form the refractory uranium species. The ashed sample was transferred into a Teflon crucible, and ^{233}U was spiked as yield tracer. The prepared samples were dissolved using 10 mL HF mixed with 10mL HNO_3 under heating on a hotplate under reflux with watch glass covered on the beaker until completely dissolution. The solution was then evaporated to dryness, and the residue was dissolved with 10mL of 3 M HNO_3. The prepared solution was loaded to a 2 mL UTEVA column. The column was rinsed with 3 M HNO_3. Uranium on the column was eluted with 30 mL 0.01 M HNO_3, the eluate was evaporated to dryness on a hotplate at 200 °C. The residue was dissolved in 5 mL of 0.5 M HNO_3. The intensity of ^{233}U and ^{238}U signals was measured using ICP-MS/MS (Agilent 8800 ICP-MS) at the Xi'an AMS Center, China. The total amount of ^{238}U in the sample was calculated based on the intensities of ^{238}U and ^{233}U and the total ^{233}U spiked to the sample. The concentration of uranium in the sample was calculated from the measured ^{238}U amount, the sample weight and the natural abundance of ^{238}U (99.27%). The chemical yields of uranium can be also estimated by the ^{233}U in the final separated sample and the ^{233}U spiked to the sample before separation, which was >80% for most samples. The detection limit of uranium concentration was estimated to be 3.0 pg/g, which are >3 orders of magnitude lower than that in the prepared samples.

3. Results

3.1. Levels and variations of iodine in the KLK sediment core

The measured iodine concentrations in the KLK sediment core vary in a range of 3.4–69.7 μg/g with a mean value of 21.4 ± 1.4 μg/g (Fig. 3a; Table S5). An obvious variation of iodine concentrations was observed in the whole KLK core (542 BCE to 2009 CE). The relatively lower and constant iodine concentrations was observed in 34–55 cm depth (mean value of 10.6 μg/g) compared to that in the depth of 0–33 cm (28.6 μg/g). Apparently, five peak values of iodine concentration identified as >1.5 times of the mean value were observed, i.e. 32.2 μg/g at the depth of 31–32 cm, 30.5–37.7 μg/g at the depth of 26–29 cm, 32.6–69.7 μg/g at the depth of 19–22 cm, 33.2–49.0 μg/g at the depth of 7–9 cm, and 34.2 μg/g at the depth of 0–1 cm (Fig. 3a).

3.2. Levels and variations of uranium in the KLK core

The uranium concentrations vary from 4.4 to 11.7 μg/g with a mean value of 7.4 ± 0.2 μg/g (Table S5). The uranium concentrations vary along the sediment profile, and a relatively high correlation between uranium and iodine concentrations ($R = 0.62$, $P < 0.001$) was observed (Fig. S2a). Accordingly, four peak values of uranium concentration similar with that of iodine appear in the core, i.e. 8.1 μg/g at the depth of 32–33 cm, 8.1–8.3 μg/g at the depth of 26–29 cm, 8.5–11.7 μg/g at the depth of 17–22 cm and 9.1 μg/g at the depth of 7–8 cm. However, two obvious inconsistences between two elements were also observed, i.e. (1) an iodine peak at the depth of 0–1 cm, but uranium at mean-value level in this layer; (2) uranium concentration increasing to a high level at the depth of 3–5 cm, but the corresponding iodine concentrations just at mean-value level (Fig. 3 b).

4. Discussion

4.1. Source, transfer processes of iodine and uranium in the KLK sediment and the key controlling factors

4.1.1. Sources of iodine and uranium in the sediment

The measured iodine concentrations (3.4–69.7 μg/g, mean: 21.4 ± 1.4 μg/g) (Fig. 3a; Table S5) are comparable with that reported in other terrestrial sediments, such as that in the Mississippi River Delta (5.7–34.3 μg/g) (Oktay et al., 2000), Taal Lake (2.97–54.7 μg/g) (Zhang et al., 2018), and Jiulong River (19.3–35.7 μg/g) (Wang, 2014). In generally, iodine in lake sediment mainly originates from the “allogenic” source of atmospheric deposition and “authigenic” source from drainage basin soils through the runoff (Gilfedder et al., 2010; Fan et al., 2021a). However, the measured level is significantly higher than that measured in atmospheric total suspended particles (TSP) from Xi'an with a distance of 1100 km (mean: ~10 μg/g) (Zhang et al., 2020) and in surrounding surface soil samples (2.5–13.5 μg/g, mean: 6.4 μg/g) (Zhang, 2018), indicating an accumulation of iodine in the studied lake sediment.

The measured uranium concentrations (4.4–11.7 μg/g, mean: 7.4 ± 0.2 μg/g) is comparable with that reported in other terrestrial sediments, such as that collected in northwest China (0.6–83.0 μg/g with most data falling in the range of 2–10 μg/g) (Han et al., 2013), the Qinghai-Tibet Plateau (mean: 3.18 μg/g) (Hao et al., 2014), the Black Sea (~3–18 μg/g) and Cariaco Basin (~3–14 μg/g) (Andersen et al., 2014). As a weathering product, uranium in lake sediment was mainly leached from the soil and pedogenic rocks in the drainage basin by the runoff. However, the level is significantly higher than that measured in surrounding surface soil samples (1.8–2.7 μg/g, mean: 2.3 μg/g) (Table S1). This indicates that the studied lake sediment might accumulate uranium from

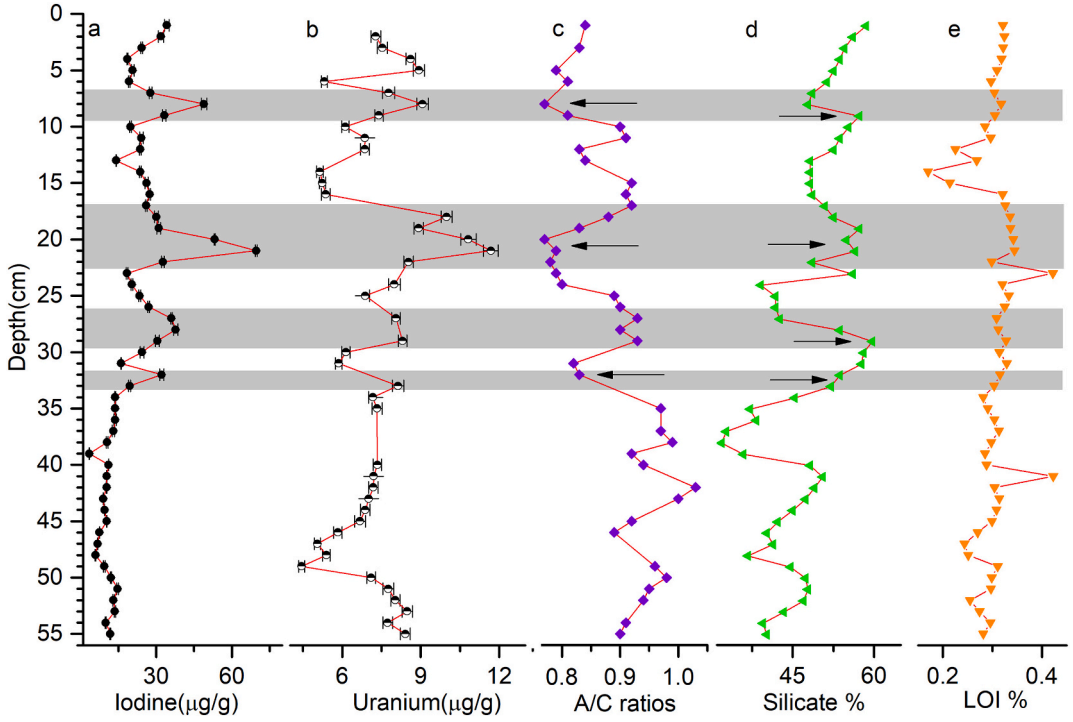

Fig. 3. Variations of iodine (a) and uranium concentrations (b) LOI values (e) with depth in the KLK core, and comparison with the reported *Artemisia/Chenopodiaceae* (A/C) ratios (c) and silicate percentage (d) at the same depth from this lake (Zhao et al., 2009). Four similar peaks of iodine and uranium concentrations in grey stripes.

surrounding environment. Due to the smaller area of Keluke Lake (56.7 km^2) compared to that of the drainage basin (7462 km^2), and the large annual runoff amount from Bayin River (1.9×10^8 m^3/a) and Balegen River (0.1×10^8 m^3/a) to the lake, the input of iodine and uranium from drainage basin soil should be the most important contributors in the studied sediment. A serial physiochemical process that happened during their transfer from lake water to sediment could help enrichment of iodine and uranium in sediment.

4.1.2. Parameters influencing the transfer of iodine and uranium from lake water to sediment

The significant correlation between iodine and uranium concentrations ($r = 0.62$, $P < 0.001$) and four similar peak events of iodine and uranium concentrations in the KLK core indicate the similar controlling factors on the transfer processes of these two elements from catchment to lake water and then sediments.

A positive, but not highly correlations of iodine concentrations with LOI ($r = 0.25$, $P < 0.10$) and a significantly positive correlation of uranium concentrations with LOI in the sediment ($r = 0.45$, $P < 0.01$) were observed in the investigated sediment core. Positive correlation between iodine and organic matter content has been observed in other sediment cores (Oktay et al., 2000; Luo et al., 2013; Takata et al., 2013). Meanwhile, high proportions (~60–70%) of iodine in sediment was observed to associate with organic matter (mainly on phenolic and thiol groups in the humic substances) (Hou et al., 2003; Qiao et al., 2012). Uranium (VI) could form strong complexes with organic matter, and dissolved uranium concentrations decreased after the addition and deposition of organic matter (Martine et al., 1999). A portion of ~10–20% uranium was observed to associate on organic matter in sediment (Qiao et al., 2012). All these findings confirmed that the organic matter (indicated by LOI) is one of the important factors in the transfer of iodine and uranium from upper water into the sediments. This might be interpreted by micro-algae and bacteria intermediated scavenging process of iodine and uranium from upper water to the bottom sediments (Martine et al., 1999). The *E*h, pH, temperature, and content of suspended matter and carbonate, etc. in lake water might also influence the deposition and accumulation of iodine and uranium in the sediment through dissolution-precipitation courses, oxidation-reduction processes, adsorption-desorption, etc., leading to the synchronous variations of iodine and uranium concentrations in the studied sediment core.

(1) E_h

Both iodine and uranium are redox sensitive elements, variation of the E_h of the water could change the valence states and species of iodine and uranium, and therefore their mobility and migration in the sediment, consequentially their concentrations in the sediment. It has been observed that the mobility of iodine in sediments could be enhanced under anoxic conditions through dissociation of iodine from organic matter and metal oxides/hydroxides (Gilfedder et al., 2010; Zhao et al., 2021; Takata et al., 2013). Uranium is highly soluble in oxygenated water because it mainly presents as uranyl ion (UO_2^{2-}) which forms soluble and non-reactive carbonate complexes (Weyer et al., 2008), especially in seawater which contains high concentration of carbonate. While in anoxic condition, uranium can be reduced to insoluble and particle active U(IV) (UO_2) (Andersen et al., 2014), and then easily transferred into sediment. It has been observed that dissolved uranium concentrations in the water column significantly decreased under reducing conditions and enriched/accumulated uranium in the beneath sediments (Martine et al., 1999). Therefore, redox condition in lake water shows a reverse impact on the concentrations of iodine and uranium in the sediment.

(2) pH

No clear relationship between iodine and pH value in lake water has been reported, although slightly decreased iodide concentration in lake water with decreased water pH from 10 to 4 was observed (Zhang, 2012). Significantly increased solubility of uranium with the decreased pH values from 11 to 7 in water was observed (Iwatsuki et al., 2004).

This is mainly due to the increased formation of highly soluble UO_2^{2+}-carbonate complexes in the water (Martine et al., 1999) with the increased pCO_2 (partial pressure of CO_2) causing by the decreased pH values. However, since the relative low concentration of carbonate in fresh water (lake), the impact of pH value should be not significant on the concentration of uranium and iodine in the KLK sediment core.

(3) Temperature

The influence of temperature on the iodine and uranium concentration in the sediment might be implemented indirectly through biological intermediated processes instead of directly transferring water soluble iodine species (I^-, IO_3^- and organic iodine compounds) and uranium species (UO_2^{2+}-carbonate complexes) from water body to the sediment due to their relative stability in fresh water (Fuge, 1996; Zhang, 2012). The increased temperature could promote primary production of hydro-biota, iodine and uranium accumulated in the biota will be transferred to the sediment with the debris of dead biota, inducing an increased iodine and uranium concentration in the sediment.

(4) Suspended matter in lake water

Besides the input of organic matter to the sediment, suspended matter (here referring to these detrital minerals, Fe/Mn oxides and hydroxides input by runoffs, but excludes the organic matter discussed above) is a major source of the sediment. It was reported that the KLK sediment is composed of fine-grained (<250 μm), light colored minerals, which mainly includes silicate (~30–60%) and carbonate (~30–60%) (Zhao et al., 2009). While such materials often contain lower iodine (carbonates: 2.7 μg/g; sandstone: 0.8 μg/g,) compared to that of the KLK core sediment (21.4 μg/g) (Fuge, 1996). A mechanism that refers to the fine floating detrital minerals in water can adsorb iodine and then settle down from the upper water column has been reported, which could help increase the iodine concentrations in bottom sediments (Takata et al., 2013). Despite the low iodine concentrations of suspended particles, high iodine fluxes in lake sediment samples were measured during the flood events with increased runoff and erosion from drainage basin (Gilfedder et al., 2010). A similar scavenging way of uranium by suspended fine detrital minerals from lake water into sediments has also been reported before (Yang et al., 2015; Hao et al., 2014).

Therefore, the organic matter content (as well as temperature) and suspended matter concentrations (including both detrital minerals and Fe/Mn oxides and hydroxides) in lake water, could be the two key factors that control the synchronous changes of iodine and uranium concentrations in the studied KLK core, and the elevated organic matter content and suspended matter concentration in lake water could facilitate more iodine and uranium deposition and the formation of these four peak events.

4.1.3. Climate impact on the formation of four peak events of iodine and uranium concentrations

The regional climate could change the input amount of iodine and uranium from the drainage basin soil, as well as the organic matter content and suspended matter concentrations in lake water through runoffs, which might be manifested together as the four peak-value events of iodine and uranium concentrations in the KLK sediment core.

(1) Input from drainage basin

The Pollen records in the Keluke sediments indicated the decreased *Artemisia/Chenopodiaceae (A/C)* ratios at the depth of high iodine and uranium concentrations (Zhao et al., 2010). Negative correlations between *Artemisia/Chenopodiaceae* ratios and iodine concentrations ($r = -0.59$, $P < 0.001$), and *Artemisia/Chenopodiaceae* pollen ratios and uranium concentrations ($r = -0.38$, $P < 0.001$) have been observed at the same depth (Zhao et al., 2010) (Fig. 3 c and Fig. S2 b and c). This indicates an increase input of iodine and uranium from the drainage basin to the lake during drier periods.

In addition to existing partly as oxides, water soluble, etc., the majority of iodine in soil was observed to exist in organic phase (bound to phenolic and thiol groups in the humic substances), which was demonstrated as the key parameter controlling the retention process of iodine in soil (Duborská et al., 2019; Hou et al., 2003). The transformation of inorganic iodine to organic iodine plays an important role in the iodine immobilization in soil systems (Shimamoto et al., 2011). Therefore, when the regional climate getting drier and soil organic matter content decreased, the organic matter bond iodine could be easily leached out by runoffs from the drainage soil, and the remained forms could be directly eroded with fine particles as well.

Besides partly associated with organic matter, uranium is mainly bound to Fe/Mn oxides, as well as minerals (clays), both consist as the fine particle fractions in soils. Only a tiny amount is in water soluble fraction (Qiao et al., 2012). The two stable oxidation states are IV and VI, respectively. The tetravalence is extremely resistant to leach, however, the oxidation of U(IV) to U(VI) results in much higher uranium motility in soil environment due to the formation of stronger uranyl carbonate complexes (Owen and Otton, 1995; Lefebvre et al., 2021; Qiao et al., 2012). Therefore, when the regional climate getting drier and soil organic matter content decreased, uranium might be easily eroded with the fine soil particles, as well as be leached at the high oxidation state (VI) as the soil aeration condition getting better.

The field observations show that significant vegetation succession occurs along the elevation gradient from the high-altitude mountain in the north of the Kekule Lake (ca. 4100 m) to the lake area (ca. 2817 m), from alpine meadow, through alpine steppe, *Artemisia*-dominated steppe, to *Chenopodiaceae*-dominated desert (Zhao et al., 2010). During drier periods, the *Chenopodiaceae*-dominated desert extended, and the catchment area of lake shrinks to the *Chenopodiaceae*-dominated desert. Due to the smaller leaves of *Chenopodiaceae*-dominated vegetation, the organic matter input to soil from vegetation reduces, and organic matter content of soil decreases as results. That leads to more organic bound iodine leached out from the catchment to the lake. Additionally, when soil organic matter content lowers down, it is easier to form oxic condition and leach more soluble uranium (VI) to the lake. Meanwhile, due to the relatively low vegetation coverage of *Chenopodiaceae*-dominated desert compared to that of *Artemisia*-dominated steppe, the fixation capacity of soil decreases and the erosion of fine soil particles by runoffs intensifies, which directly leads to finer particle associated iodine and uranium eroded from the catchment and increase input to the lake. Similarly, a less developed wetland under dry climate has been reported to be less efficient in terms of uranium fixation in the drainage basin resulting in exceeding uranium inputs into the lake (Lefebvre et al., 2021).

(2) Transfer process of iodine and uranium in lake water

The correlation between iodine concentrations and LOI values ($r = 0.25$, $P < 0.10$, not high significant) and uranium concentrations and LOI values ($r = 0.45$, $P < 0.01$, significant) were observed in the KLK core. As the climate getting drier and lake water level declined (not become to saline lake), the organic matter content in lake water was found to first increase, which is stimulated by the increased light penetration and the consequent increased aquatic plant biomass (Lan et al., 2018). The increased organic matter content in lake water would consequently adsorb more iodine and uranium deposited into beneath sediments.

Additionally, a good correlation of iodine concentrations with the percentages of silicate (as an important source of suspended matter) was observed at the same depth in KLK sediments ($r = 0.55$, $P < 0.001$), as well as a relatively weak correlation of uranium concentrations with the percentages of silicate at the same depth ($r = 0.27$, $P < 0.1$) (Fig. 3d and

Fig. S2 d and e) (Zhao et al., 2009). As the climate getting drier, the fine particles erosion in the drainage basin become intensified, causing both higher suspended matter concentrations in lake water and direct input of particulate iodine and uranium in sediments. The increased suspended matter concentrations in lake water can adsorb more iodine and uranium and then deposit them into sediments.

The correlation between iodine and uranium concentrations was interrupted at the depth of 0–5 cm (since 1944 CE) compared to other parts of the sediment core, with iodine concentration peaking at the depth of 0–1 cm but uranium concentration just at a mean-value level, while uranium concentrations peaking at the depth of 3–5 cm, but the corresponding iodine concentrations just at the mean-value level. This might be attributed to the accelerated human activities in the recent decades. The studied area was a typical animal husbandry region in history with agricultural activities started to develop since 1950s, which therefore changed the usage and coverage of the soil. Similarly, the interrupt of synchronous changed paleoenvironmental proxies have been reported in a serial lake sediment due to the accelerated human activities (Zhao et al., 2009; Lefebvre et al., 2021).

4.2. *Climate change records of iodine and uranium concentrations in the sediment*

The above discussion underlines the climate impact on the variation of iodine and uranium concentrations in the lake sediments collected from arid areas. In return, the varied iodine and uranium concentrations can be used to reconstruct the climate changes during the past ~2200 yr in this region. The four dry events were only roughly identified by the widest range of both high-level iodine and uranium concentrations at 31–33 cm, 26–29 cm, 17–22 cm and 7–9 cm depth, corresponding to the dating ages of ~503–558 CE (named as H-1), ~747–1063 CE (H-2), ~1449–1626 CE (H-3), and ~ 1900–1914 CE (H-4), respectively. These should result from the relatively dry climate with increased leach/erosion and deposition intensity of iodine and uranium (Fig. 4). These events also coincide with the reported climatic changes in the Qaidam Basin (Wang et al., 2008; Yao et al., 1996; Dong et al., 2021).

In the analyzed sediment core, a relatively low and stable iodine and uranium concentrations occurred in wet period during ~542 BCE-503 CE, and the first peak value of both uranium and iodine concentrations were observed in a dry period during ~503–558 CE (H-1). Similar climate characteristics have been recorded by Rb/Sr ratios in an aeolian deposit sequences (~550 BCE-550 CE) from this area, with wet climate developed in ~350 CE-350 BCE, and dry climate in ~350–550 CE (Dong et al., 2021). Such wet and dry events have also been recorded in Guliya ice core by the glacial accumulation rate in 300–400 CE (no samples of earlier years) and 401–560 CE, respectively (Yao et al., 1996).

The dry period during ~747–1063 CE (H-2) was recorded in Guliya ice core (721–980 CE) by low glacial accumulation rate and in lacustrine sediments (850–1110 CE) by low total organic carbon content (Yao et al., 1996; Wang et al., 2008). The driest event of H-3 with the highest uranium and iodine concentrations (~1449–1626 CE) has been widely recognized and identified in tree rings (1429–1519 CE) by narrow tree ring width and in the same lacustrine sediments (~1500 CE) (Shao et al., 2004; Wang et al., 2008), but missing in Guliya ice core and instead of a cold event in 1451–1500 CE (Yao et al., 1996). The intervening wet periods, including ~558–747 CE and ~ 1063–1449 CE, also responded in the intervals of ~527–650 CE and 1200–1430 CE from the same tree ring study by wide tree ring width (Shao et al., 2004, 2006), and of ~561–740 CE and 1271–1600 CE from the same Guliya ice core study by high glacial accumulation rate (Yao et al., 1996).

The H-4 dry event occurred in ~1900–1914 CE has been recorded in Guliya ice core (1811–1930 CE) (Yao et al., 1996), lacustrine sediments (~1850 CE) (Wang et al., 2008). This dry period was also recorded in the historical documents (1875–1940 CE) (Hou et al., 2008). The intervening wet period of 1626–1900 CE is comparable with the wet period of ~1520–1633 CE in tree ring samples (Shao et al., 2004), of ~1650–1720 in historical documents (Hou et al., 2008), and of 1641–1810 CE in Guliya ice core samples (Yao et al., 1996). The slight

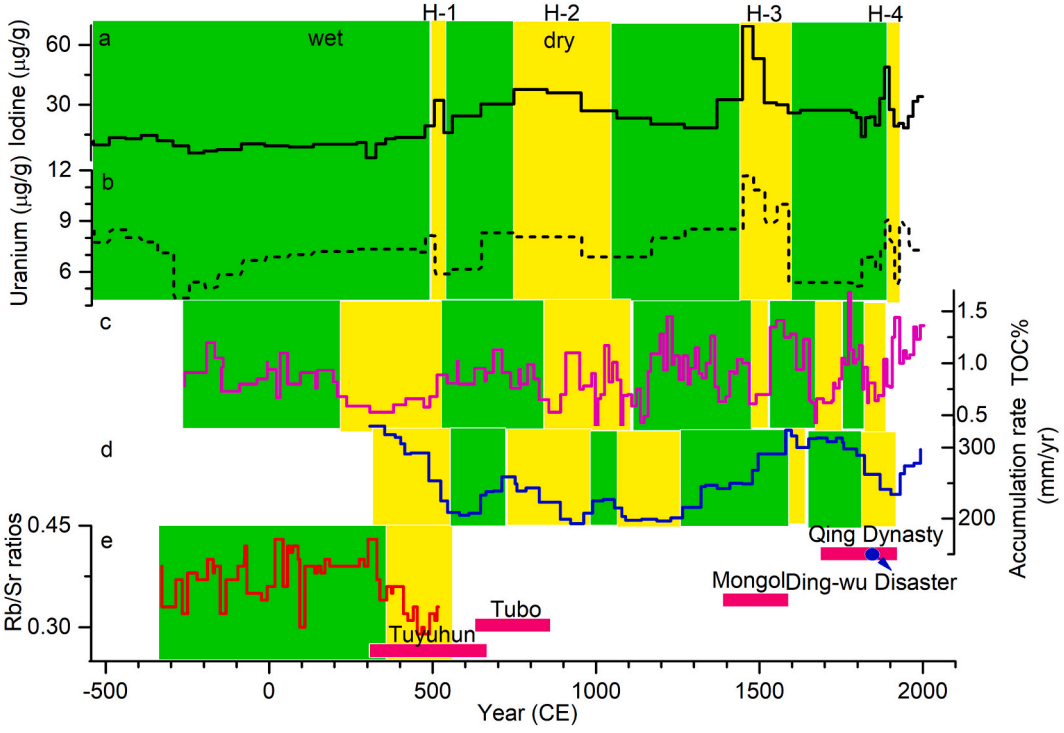

Fig. 4. Four dry events named as H1-H4 and indicated by high uranium and iodine concentrations in yellow stripes, and the internal wet periods in green stripes (a, b); Comparison with the dry-wet periods indicated by TOC in Kusai Lake sediments (c) (Wang et al., 2008), accumulation rates of snow in the Guliya ice core (d) (Yao et al., 1996), Rb/Sr ratios in aeolian sediments from Qaidam basin (only a short period before 530 CE) (e) (Dong et al., 2021), and cultural evolutions in this area (pink stripes). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

shifting of time bounds among different records is often observed, this is mainly attributed to the uncertainties of natural records or/and dating results. The cross-correlation of dry events defined in the Guliya ice-core with the 4 dry events of high-level of both iodine and uranium concentrations (H1-H4) confirmed the iodine-uranium pair proxy for dry-wet climate, which also confirmed the dating results of the studied sediment core in return.

It has also been suggested that when soils were more developed under a humid climate, it could accumulate more uranium and result in higher uranium supply to the lake with exogenous organic matter from drainage basin soil (Lefebvre et al., 2021). This indicate the uranium and iodine multi proxy could not be effective in all climate conditions but in the arid areas, and when applying such geochemical proxies, the regional geochemical circulation should be discussed firstly.

Additionally, the cultural evolution happened in this area at the similar periods with the four dry events indicated by peak uranium and iodine concentrations. As the first dry event (H-1) occurred, the Tuyuhun Kingdom (313–663 CE) became an important regional regime in the north Tibetan Plateau; In the H-2 event, the Tubo Empire (618–842 CE) defeated the Tuyuhun Kingdom, and governed this area; as the driest H-3 event occurred, several Mongolian tribes occupied successively the west area of Qinghai Lake (the studied area) since 1495, and a relatively strong Mongolian tribe was established in the middle sixteenth century, which was ended in 1591 (Han, 2000). After that, the studied area was governed by the Qing Dynasty until 1912. Although no social changes during this period, the Ding-wu Disaster, an exceptionally severe drought happened in 1876–1879 with mass human migrations towards the west from the Central China (Xia, 1993), which is close to the H-4 event (~1900–1914 CE). This indicates that the dry climate could act as a crucial external factor for the evolution of human societies in the northeastern edge of the Qaidam Basin just as in the worldwide, and the harsh living condition caused by dry climate should be an important factor to stimulate the social unrest and lead a new regime.

5. Conclusions

Based on the measured iodine and uranium concentrations in the Keluke lake sediment core collected from the Qaidam Basin and the discussion above, it can be concluded that (1) the iodine concentrations vary within 3.4–69.7 μg/g, and uranium concentrations in 4.4–11.7 μg/g; (2) a relatively high correlation between uranium and iodine concentrations ($R = 0.62$, $P < 0.001$) was observed with four similar peak value events appearing at the depth of 31–33 cm, 26–29 cm, 17–22 cm, and 7–9 cm; (3) both iodine and uranium were mainly originated from the drainage basin soil, and the organic matter content and suspended matter concentrations in lake water were the two key factors that controls the synchronous change of iodine and uranium concentrations in the studied core; (4) during drier periods, both the leaching and erosion amounts of iodine and uranium from drainage basin soils increased due to the extended xerophytic plants compared to the steppe/meadow vegetation and the decreased soil organic matter content. Accompanied with the simultaneously increased suspended matter concentrations and organic matter content in lake water, more iodine and uranium was deposited into sediments, which forms four peak events of iodine and uranium concentrations; (5) the reconstructed four dry events during the past ~2200 yr coincide with the reported climatic changes in Kusai lake sediments, Guliya ice core, and aeolian sediments in the Qaidam Basin, and also correspond well with the society evolution in this area, indicating the validity of iodine-uranium concentrations as a new dry-wet climatic proxy in arid lake sediments.

CRediT authorship contribution statement

Xue Zhao: Conceptualization, Investigation, Writing – review & editing. **Xiaolin Hou:** Writing – review & editing, Supervision, Funding acquisition, Project administration. **Zhao Huang:** Visualization, Methodology. **Chengjun Zhang:** Investigation, Validation. **Peng Cheng:** Methodology, Software.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Sample information and uranium analytical results of five surface soil samples, measurement results of $^{210}Pb_{ex}$, and dating results with this method, analytical results of iodine and uranium concentrations in the KLK sediment core, and correlations between iodine and uranium concentrations, iodine and A/C (Artemisia/Chenopodiaceae) ratios, uranium and A/C ratios, iodine and silicate percentage, uranium and silicate percentage were included in the Supplementary file.

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