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Trace metals as markers for historical anthropogenic contamination: Evidence from the Peshawar Basin, Pakistan

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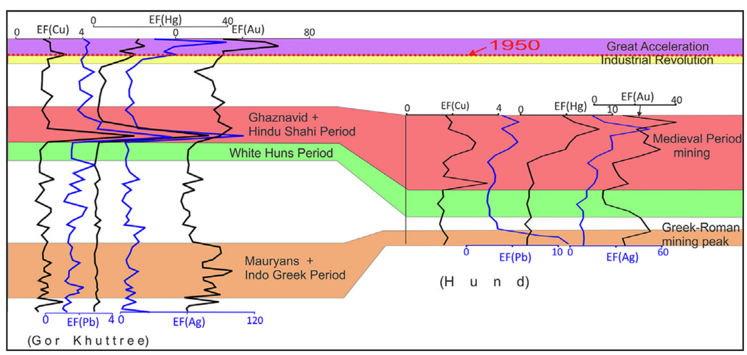
HIGHLIGHTS

- Trace elements in sediment used as signals for historical anthropogenic pollution.
- Early anthropogenic signals in the Pakistan Peshawar Basin start at 500 BCE.
- Start of early anthropogenic signals correlated with Greek and Roman periods.
- Ag and Au anomalies in sediment indicate precious metal use since 400 BCE.
- Peshawar Basin provides signals for Mid-20th century Great Acceleration.

GRAPHICAL ABSTRACT



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ABSTRACT

Trace element concentrations in the youngest Holocene sedimentary archives, historical mining, and archaeological sites are reliable indicators for historical anthropogenic contamination. The Pleistocene-Holocene strata and the overlying archaeological sites of the Peshawar Basin, NW Pakistan provide sedimentary archives to explore historical anthropogenic controls on the distributions of trace elements. The basin with 2500 y of human civilization was sampled using archaeological trenches at Gor Khuttree and Hund, and six sections of youngest Pleistocene-Holocene strata along river banks. Geochemical analysis of high-resolution samples were conducted for both the lacustrine-floodplain sediments and archaeological sites. Results from various horizons of the archaeological sites provide signals for anthropogenic control on the distribution of As, Zn, Cu, Mo, Pb, Hg, Ag, and Au during the Meghalayan Stage of Holocene that gain progressive strength since the 18th century. The geochemical proxies negate direct mining of Cu-Pb and Zn in the area. The consistent, anthropogenic Ag and Au contribution to the system throughout the basin's archaeological history is a significant finding. When correlated against the anthropogenic mercury contamination, it appears that Hund was a major silver-gold panning site throughout its known history whereas Gor Khuttree was the major silver-gold processing center. The Peshawar Basin anthropogenic signals contribute to widespread European early Anthropocene signals at around 2000 BP related to the Greek and Roman mining. Signals during the Hindu Shahi period correlate well with the Medieval period mining and smelting peak signals observed in Europe and China. Hg, Ag, and Au concentrations in the area since the start of the 19th century CE correlates to the start of industrialisation. During the mid-20th century, these geochemical signals from the Gor Khuttree reflect anthropogenic contributions to the local system and correlate to the suggested base of a formalised Anthropocene.

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1. Introduction

Human interference with the Earth System, the atmosphere, hydrosphere, biosphere and geosphere has produced multiple signals, initiating debates on the concept of a new geological epoch "Anthropocene" defined by the human factor (e.g. Crutzen and Stoermer, 2000; Crutzen, 2002; Lewis and Maslin, 2015; Waters et al., 2016; Steffen et al., 2018). Anthropogenic signals progressively gained strength towards the end of the Pleistocene, and the beginning of the Holocene displays widespread human activities dating back to the advent of agriculture, domestication of animals, and extensive deforestation in the Holocene (Foley et al., 2013; Ruddiman, 2013) leading to the early Anthropocene hypothesis (Ruddiman, 2003). The first anthropogenic trace metal peaks are signals for historical humans controls on the distribution of various metals (e.g., Fiałkiewicz-Kozieł et al., 2018), and thus early Pb pollution records were suggested as markers for the early Anthropocene (Wagreich and Draganits, 2018). However, globally the magnitude of early Pb pollution is minor by comparison to the scale of the more modern environmental Pb enrichment that begins with the Industrial Revolution (Marx et al., 2016). By the third millennium BCE (the formalised Meghalayan Stage), Mesopotamia, the Nile Valley, and the Indus Basin of Pakistan established distinct cultures (Zalasiewicz et al., 2011). Anthropogenic interference is complex, and the impact on different environmental compartments (air, water, soil) provides variable scales of changes from local to regional and global (Gałuszka et al., 2014). Anthropogenic pollution signals are best preserved in stratal archives such as sedimentary successions, hence often referred to as archives of contamination (Choi and Wania, 2011).

Freshwater depositional environments are subject to various pollution sources and intensified land use, and therefore, such sediments show elevated levels of metal contamination (Zolitschka et al., 2015). Similarly, archaeological sites, the primary places for reconstructing cultural histories, provide also stratigraphic records to understand anthropogenic impacts on Earth's ecosystems (Erlandson and Braje, 2013).

Anthropogenic fluxes have strongly increased pollutants and trace metals like Ag, Cr, Cu, Ni, Pb, and Zn which have been widely discussed in this context (Rauch and Pacyna, 2009; Rockström et al., 2009; Gu et al., 2013; Dean et al., 2014; Schlesinger, 2014; Zalasiewicz et al., 2014, 2015; Gałuszka et al., 2017; Fiałkiewicz-Kozieł et al., 2018; Gałuszka and Migaszewski, 2018; Waters et al., 2018). Similarly, Cu, Pb, Ag, Au, and other heavy metals enrichment in different environments due to mining, smelting, uses as tools or ornaments, and fossil fuel combustion has remained the subject of many studies (Nriagu, 1989; Shotyk, 1996, 1998; Renberg et al., 2000; Reuer and Weiss, 2002; Bindler et al., 2004; Le Roux et al., 2005; Bindler, 2006; Lindberg et al., 2007; Komarek et al., 2008; De Vleeschouwer et al., 2007, 2009; Yang et al., 2010).

The present study uses geochemical proxies from bulk sediment composition analyses of two prominent archaeological sites at Hund (Swabi) and Gor Khuttree (Peshawar) in search for the historical anthropogenic signals in the Peshawar Basin, NW Pakistan. Hund was the last capital of the Gandhara civilization in the Peshawar Basin during the Hindu Shahi period (822–1098 CE) while Gor Khuttree is situated in the central part of Peshawar city, the capital of the Gandhara civilization during various periods and the oldest living city of the Indian subcontinent (Samad, 2011). Geochemical analysis of the youngest Pleistocene–Holocene lacustrine-floodplain sedimentary archives has also been included to determine the natural background concentrations for the various elements used as proxies for anthropogenic signals. Additionally, ^{14}C age dating at the archaeological sites was included to constrain ages of the geochemical trends and to correlate to different cultural periods. This work focused on the possible correlation of the anthropogenic signals from the Peshawar Basin, Pakistan with the widely discussed anthropogenic signals from Greek-Roman mining peak, Medieval period mining and smelting, Industrial Revolution, and Great Acceleration.

2. General geology and minerals deposits

The Peshawar Basin, located in NW Pakistan, is situated in the southern foothills of the western Himalayan Orogen (Kazmi and Jan, 1997). The sedimentary basin developed due to the Attock–Cherat Range uplift around 2.8 Ma ago (Cornwell, 1998) and covers an area of 8300 km^2 (Rendell, 1993). The structural geology and tectonic history of the basin were discussed in detail in Hussain et al. (1989, 1991) and Hussain and Yeats (2002).

Widespread Pleistocene–Holocene strata occur within the basin and consist of thick, lacustrine-floodplain silts and clays, with interlayered sands and gravels, and are overlain by loess deposits (Burbank and Tahirkheli, 1985; Bibi et al., 2019a,b). The Indus, Kabul, and Kalpani rivers are the major drainage network of the basin (Fig. 1a). Terraces along these rivers provide natural outcrops of Quaternary sediments.

Historical mining activity within the basin and its hinterland is known from at least 3000 BCE onwards (Biswas, 2001). Porphyry Copper-Lead (Cu-Pb) and Alpine chromite deposits occur in the N-NW, whereas oolitic-pisolitic Iron deposits occur in the E-NE of the study area. However, none of these metal deposits have contributed to the basin as placers. Located to the NW of the study area, the famous Mes-Aynak Cu–deposits with the discovery of a 5000-year-old (Bronze Age) Cu–smelter (Biswas, 2001), is a proven source for anthropogenic Cu use in the area. The banks of Indus and Kabul rivers in the Peshawar Basin have long remained key points for gold (Au) panning (Shah et al., 2007). In addition, lignitic sub-bituminous coal deposits occur to the south in Attock–Cherat Range and the basin is known for clays and bricks industry (Kazmi and Abbas, 2001).

3. Archaeology

3.1. Gandhara civilization

Caves at Shanghao near Mardan (110 km north of Peshawar) provide evidence for human inhabitants at Gandhara area around 3000 years BP (Salim, 1986). The Gandhara Kingdom (Fig. 1b) included the areas now situated in modern-day Afghanistan, N-NW Pakistan, and India, was one of the sixteen kingdoms of ancient India (Behrendt, 2007; Devi, 2007), and dates back to the time of the Rig-Veda (ca. 1500–1200 BCE) and Zoroastrian Avesta (Keith, 1993). In the 6th century BCE the Achaemenid Empire conquered the study area, followed by Alexander the Great in 327 BCE (Salomon, 1999). Afterward, the area remained part of the Maurya Empire (305–180 BCE). A detailed historical overview of the area is given in Fig. 2. In the following lines; details on two prominent historical sites is given.

3.1.1. Hund (Swabi)

Hund, now a small village in Swabi district, is located 80 km east of Peshawar, Khyber Pakhtoonkhwa Province, Pakistan (Fig. 1b). The site provides a rich history, i.e., this was the place where Alexander the Great crossed the Indus River. During the Hindu Shahi period Hund was the third and last capital of Gandhara after Charsadda (Pushkalawati) and Peshawar (Parshapura) under the ruler Anandapala (Ali et al., 2014). When Mahmud of

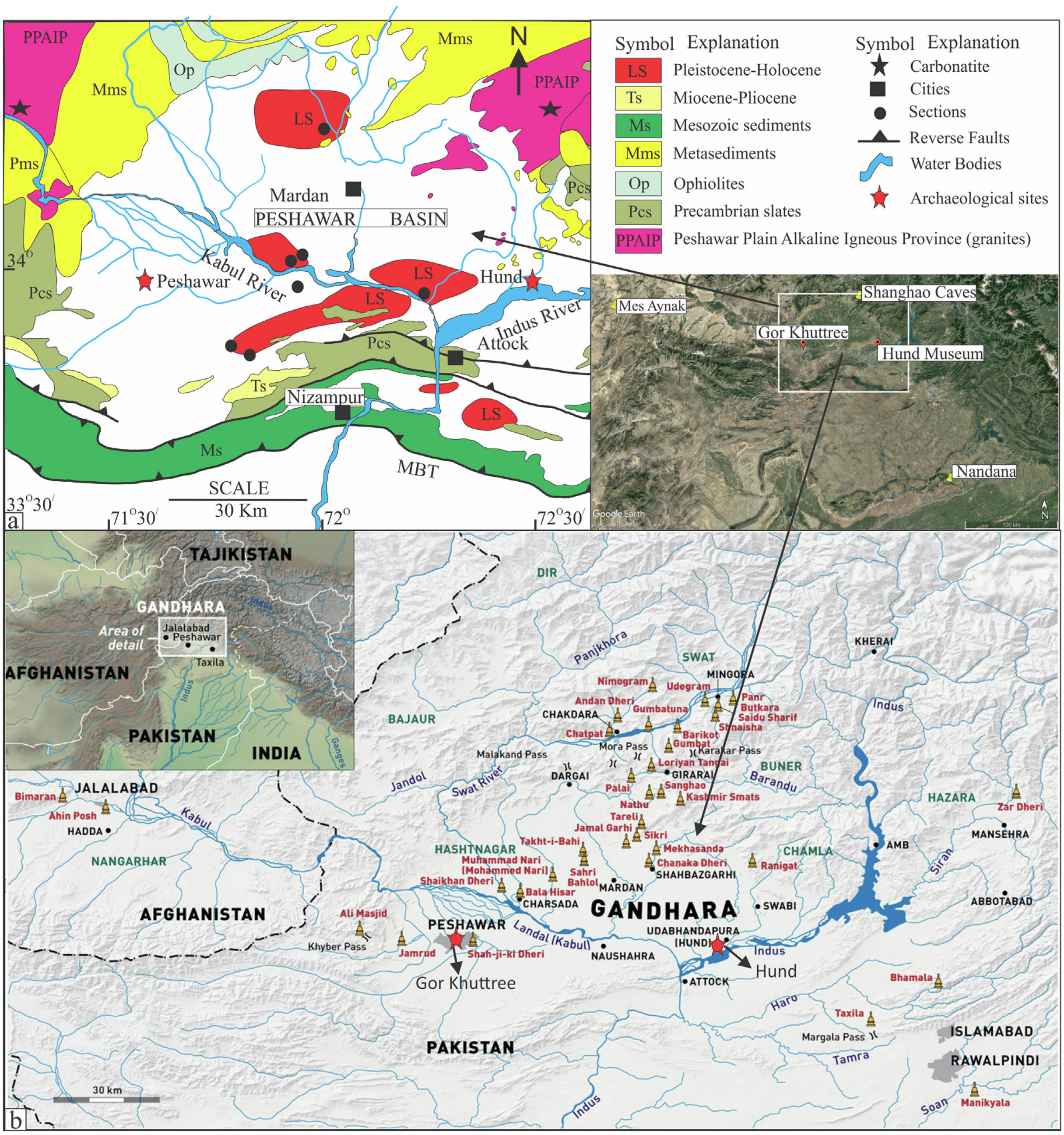

Fig. 1. (a) Geological map of the Peshawar Basin and the surrounding area with the geographic position of Pakistan and tectonic map of north Pakistan. The white colour in the geological map represents the area covered by the youngest alluvium under human use (agriculture and population and therefore are reworked). MBT = Main Boundary Thrust (Bibi et al., 2019a,b). (b) Location map of the Gandhara Civilization. The inset indicates the important locations. The important archaeological sites are labeled and the Gor Khuttree site (Peshawar) and Hund (Swabi) are marked with red star symbols (Naveed, 2015). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Ghazna defeated Anandapala (1001–1010 CE) the capital was shifted to Nandana (Salt Range).

The Chinese Buddhist pilgrim Xuan Zang visited the Gandhara area in 644 CE and mentioned Hund by the name "wo-to-kia-han-cha". Sarda inscriptions found at Hund reveal the city by the name of "Udabhandapura". Similarly, the 12th century CE history of Kashmir "Rajatarangini" (1151 CE, written by Kalhaṇa) mentions the city by the same name (Stein, 1892). Excavation in the site has revealed coins, beads, pottery, and weaponry dating back to the Kushan (1st–4th century CE) and Hindu Shahi (800–1100 CE) periods.

3.1.2. Gor Khuttree (Peshawar)

Peshawar is considered to be the oldest living city in the Indo-Pakistan subcontinent. According to the Hindu religious literature the city was founded around 7000 BP. Al-Biruni (Samad, 2011) mentioned it first as "Pershawar". Gor Khuttree, located in central part of the ancient walled city, is the highest place in Peshawar. The present-day structure of the Gor Khuttree complex was constructed by Jahanara Begum (daughter of the Mughal emperors Shah Jahan) in 1640 CE (Jaffar, 2008). Excavations led by the Department of Archaeology, University of Peshawar initiated in

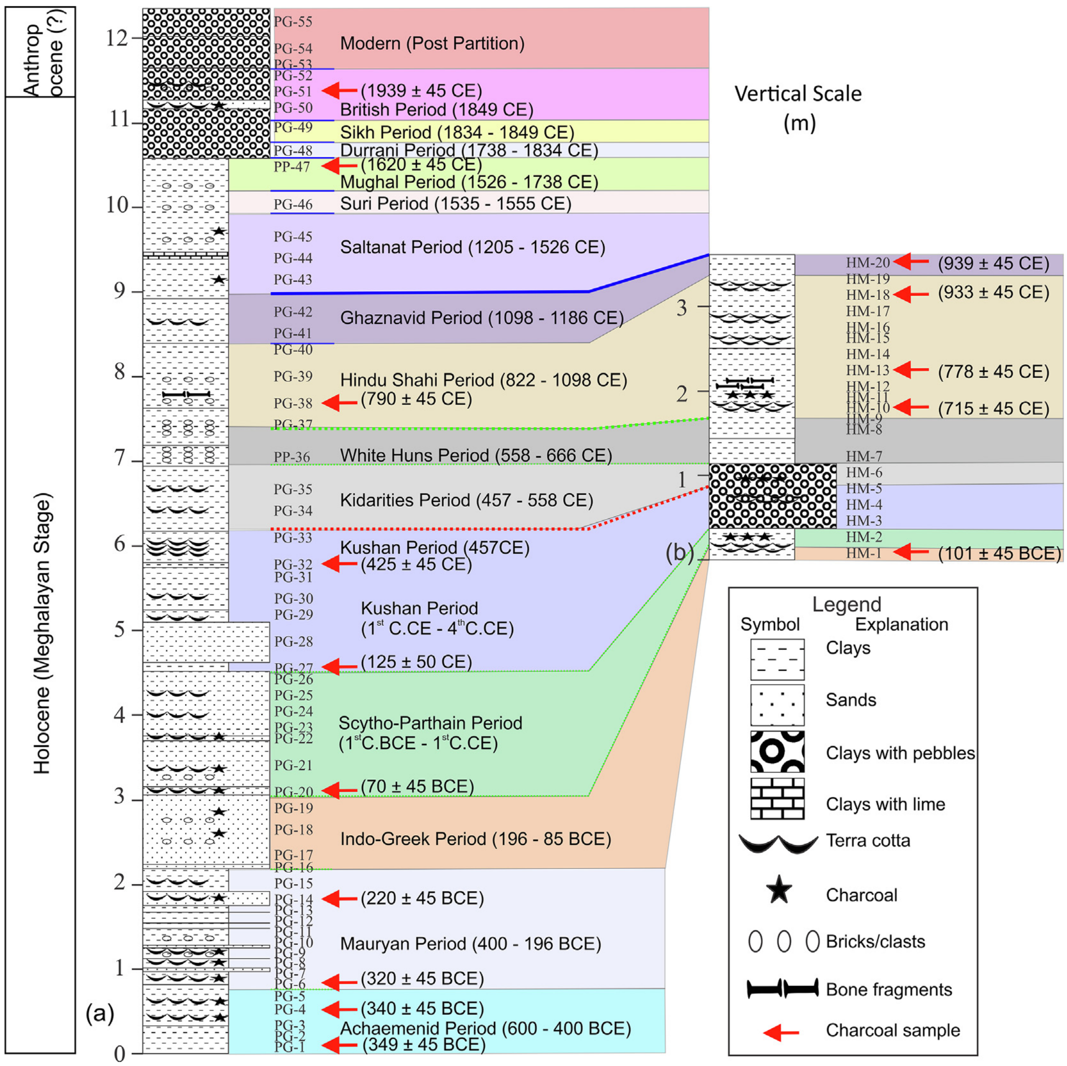

Fig. 2. Lithological log of the archaeological trenches at (a) Gor Khuttree and (b) Hund. The chronological correlation between the two archaeological sites is also provided (Ali et al., 2005; Samad, 2011). Sample positions are indicated and the Red arrows mark the position of charcoal samples with the calibrated radiocarbon ages. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

1992–93; the more than 12 m deep trench dates the city age to at least 300 BCE (Ali et al., 2005; Petrie et al., 2008).

4. Material and methods

4.1. Outcrops and sampling

Six outcrop stratigraphic sections (Fig. 1a) of Pleistocene–Holocene strata in the basin were selected and high-resolution samples (10–50 cm sampling interval), were collected from all the lithofacies for geochemical analysis. These outcrops are located at Garhi Faizullah (33°51′52.08″N, 71°42′42.362″E), Nowshera south (34°00′03.06″N, 71°58′13.50″E), Kabul River (34°00′30.60″N, 71°58′13.50″E), Nowshera-Risalpur road section (34°01′46.98″N, 71°59′57.45″E), Jalala (34°19′52.14″N, 71°54′526.16″E), and Jahangira (33°58′38.81″N, 72°12′52.17″E). The sediments samples were collected covering the entire variation including the floodplain clays lithofacies, lacustrine-fluvial plain lithofacies, alluvial fan lithofacies, glacio-fluvial lithofacies, and loess lithofacies (see Bibi et al., 2019a). Since these sediments had sedimentation rates of 2–15 cm/ka (Burbank, 1983; Bibi et al., 2019a,b), the sampling interval covers on average 1–3 ka time intervals. However, the sampling interval was reduced to only few cm in the very thin-bedded strata and expanded to around 70 cm in the relatively thicker beds. Each sample was collected from vertically <2 cm thickness to make sure that it represented specific and short deposition episode. Furthermore, fresh samples were collected by digging into the sediment and removing any material with signs of surficial weathering. Also, contamination by any possible post-depositional source (e.g., penetrated bullet or large metal pieces) or any other recent anthropogenic activity were removed. For fine-grained strata the average sample size was 0.5 kg that was increased to around 2 kg in coarse-grained sediments. In total, 80 samples were collected to determine the background geochemical composition of sediments in the basin.

4.2. Archaeological sites

Samples at the archaeological sites of Hund (34°00′48.30″N, 72°25′59.42″E) and Gor Khuttree (34°01′05.20″N, 71°34′08.82″E) were collected from the unconsolidated alluvium with a rich artifact content. These alluvium facies consisted of interbedded clays, silt, with multiple sandy and pebbly horizons correlatable to the sedimentary archives. All the above-mentioned precautions for sampling in sedimentary archives were strictly adopted during sampling at the archaeological sites. Additionally all the larger size clasts including stone pebble, bricks, bone fragments, and other such objects were removed by mechanical handpicking and followed by sieving.

A 12.5 m thick section (excavation site) at Gor Khuttree Complex, Peshawar, was sampled for geochemical analysis to detect anthropogenic signals in the area using 60 high-resolution samples (Fig. 2a). The 3.5 m thick section (excavation site) at Hund Museum was sampled at high resolution (10–20 cm interval) and 25 samples were collected for geochemical analysis (Fig. 2b). Here the high-resolution samples mean that the sampling interval covered on average 20–40 years. Again, the sampling interval was reduced to few cm in the thin beds while it was expanded to around 30 cm in the thick beds. The sample size mentioned for outcrops was also followed here. It is further mentioned that 5 samples were collected as duplicates from Gor Khuttree and from Hund, respectively, to see the lateral variation in the same beds. Therefore, these duplicate samples are not labeled on the litho-logs (Fig. 2). Thus the total number of samples on the litho-log is 55 for Gor Khuttree and 20 for Hund (Fig. 2). Unfortunately, no drill core was collected at the archaeological sites during the open excavation, and the authors were not allowed to collect cores at the base of these sites.

4.3. Bulk geochemistry

Coarse-grained particles were removed from each of the 165 samples first by mechanical handpicking and followed by sieving to avoid the unwanted impact of grain-size. Each sample was powdered and homogenised for bulk rock geochemistry (Supplementary material 1). Inductively Coupled Plasma Optical Emission Spectroscopy/Mass Spectrometry (ICP- OES/MS) was used at the Bureau Veritas, Canada for the analysis. All the major, minor oxides, trace and rare earth elements (REE) were determined. Since this work is based on the selected trace metals (As, Zn, Mo, Cu, Pb, Hg, Ag, and Au) therefore, data of only these elements are included. Al is used as the reference element. Each sample was fused in Lithium metaborate/tetraborate and digested in nitric acid and aqua regia to extract the chemical species using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES; Spectro Ciros Vision) and ICP-MS (Perkin Elmer ELAN 9000, Sciex). Reference materials SO18, DS 10, and OREAS 45 EA were used for calibration. 15 samples were analysed in duplicate and the average analytical error in the analyses is 0.1%.

4.3.1. Geochemical parameters

Several geochemical parameters are widely used to evaluate the anthropogenic influence and thus develop contamination indices for sediments (Gałuszka et al., 2014). Presently, As, Zn, Mo, Cu, Pb, Hg, Ag, and Au were selected as proxies for anthropogenic signals. The background values used in the geochemical proxies were the average values for each element in background sediments. Cu, Pb, Ag, and Au have been historically mined by human and As, Zn, Cu, Mo, Pb, and Hg have been used in many studies as proxies for anthropogenic signals based on the following geochemical parameters:

4.3.1.1. Enrichment Factor (EF).

$$\boldsymbol{EF} = (\boldsymbol{A_e} \times \boldsymbol{B_c}) \div (\boldsymbol{A_c} \times \boldsymbol{B_e}) \tag{1}$$

where A_e is element concentration in the sample, B_e is reference element concentration in the sample, A_c is the Clarke value of the element, and B_c is the Clarke value of the reference element (Halstead et al., 2000; Sutherland, 2000). The Clarke value is generally considered as the average shale value but in this equation, these refer to the average background sediment concentrations.

4.3.1.2. Degree of Contamination (C_{deg}). In order to calculate C_{deg} first the Contamination Factor (CF) for each element is obtained when the element concentration in the sample is divided by the background concentration of the element. C_{deg} is the sum of all the CF values for the given sample (presently eight pollutant species (As, Zn, Mo, Cu, Pb, Hg, Ag, and Au) as defined (Hakanson, 1980).

$$\boldsymbol{C_{deg}} = \boldsymbol{CF}_1 + \boldsymbol{CF}_2 + \boldsymbol{CF}_3 + \cdots + \boldsymbol{CF_n} \tag{2}$$

where CF = $C_{(sample)}/C_{(background)}$,

4.3.1.3. Pollution Load Index (PLI). PLI (Tomlinson et al., 1980) is determined using the contamination factors (CF) of different elements in the same sample with respect to their background values:

$$\boldsymbol{PLI} = \sqrt[n]{\boldsymbol{CF}_1 \times \boldsymbol{CF}_2 \times \cdots\cdots\cdots\cdots.. \times \boldsymbol{CF_n}} \tag{3}$$

where n is the number of elements used in the calculation (8 in the present case).

4.3.1.4. Geoaccumulation Index (I_{geo}). I_{geo} (Müller, 1969), is a quantitative measure of the extent of pollution of the sample:

$$\boldsymbol{I_{geo}} = \boldsymbol{log}_2(\boldsymbol{C_e} \div 1.5\boldsymbol{GB}) \tag{4}$$

where C_e = concentration of the examined element in the sample, GB = geochemical background concentration, and 1.5 = correction factor for possible variations in the geochemical background.

4.4. Radiocarbon dating

In total, 15 samples were selected for radiocarbon dating, including 5 samples from the Hund and 10 from the Gor Khuttree excavation sites (Fig. 2). The analysis was conducted at CEntro di DAtazoni e Diagonostica (CEDAD) (Accelerator Mass Spectrometer (AMS) and radiocarbon dating facility, University of Lecce, Italy).

All samples consisted of charcoal that was mechanically picked from fresh exposures. Each of the mechanically handpicked charcoal samples was converted to carbon dioxide (CO_2) by combustion in sealed quartz tubes. The obtained CO_2 was converted into graphite at 550 °C by using ultra-high purity hydrogen as reducing medium and 2 mg iron powder as catalyst. The radiocarbon concentrations were determined in the AMS by comparing the ^{12}C, ^{13}C currents and the ^{14}C counts obtained from the samples with those obtained from standard materials supplied by the International Atomic Energy Agency (IAEA) and National Institute of Standard and Technology (NIST).

The "conventional radiocarbon age" was calculated with a $\delta^{13}C$ correction based on the $^{13}C/^{12}C$ ratio measured directly with the accelerator. These ages were converted into calendar years using the software OxCal Ver. 3.5 based on the last atmospheric dataset (Reimer et al., 2013). These ages were then calibrated against the ages for the sampling layers (Fig. 2) determined by the archaeology department (Ali et al., 2005) based on the artifact material and radiocarbon dating.

5. Results

5.1. Outcrops

The Pleistocene–Holocene sediments of the Peshawar Basin in all the studied sections consisted of well-preserved, unconsolidated strata with no indications of reworking. In the southern part near the Attock–Cherat Range (Garhi Faizullah) the basal part of the strata consisted of fining-upward, clay-rich, rhythmic cycles that constituted the floodplain lithofacies (Bibi et al., 2019a). These were overlain by multiple, poorly-sorted pebbles and boulder rich horizons of alluvial fan facies. Basinward, in the Jahangira section, Nowshera–Risalpur road section, and Jalala section fining-upward cycles of greenish-gray, coarse and micaceous sand occurred at the same level and constituted the lacustrine-fluvial floodplain lithofacies. Multiple conglomerate horizons with well-rounded pebbles of granitic and basic igneous rocks with a diameter range of 5–20 cm occurred within this interval. In places, poorly sorted, angular clasts of quartzite and slate that were comparatively larger in diameter than the rounded clasts, were also common. These constituted the glacio-fluvial lithofacies. These deposits were overlain by loess deposits in upper parts of all the sections. The loess lithofacies was generally orange to red colour and yielded calcrete concretions and multiple carbonate-rich horizons. The oldest part of the deposits yielded a Thermoluminescence dating (TL) age of 2.8 Ma (Burbank and Tahirkheli, 1985). The uppermost part of the loess lithofacies is as young as 3–17 ka (Bibi et al., 2019b).

5.2. Excavation sites

The two excavation sites consist of construction remains and also provide stratified alluvial deposits rich in artifacts. In the following lines details of the two archaeological sites have been provided:

5.2.1. Gor Khuttree

The 12.5 m trench at the Gor Khuttree revealed a stratigraphy of the area in the form of alluvial deposits rich in artifacts. The alluvial deposits were dominated by unconsolidated clays of gray and yellowish-orange colours. The lowermost part consisted of orange-red clays and apparently, it resembled the uppermost part of the loess lithofacies of the background sediments (Fig. 2a). Multiple clays and sandy layers, with charcoal particles, terra cotta clasts, and red brick clasts were observed.

The construction consisted of stone walls with dressed blocks similar to those observed in Hund while walls made from solid red bricks were present at different levels. The sampling focused on the clays, associated thin green sands and charcoal patches at various intervals of the site.

5.2.2. Hund

The excavation site at Hund consisted of construction remains of various ruling dynasties and a well-preserved alluvial deposits section. The trench in the Hund Museum provided a 3.5 m section with stratigraphy in the form of unconsolidated alluvial deposits (Fig. 2b). Multiple layers consisting dominantly of fine-grained orange-red clays with terra cotta clasts, charcoal particles, bone fragments, and rock fragments were observed.

Based on variation in the construction style, the site was subdivided into four (4) different construction periods: (1) Walls made of undressed and rounded stones (2) Well-constructed walls that met at right angle and consisted of well-dressed stones (3) The third cultural phase had construction style resembling the first one. (4) The topmost part consisted of construction similar to the second episode but lacked the typical symmetry.

5.3. Geochemical data

5.3.1. Background concentration

Background values were determined by the average of all the outcrop section samples (80 samples). The background sediments were enriched in As (Mean = 6.8 mg kg^{-1}) in comparison to the Upper Continental Crust (UCC, taken from Rudnick and Gao, 2003) (As = 4.8 mg kg^{-1}) and Cu (Mean = 33.9 mg kg^{-1}) against the 28 mg kg^{-1} value for the UCC. Ag and Au had background values similar to the UCC (0.05 mg kg^{-1} and 1.5 μg kg^{-1} respectively). The mean average Pb, Zn, Mo, and Hg (10.7 mg kg^{-1}, 60.9 mg kg^{-1}, 0.7 mg kg^{-1}, and 0.01 mg kg^{-1} respectively) were depleted in comparison to the UCC (17 mg kg^{-1}, 67 mg kg^{-1}, 1.1 mg kg^{-1}, and 0.05 mg kg^{-1} respectively) (Fig. 3).

5.3.2. Excavation sites

Arsenic (As) had a range of 5.5–13.1 mg kg^{-1} at Gor Khuttree (mean = 9.3 mg kg^{-1}) and 4.8–11.5 mg kg^{-1} at Hund (mean = 8.5 mg kg^{-1}). The lowest values occurred in the upper part at Hund site (HM-18) while the upper part at Gor Khuttree yielded the highest value of 13.1 mg kg^{-1} (Fig. 3).

Zinc (Zn) displayed a range of 55–106 mg kg^{-1} at Gor Khuttree (mean = 75 mg kg^{-1}) and 58–169 mg kg^{-1} at Hund (mean = 82.8 mg kg^{-1}). The lowest value (55 mg kg^{-1}) occurred in the lower part of Gor Khuttree (PG-2) and the highest value (169 mg kg^{-1}) in the upper part at Hund (HM-16). Both sites displayed increasing up section trends with relatively low values in the middle parts (Fig. 3a, b).

Molybdenum (Mo) had a range of 0.6–2.9 mg kg^{-1} in Gor Khuttree that expanded to 1.2–10.4 mg kg^{-1} at Hund. Both sections displayed two cycles of roughly increasing up section trends (Fig. 3a, b).

Copper (Cu) displayed a general 30–40 mg kg^{-1} range in both sites. However, anomalous values of 587 mg kg^{-1} (PG-38 at Gor Khuttree) and 83 mg kg^{-1} (HM-10 at Hund) occurred in the two sites (Fig. 3a, b).

Lead (Pb) displayed a 20–40 mg kg^{-1} range (mean = 22.4 mg kg^{-1}) at Gor Khuttree with an extremely high value of 309 mg kg^{-1} in the middle part (PG-38). Hund displayed a higher range of 30–70 mg kg^{-1} (mean = 58.4 mg kg^{-1}). The lowermost part of Hund site displayed the highest Pb content of 270 mg kg^{-1} (HM-1). Similarly, the upper part yielded values in excess of 70 mg kg^{-1} (Fig. 3; HM-16, 19).

Mercury (Hg) content possessed a 0.01–0.4 mg kg^{-1} range in both sites in progressive increasing trends. The middle part of Gor Khuttree possessed higher Hg content of 1.25 mg kg^{-1} (PG-38) (Fig. 3).

Silver (Ag) content had a general 0.2–2 mg kg^{-1} range for both sites in increasing up section trends. The middle part at Gor Khuttree (PG-38) displayed extremely high value of 4.2 mg kg^{-1} (Fig. 3).

Gold (Au) content displayed a range of 4–58 μg kg^{-1} for both sites in general up section increasing trends (Fig. 3a, b). The Gor Khuttree site possessed a mean Au content of 18.7 μg kg^{-1} that increased to 20.8 μg kg^{-1} in the Hund.

5.4. Geochemical proxies

5.4.1. Enrichment factor (EF)

Only one sample (HM-16 at Hund) displayed EF > 3 for Zn (Fig. 4b). Only two samples for Cu yielded higher EF of 29 (PG-38 at Gor Khuttree) and 3.5 (HM-10 at Hund), respectively. As displayed a maximum value of 3.7 in the upper part at Gor Khuttree (PG-51) and 2.8 (HM-14) at Hund. Mo at Gor Khuttree showed a number of samples with EF > 3 in an increase up section trend. The Hund site displayed a mean EF of 3.6 and the highest EF value was 10.8 (HM-6). Around 40% of the samples had EF > 3 (Fig. 4a, b).

Both sites displayed EF < 1 for Hg in the lower and middle parts, however, their upper parts had very high Hg EF > 8 (Fig. 4a, b). The highest EF = 34.6 for Hg occurred in the middle part of Gor Khuttree site (PG-38) whereas the highest EF at Hund was 8.6 (HM-18). Although Pb displayed a general EF < 3 for both sites, the middle part of Gor Khuttree had an EF = 25 (PG-38) and the lowermost part of Hund had 18.5 (HM-1). Ag displayed extremely high EF values for both sites, with mean average EF in excess of 15. Very few

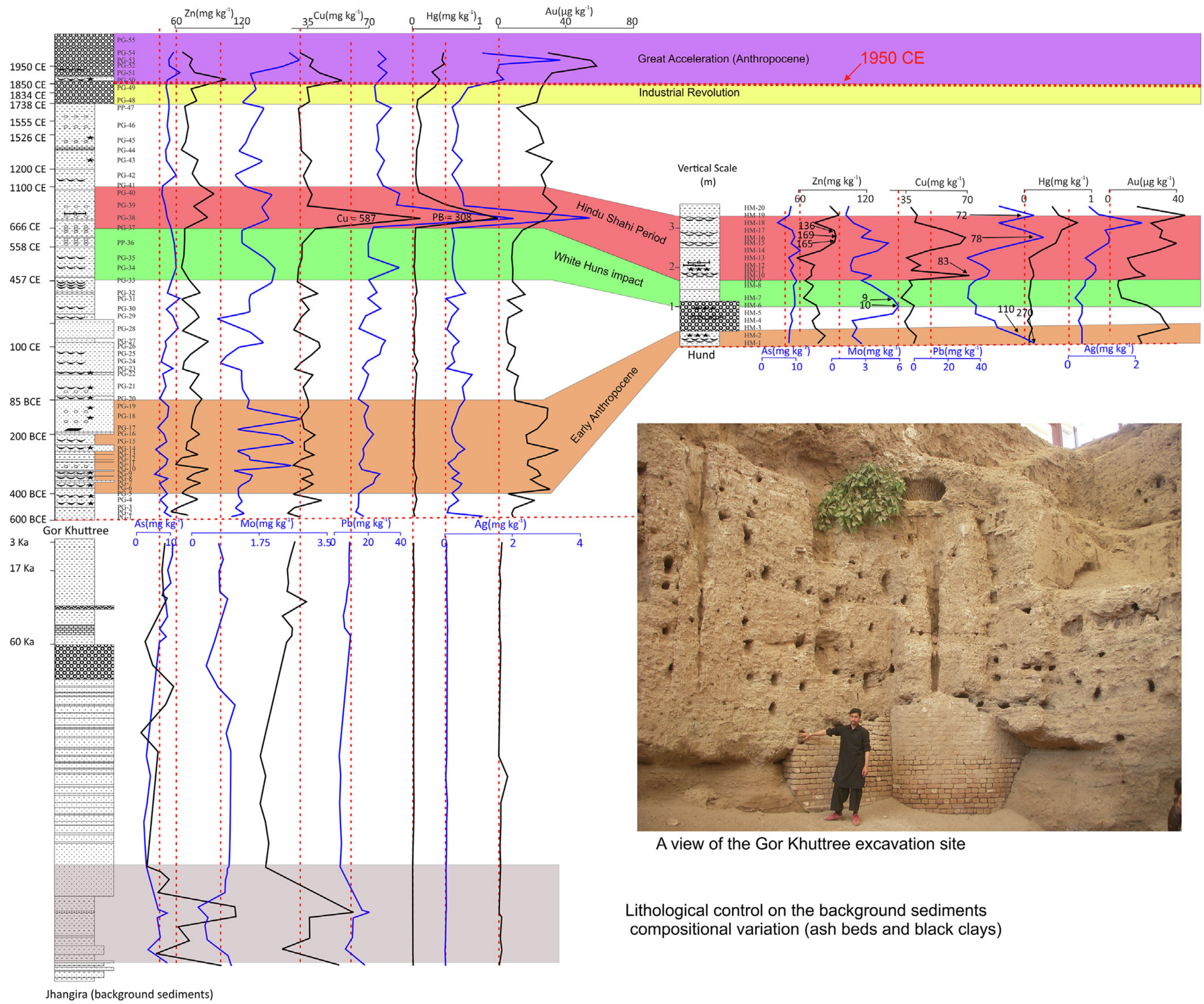

Fig. 3. Logs of the (a) Jahangira-Gor Khuttree and (b) Hund showing the background concentrations of elements used as Anthropocene proxies and their concentration in the Gor Khuttree and Hund cites. The dashed red lines represent the average background concentrations in each case. A photograph of Gor Khuttree site is provided to have an idea of the sampled material. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

samples at Gor Khuttree had EF < 5, with the highest EF = 109.8 (PG-38) and persistently EF > 30 occurred in the upper part. The Hund site did not display EF < 6.6 with the exception of one sample (HM-7 EF = 4.4) and the highest EF = 52.3 (HM-18). Au had extremely higher EF values similar to those for Ag with both sites displaying mean average EF > 18 (Fig. 4a, b). The maximum EF value for Au at Gor Khuttree was 60.9 (PG-52) while at Hund it was 40.1 (HM-19).

5.4.2. Degree of contamination (C_{deg})

Gor Khuttree revealed a C_{deg} range of 11.6–249.7 with a mean average of 39.5 (Fig. 5a). <10% of the samples had C_{deg} < 16 and>45% had C_{deg} > 32 while the remaining had C_{deg} in the 24–32 range. PG-38 displayed the highest C_{deg} = 249.7 (Fig. 5a). Hund possessed a mean average C_{deg} = 48.4 and an overall C_{deg} range of 27.9–96.8. Like the Gor Khuttree, Hund displayed a general increasing up section C_{deg} trend (Fig. 5b).

5.4.3. Pollution Load Index (PLI)

PLI ranged at Gor Khuttree from 1.3 to 12 (mean = 3.3). The lower and middle parts had PLI < 3, the upper part of the section yielded higher values (Fig. 5). The highest value (12) occurred in the middle part (PG = 38). Hund had a PLI range of 2.5–6.4 (mean = 4) with HM-15 had PLI = 6.4.

5.4.4. Index of geoaccumulation (I_{geo})

Both sites yielded I_{geo} values of < 1 for As, Zn, and Cu also but with the exception of I_{geo} = 3.5 (PG-38). Only 16% of the samples at Gor Khuttree, mostly in the upper part, displayed Mo I_{geo} > 1 (Fig. 6a). Hund had a maximum I_{geo} of 3.3 for Mo and 40% of the samples had I_{geo} < 1. Pb generally yielded relatively higher I_{geo} values for Gor Khuttree and PG-38 possessed the highest I_{geo} = 4.3. Hund displayed an I_{geo} range of 1–4.1 (mean = 1.7) and HM-1 had the highest value of I_{geo} = 4.1 (Fig. 6b).

Hg had an I_{geo} range of 0–6.4 for Gor Khuttree with around 50% of the samples possessed I_{geo} > 1 (PG-38 had 6.4). The site yielded values in excess of 4 in the upper part (Fig. 6a). Hund yielded an I_{geo} range of 1–4.5, with the upper part displaying persistent high values of I_{geo} > 3. Ag had an I_{geo} range of 0–5.8 for Gor Khuttree. Only 11% of the samples had I_{geo} values of < 1 (lower half of the section) with PG-38 had I_{geo} = 5.8 and the upper part yielded I_{geo} values > 3. Ag revealed an I_{geo} range of 2–4.9 range (mean = 3) for Hund. The upper half of the section had consistent high I_{geo} values > 3 (HM-19 I_{geo} = 4.9). Au produced an I_{geo} range of 0.9–4.7 (mean = 3.1)

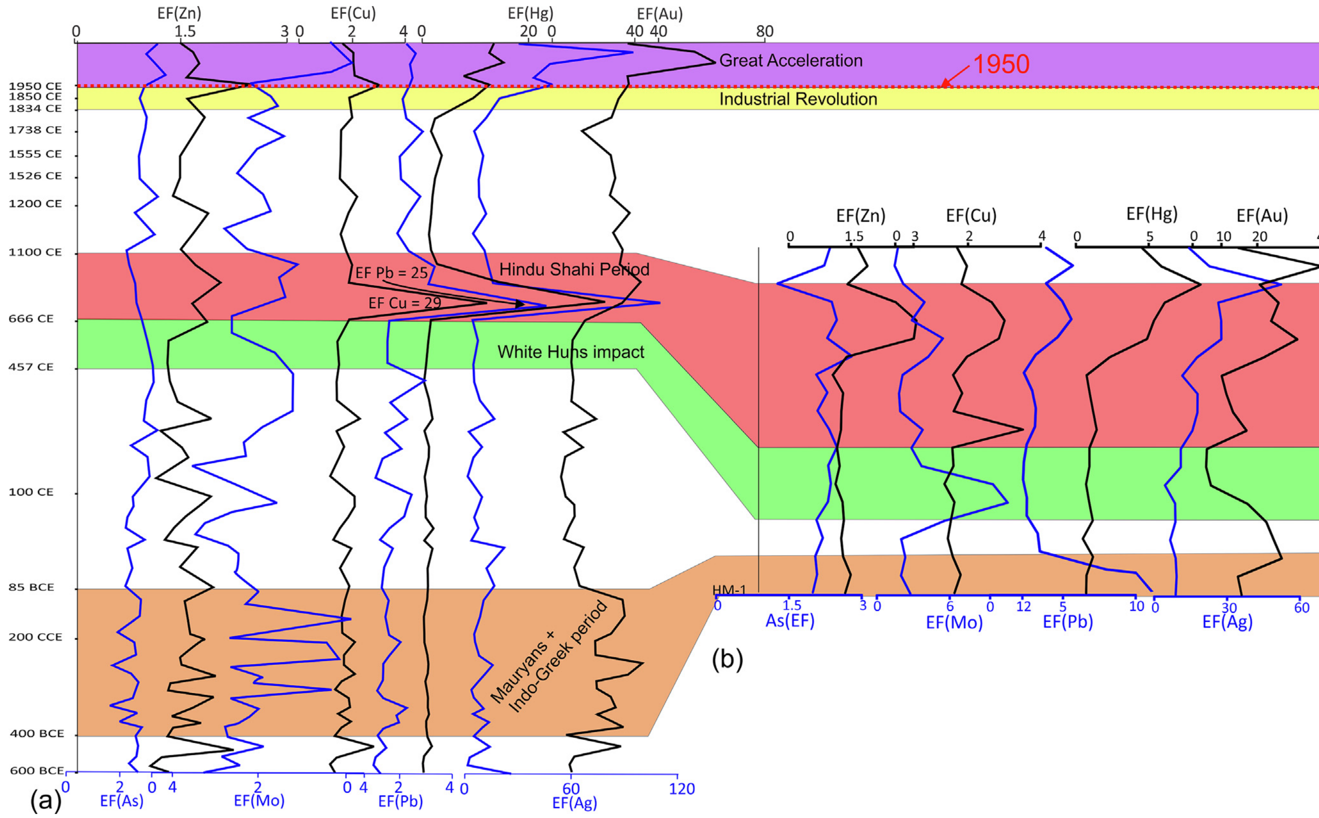

Fig. 4. Enrichment Factors (EF) for the various elements at (a) Gor Khuttree site and (b) Hund site. The chronological correlation is also provided for the labeled time intervals.

for Gor Khuttree and the upper part had the highest I_{geo} value of 4.7 (PG-52). Hund had an I_{geo} range of 1.3–4.3 (mean = 3.1). Around 65% of the samples had $I_{geo} > 3$ (HM-19 had $I_{geo} = 4.3$) (Fig. 6b).

5.5. Radiocarbon dating

The lower part of Gor Khuttree revealed minimum calibrated age of 349 ± 45 BCE. The middle part yielded a calibrated minimum age of 790 ± 45 CE. The uppermost part produced an age of 1939 ± 45 CE. The lower part of Hund yielded a calibrated age range of 101 ± 45 BCE. The middle part yielded an age range of 778 ± 45 CE while the upper part had 939 ± 45 CE age (Fig. 2).

6. Interpretation of the results

It is important to establish the chronological context of various stratigraphic layers before discussing the geochemical proxies of these layers.

6.1. Ages of the sites

Radiocarbon dating at Gor Khuttree suggested that the existing Peshawar city dated back to at least 5th century BCE and provided archaeological records as young as mid-20th century CE. Similarly, Hund dated back to at least the 4th century BCE and concluded at around 1000 CE (Alexander the Great crossed the Indus River at the very same site during his invasion on India in 324 BCE, Khan et al., 2012). Radiocarbon dating and outcrop based correlation of the two sites provides a detail of the various cultural periods in the area (Fig. 2; Table 1).

6.2. Geochemical proxies

6.2.1. Enrichment factor (EF)

As, Zn, and Mo indicate only minimal enrichment (Halstead et al., 2000) in both sites. Cu, Pb, and Hg yielded very high enrichment ($20 \leq EF \leq 40$) at Gor Khuttree only during the Hindu Shahi period. Hg also indicated significant enrichment ($5 \leq EF \leq 20$) at Gor Khuttree during the Sikh, British, and post-partition periods (Table 1). At Hund, Pb yielded significant enrichment during the Indo-Greek, Scytho-Parthian, Hindu Shahi, and Ghaznavid periods. Hg also displayed significant enrichment at Hund during the Hindu Shahi and Ghaznavid period.

At Gor Khuttree Ag yielded high enrichment factors during the Achaemenid period that dropped to significant enrichment during the Mauryan and Indo-Greek periods. Ag again reached very high enrichment during the Scytho-Parthains period followed by significant enrichment from the Kushan period until the white Huns period. Upsection at this site Ag yielded extremely high enrichment ($EF \geq 40$) during the Hindu Shahi period (Table 1) that again dropped to significant enrichment during the Ghaznavid period to Durrani periods. During the Sikh period, Ag displayed very high enrichment that further increased to extremely high enrichment during the British and post-partition periods (Table 1). On the other hand, at Hund, Ag displayed significant contamination until the White Huns period followed by an extremely high enrichment trend during the Hindu Shahi period correlatable to Gor Khuttree. The Overlying Ghaznavid period yield very high Ag enrichment at Hund (Table 1).

Au displayed very high enrichment at Gor Khuttree until the Indo-Greek period that dropped to significant enrichment during the Scytho-Parthians to White Huns periods. During the Hindu Shahi to Sikh periods, Au yielded very high Au enrichment. The British and post-partition periods at Gor Khuttree displayed extremely high Au enrichment (Table 1). Until the White Huns period

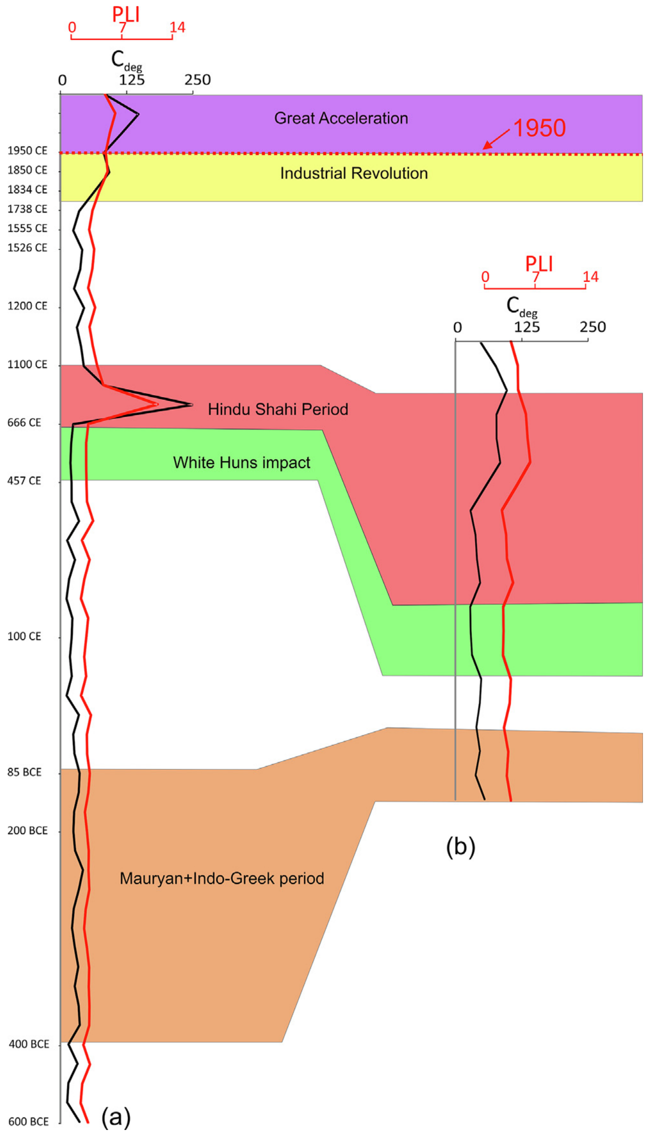

Fig. 5. (a) Degree of Contamination (C_{deg}) at Gor Khuttree site (black line) and Pollution Load Index (PLI) at Gor Khuttree site (red line), (b) degree of Contamination (C_{deg}) at Hund site (black line) and Pollution Load Index (PLI) at Hund site (red line). The important periods of anthropogenic influences are highlighted and correlated where possible. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Au displayed significant enrichment at Hund with very high enrichment during the Kushan period. The overlying Hindu Shahid and Ghaznavid periods again displayed very high Au enrichment.

6.2.2. Degree of contamination C_{deg}

Gor Khuttree displayed considerable C_{deg} ($16 \le C_{deg} \le 32$) until the White Huns period. Consistently very high C_{deg} ($C_{deg} \ge 32$) is indicated at this site since the Hindu Shahi period with the exception of the Mughal period that yielded considerable C_{deg} (Table 1). Hund yielded very high C_{deg} throughout its history with the exception of a considerable C_{deg} during the Kidarities–White Huns periods (Table 1).

6.2.3. Pollution Load Index (PLI)

The PLI data indicated that the entire geochemical history of both the archaeological sites was anthropogenically polluted (Table 1) (*sensu* Tomlinson et al., 1980). Gor Khuttree section showed relatively low pollution until the White Huns period. The overlying Hindu Shahi period displayed the highest pollution that was followed by very high pollution in an increasing up section trend (Fig. 5a). Hund, on the other hand, supported higher pollution in the area than Gor Khuttree throughout its history in increasing up section anthropogenic influence (Fig. 5b).

6.2.4. Index of Geo-accumulation (I_{geo})

The I_{geo} values for As and Zn did not indicate pollution (e.g., values below 1; Müller, 1969) in the area (Table 1). Mo indicated significant pollution ($1 \le I_{geo} \le 2$) at Gor Khuttree during the Mauryan, Indo-Greek, Kidarrites periods, and British period and onward. On the other hand, Mo at Hund yielded significant pollution until the Scytho-Parthian period and extremely high pollution during the Kushan period (Table 1). Pb at this site indicated significant pollution during the Kidarrities and Mughal periods with extremely high pollution ($I_{geo} > 4$) during the Hindu Shahi period. At Hund Pb displayed significant pollution throughout its history with very high pollution ($I_{geo} \le 4$) only during the Hindu Shahi period. Cu indicated extremely high pollution only during the Hindu Shahi period at Gor Khuttree site and lacked significant pollution at Hund (Fig. 7a).

Hg yielded elevated concentrations ($I_{geo} \le 1$) significant pollution at Gor Khuttree during the Kidarrities period and higher concentrations ($I_{geo} \ge 6$) during the Hindu Shahi period and throughout the post-Durrani periods (Table 1). Hund revealed significant Hg pollution ($1 \le I_{geo} \le 2$) until the White Huns period and higher Hg pollution ($I_{geo} > 3$) afterward.

At Gor Khuttree site, Ag yielded higher pollution ($I_{geo} > 3$) during the Achaemenid period that dropped to still very elevated pollution ($1 > I_{geo} > 2$) during the Indo-Greek period and regained higher concentrations ($I_{geo} > 3$) during the following Scytho-Parthians period. The overlying Kushan period reflected elevated concentrations ($I_{geo} > 1$) of Ag pollution and the overlying Kidarrities to White Huns periods observed a drop to significant Ag pollution (Table 1). The Hindu Shahi period again yielded higher cocentrations ($I_{geo} > 5$) of Ag that dropped to $1 > I_{geo} > 3$ Ag pollution during the Ghaznavid to Durrani periods with a drop to significant pollution ($I_{geo} > 2$) during the Mughal period. The Sikh period and onward history of Gor Khuttree reflected higher concentrations ($I_{geo} > 4$) of Ag in an increasing upscetion trend. Hund yielded higher Ag pollution ($I_{geo} > 3$) until the Kidarrities period that dropped during the White Huns period ($1 > I_{geo} > 2$). The overlying Hindu Shahi and Ghaznavid periods indicated higher concentrations of Ag ($I_{geo} > 3$) pollution. Gor Khuttree yielded higher concentrations of Au ($I_{geo} > 3$) during most parts of its history with a drop to elevated concentrations ($1 > I_{geo} > 2$) during the Kushan and Mughal periods. The Kidarrities and White periods replicated the Ag pollution trend for Au. Hund displayed a simple higher concentrations ($I_{geo} > 3$) reflecting high Au pollution trend throughout its history with a drop only during the White Huns period (Table 1, Fig. 6).

7. Discussion

7.1. Evolution of the anthopogenic influence through time

The combined results of the used geochemical proxies indicate that the Peshawar Basin experienced consistent anthropogenic influence during the past 2500 y in the distribution of various geochemical pollutants especially Ag, Au, and Hg with contributions from Pb, Mo, and Cu during multiple intervals (Table 1). During the Achaemenides period (600–400 BCE) Gor Khuttree observed elevated concentrations Ag (EF > 25), Au (EF > 25) supporting

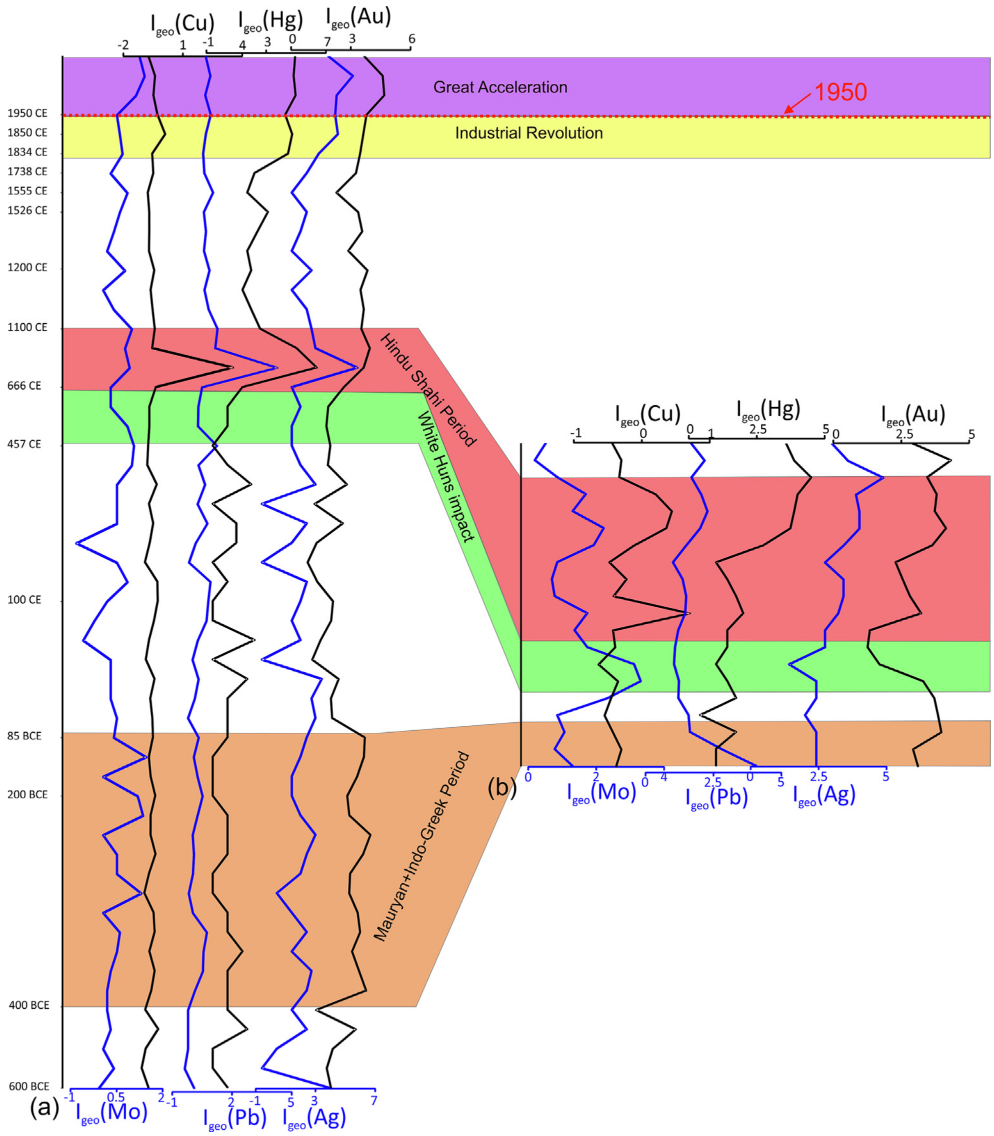

Fig. 6. Index of Geoaccumulation (I_{geo}) for the various elements at (a) Gor Khuttree site and (b) Hund site. All the important anthropogenic influence periods are highlighted and correlated.

anthropogenic pollution. During the Mauryan (400–200 BCE)-Indo-Greek (200–85 BCE) periods both Gor Khuttree and Hund observed elevated concentration of Ag (EF > 16) and Au (EF > 34) anthropogenic pollution. An interesting finding is the elevated Mo (EF > 2) pollution in both sites and also the pollution of Hg (EF > 1) and Pb (maximum value of EF > 18) at Hund during this time interval (Table 1).

This apparently provides signals for an early Anthropocene in the basin in the sense of Wagreich and Draganits (2018). During the Scytho-Parthian period (1st century BCE – 1st century CE) Au and Ag at Gor Khuttree displayed very high (EF > 11) and extremely high (EF > 22) pollution respectively. Hund was subjected to very high Ag (EF > 9) and extremely high Au (EF > 13) pollution during this time with higher concentrations of Mo (EF > 2). Pb and Hg continued the significant pollution at Hund during this time. During the Kushan period (1st century CE – 4th century CE) Ag and Au observed higher concentrations (EF > 16 for both species) at Gor Khuttree while Hund maintained trend mentioned for the previous period excluding any significant Mo pollution (Table 1).

The Kidarrities – White Huns period (457–666 CE) in the basin displayed a major drop in the anthropogenic pollution. All the Ag, Au, Hg, Pb, and Mo yielded significant pollution (EF < 8) at both sites in contrast to the higher concentrations observed during the older periods (Table 1). This correlates well to destruction during this major period of war when cities were burnt and life was destroyed especially by the invading White Huns (558–666 CE). Hwen Ts'ang, a famous Chinese traveler, described this devastation during his visit to the area in the 7th century CE (Ali et al., 2014). This is also reflected by the presence of broken brick and stone layers in the archaeological sites discussed earlier.

The Hindu Shahi period observed the most widespread and higher concentrations of anthropogenic pollution of all Ag, Au, Hg, and Pb in the two sites, and additionally Cu at Gor Khuttree (C_{deg} > 249). This period reflected the recovery from the destruction during the White Huns period in the area. This pollution trend continued during the Ghaznavid period at Hund with a slight drop in Pb pollution (EF < 5). The Ghaznavid - Durrani periods at Gor Khuttree reveal higher anthropogenic Au pollution (EF > 26) and very high Ag (EF < 11) pollution (Table 1) with the exception of the Mughal period. The Mughal period observed a significant drop in pollution (C_{deg} < 25) because the Peshawar Basin was mostly in a war-like situation during this time between the Mughals and the

Table 1
A summary of the anthropogenic pollution history of various cultural periods in the Peshawar Basin (Samad, 2011). Yellow colour represents significant values for each proxy, blue refers to very high values whereas red refers to extremely high values for each proxy. Elements responsible for these pollution values are mentioned in each case.

Historical Periods	Gor Khuttree								Hund							
	EF			C_{deg}	PLI	I_{geo}			EF			C_{deg}	PLI	I_{geo}		
Pakistan (1947 onward)	Hg		Ag Au			Mo		Hg Ag Au								
British 1849-1947 CE	Hg		Ag Au			Mo		Hg Ag Au								
Sikh 1834-1849 CE	Hg	Ag Au						Hg Ag Au								
Durrani 1738-1834 CE	Ag	Au					Ag	Au								
Mughal 1526-1738 CE	Ag	Au				Pb Ag	Au									
Suri 1535-1555 CE	Ag	Au					Ag	Au								
Saltanat 1205-1526 CE	Ag	Au					Ag	Au								
Ghaznavid 1000-1186 CE	Ag	Au					Ag	Au	Hg Pb	Ag Au				Pb		Hg Ag Au
Hindu Shahi 822-1000 CE		Cu Pb Hg Au	Ag					Cu Pb Hg Ag Au	Hg Pb	Au	Ag				Pb	Hg Ag Au
White Huns 558-666 CE	Ag Au					Ag Au			Ag Au					Pb Hg Ag Au		
Kidarrities 457-558 CE	Ag Au					Mo Pb Hg Ag Au			Ag Au					Pb Hg Ag Au		
Kushan 1st C.CE-4th C.CE	Ag Au						Ag Au		Ag Au					Pb Hg	Ag	Au
Scytho-Parthians 1st C. BCE-1st C. CE	Au	Ag					Au	Ag	Ag	Au				Pb Hg	Ag	Mo Au
Indo-Greek 185-85BCE	Ag	Au				Mo	Ag	Au	Pb Ag Au					Pb Mo Hg	Ag	Au
Mauryan 324-185BCE	Ag	Au				Mo	Ag	Au	Pb Ag Au					Pb Mo Hg	Ag	Au
Achaemenid 600-400BCE		Ag Au						Ag Au								

Pathan rulers of Afghanistan (Richards, 1995). During the Sikh period Gor Khuttree observed high Au, Ag, and Hg pollution ($C_{deg} > 63$) that continued during the following British period in an increasing upsection trend. Additionally, the British period also observed significant anthropogenic Mo contribution (EF > 3) to the system. This provides a correlation to the onset of the Industrial Revolution (Reisman, 1998) in the area. The high Au, Au, and Hg pollution ($C_{deg} > 35$) and the significant Mo pollution (EF > 3) continued during the British period and also during the post-partition modern times (after 1947). The higher concentrations of Au, Ag, Hg, and Mo ($C_{deg} > 37$) during the transition between the British period and the overlying post-partition period provides the candidate signal for the suggested mid-20th century Great Acceleration Anthropocene base (Zalasiewicz et al., 2017, 2018) in the area and can be considered as correlating to a stratigraphically defined base of the Anthropocene (Waters et al., 2018).

7.2. Controlling factors

Low anomalies (EF < 1) for As, Zn, and Cu argue against the direct mining/extraction of these elements. Historically, it is proven that there was no direct source of Cu in the area and the major Cu-mining site in the Gandhara Kingdom was at Mes-Aynak (Fig. 1), about 200 km towards the west in present-day Afghanistan (Handelsman, 2012; Dalrymple, 2013). The overall low to moderate Pb and Mo anomalies at Gor Khuttree favour the passive use of these elements and their alloys. On the other hand the strong Pb and Mo anomalies at Hund hints for the strategic and military significance of Hund throughout its history. The recent discovery of a metal processing factory during the Kushan period (work still in progress) and the latter shifting of the Gandhara capital to the site during the mid-9th century CE (Arif and Ul-Hasan, 2014) strengthen the interpretation.

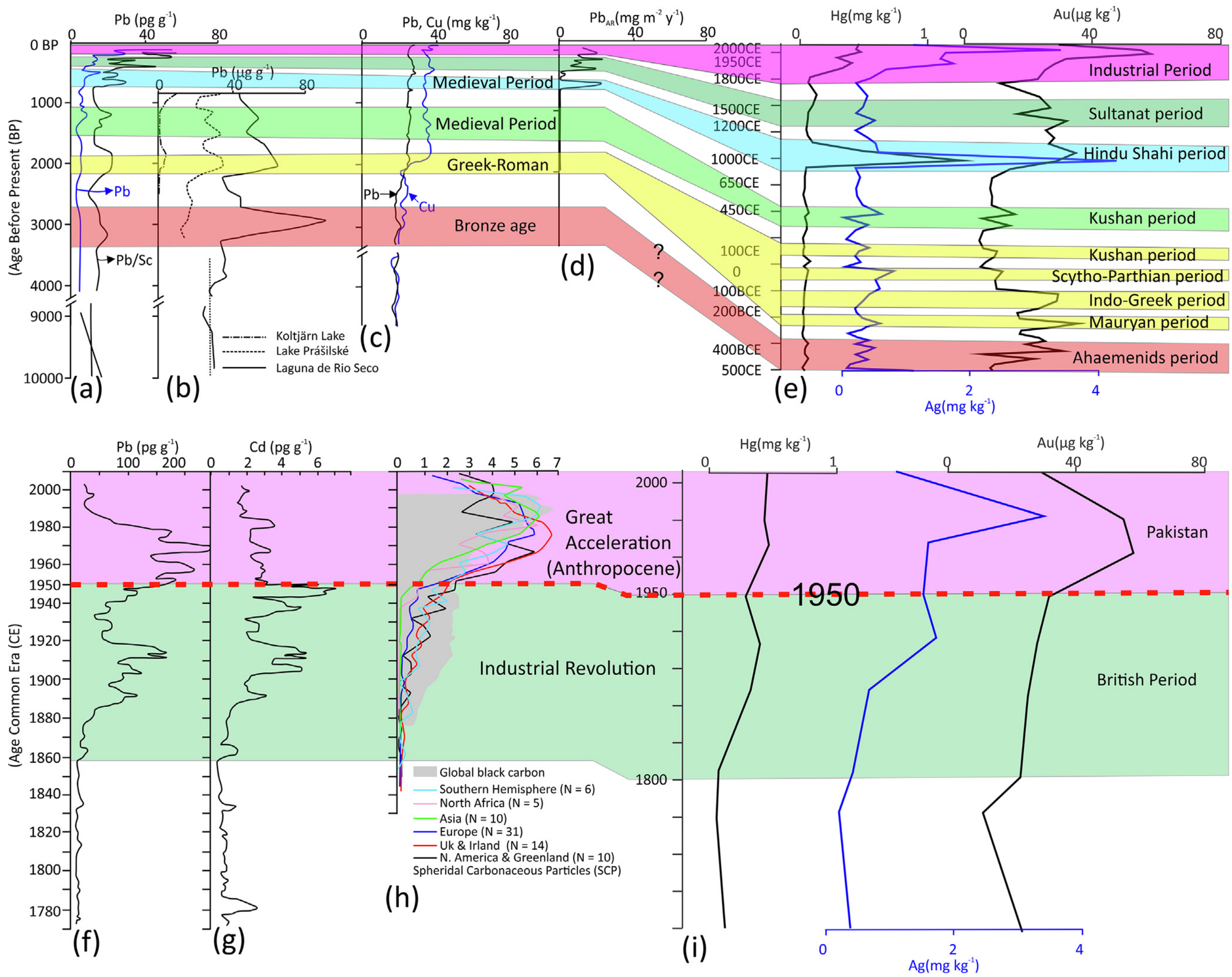

Fig. 7. Generalised correlation among various anthropogenic signals from globally studied sites and the Gor Khuttree section (Peshawar Basin). (a) Pb concentration from Arctic ice core section at Devon Island, Canada (Zheng et al., 2007), (b) Pb content from different regions and archives: solid line shows the lacustrine Laguna de Rio Seco section (dotted line marks Holocene pre-mining background value for Laguna de Rio Seco; García-Alix et al., 2013); alternating dots and dashes indicate lead contents from Lake Koltjärn, Sweden, (Bränvall et al., 2001) and dashed line from Lake Prášilské, Czech Republic, (Veselý, 2000), (c) comparison of variations of Cu and Pb in the sediments between Liangzhi Lake, China in the past 7000 years (Lee et al., 2008), (d) Pb accumulation rates (Pb_{AR}) in Puścizna Mała, Poland (Fiałkiewicz-Kozieł et al., 2018), (e) Hg, Ag and Au concentrations during various historical period in the Gor Khuttree (Peshawar), (f, g) concentrations (5-year running means) in Greenland ACT2 ice core from 1772 to 2003 CE of lead (Pb) and cadmium (Cd) respectively (McConnell and Edwards, 2008), (h) Spherical Carbonaceous Particles (SCP) sediment profile data plotted for different regions; N represents the number of lake cores is each case (Rose, 2015), (i) Hg, Ag and Au concentrations since the industrial revolution in the Gor Khuttree (Peshawar). A tentative correlation can be observed between the Late Bronze Age (LBA) anthropogenic peaks in (a-d) and the Gor Khuttree site. Note the correlation between the Greek-Roman peaks, Medieval Period and the Industrial Revolution (a-d) and the contemporaneous peaks at the Gor Khuttree (e). The anthropogenic peaks for the industrial revolution and mid-20th-century Great Acceleration show good correlation between the spikes in Greenland ACT2 ice core (f-g), global SCP record (h) and the Gor Khuttree (i).

7.2.1. Gold-Silver panning and processing

Au and Ag placers panning are historically known along the banks of Indus and Kabul rivers in the Peshawar Basin (Shah et al., 2007; Ali et al., 2015). The location of Hund on the bank of Indus River and the very high Ag and Au anomalies hint for the Ag-Au panning at the site. The very high Hg anomalies are further evidence since Hg has historically been used in such panning to separate the precious metals (De Lacerda and Salomons, 1998; Veiga et al., 2006).

The Achaemenid period at Gor Khuttree with its extremely high Au-Ag pollution, but no contribution from Hg points towards the passive use of these metals around 500 BCE. This was the peak time of the Achaemenid Empire and the area experienced a boom in trade (Briant, 2005). Peshawar was a major trade center during this time and was famous for its jewelry and handicrafts, especially Au and Ag jewelry (Samad, 2011). Hund does not go to this depth in time. At Gor Khuttree (Peshawar) continued passive use of Au and Ag until the Kushan Period as confirmed by high Au-Ag pollution but lacking contribution from Hg. On the other hand, the extremely high Au pollution at Hund during the Mauryan-Khushan Periods associated with very high Ag pollution and persistent significant Hg pollution provides a clear indication for Au-Ag panning at this site. The drastic drop in Au, Au, and Hg pollution during the Kidarrites – White Huns period in both sites points to major turnover in the anthropogenic activity in the area. This was a critical time for the area and its inhabitants when cities were burnt and people were massacred (Ali et al., 2014). This had a significant impact on the Au-Ag panning at Hund, and the jewelry and handicrafts industry at Peshawar (Gor Khuttree) as White Huns were mostly nomadic tribes having totally different priorities (Ali et al., 2014).

The following Hindu Shahi period in the Peshawar Basin was a time of recovery from the previous dark period. During this period both the Gor Khuttree and Hund sites exhibited C_{deg} of Au, Ag, Hg, Pb, and Cu above 249. This indicates a rapid increase in the anthropogenic deterioration of the local environment via Au-Ag panning at Hund and Au-Ag processing at Gor Khuttree. The Cu and Pb peaks also hint for the use of metal alloys as perhaps the Hindu Shahi rulers now wanted to be better prepared for the nomadic invaders. The shifting of the capital from Peshawar to Hund during the mid-9th century CE strengthens this interpretation (Sehrai, 1979). Hund continued the same pollution trend during the Ghaznavid period and the archaeological record of Hund excavation site concludes here (Table 1; Fig. 2).

At Gor Khuttree the processing of Au-Ag and the jewelry and handicrafts markets continued to flourish during the Ghaznavid to Suri periods. The location of this site away from both the Kabul and Indus rivers favors this assumption, while no significant Ag-Au anomalies have so far been reported from the Bara River, the closest stream to the site. The historical location of Peshawar along the Khyber Pass, as the first major stop on the entrance to the Indian subcontinent on the famous silk route, presents the site as major trading center (Docherty, 2008). The presence of the still active storyteller market (Qissa Khwani bazaar) indicates that traders used to rest at this site and discuss their stories at the very place (Prasad, 1977). The presence of gold market (Sarafa Bazaar) and copper market (Misgaraan Bazaar) that have been mentioned in historical records for thousands of years (Prasad, 1977) are clear evidence for Cu, Ag, and Au processing in the area. The drop in pollution at Gor Khuttree during the Mughal period has already been discussed. The extremely high Hg contamination in the area following the first quarter of the 18th century and the parallel Ag-Au contamination provide clear signals for the Industrial Revolution in the area. Here the base of the Sikh period is considered as the onset of the Industrial revolution in the area (Fig. 7). The continuous addition of significant Mo and extremely high Hg, Au, and Ag to the local environments during the mid-20th century (top of British period and lower part of post-partition period) can be considered as candidate for the Great Acceleration, and thus, the base of the Anthropocene in the area (Fig. 7).

7.3. Regional and global correlation and the Anthropocene concept

As the anthropogenic signals are linked to the local human cultures, therefore, their correlation often has two major limitations: (1) chronology of such signals is often diachronous and (2) the chemical species vary from culture to culture both in their nature and intensity. However, the search for early anthropogenic influence on the Earth's system from widespread localities has provided evidence for regional and global correlation of the anthropogenic signals.

7.3.1. Late Bronze age to early Iron age signals

During the Late Bronze Age (LBA) anthropogenic activity of mining and smelting in the Old World peaked (c. 3600–3200 BP) with Cyprus producing large Cu amounts that continued into the Iron Age (Kassianidou, 2013). A noticeable increase in Pb pollution occurred roughly during the same time (e.g., Zheng et al., 2007; Wagreich and Draganits, 2018 and references therein). The associated smelting activities produced anthropogenic signals which can be observed from Pb concentration and Pb/Sc ratio in the Arctic ice core section at Devon Island, Canada (Fig. 7a) due to mining and smelting in the Iberian Peninsula (Zheng et al., 2007; Krachler et al., 2009). Similar Pb spikes have also been recorded from Greenland ice cores (Hong et al., 1996).

Similarly Pb concentration records from different regions and archives including the lacustrine Laguna de Rio Seco section of Spain (García-Alix et al., (2013), Lake Koltjärn of Sweden (Bränvall et al., 2001) and Lake Prášilské of Czech Republic (Veselý, 2000) provided early anthropogenic signals (Fig. 7b) that can be correlated with the Arctic data (Fig. 7a). Furthermore, Cu and Pb data in the sediments of Liangzhi Lake China indicate a nearly contemporaneous anthropogenic distribution of these elements during the Xia Dynasty and the Han Dynasty (Lee et al., 2008).

Since the oldest archaeological evidences in the Peshawar Basin are much younger (~600B CE; Ali et al., 2005) than the famous Indus Valley Civilization (IVC) of Pakistan (3300–1300 BCE; Wright, 2009) therefore, correlation with the IVC is uncertain. However, the Ag, Au, and Pb spikes in the lowermost part at the Gor Khuttree provide a tentative correlation between the Achaemenid period and the lower part of Mauryan period of the Peshawar Basin and the above-mentioned peaks (Fig. 7a-e).

7.3.2. Greek to Roman mining peak

The Greek, Hellenistic, and Roman periods were times of large scale (proto-industrial) metals production unprecedented before (e.g., Borsos et al., 2003; Wagreich and Draganits, 2018). Major Pb spikes in all the above mentioned localities (Arctic ice core section at Devon Island, Canada, lacustrine Laguna de Rio Seco section of Spain, Lake Koltjärn of Sweden, Lake Prášilské of Czech Republic, and Pb and Cu in the sediments of Liangzhi Lake China) indicate simultaneous anthropogenic addition of Pb to the Earth's system peaking at around 2000 BP (Fig. 7a–e). These spikes correlate to Pb production (a first maximum 80,000 tons/year according to Hong et al., 1994) during the golden period of the Roman Empire (Wagreich and Draganits, 2018). Similarly, the Ag mines around Rio Tinto were intensively exploited in Roman times (Rosman et al., 1997).

Peshawar Basin provides multiple spikes of Mo, Pb, Ag, and Au during the contemporaneous periods (Figs. 1–3; 7) During this time the Gandhara area observed interference from the west especially the Greeks (c. 400–200 BCE) before the onset of the major Indo-Greek period (c. 196–85 BCE). Thus the Peshawar Basin provides early anthropogenic pollution peaks of Au and Ag that can be correlated to the Pb peaks widely reported during the Roman times (Fig. 7) and also provide strong Mo and Pb enrichment during this time (Figs. 2 and 3). The same geochemical signals have been used as potential stratigraphic signals for an early Anthropocene (Wagreich and Draganits, 2018).

7.3.3. Medieval period

The Medieval period (c. 500–1500 BP) yields multiple anthropogenic signals (Fig. 7a–d). The earliest major Medieval period Pb concentration is observed in Arctic ice core section at Devon Island, Canada around 1500–1600 BP (Fig. 7a) and can be correlated to the lacustrine Laguna de Rio Seco section of Spain, Lake Koltjärn of Sweden and Lake Prášilské of Czech Republic (Fig. 7b) and Pb and Cu in the sediments of Liangzhi Lake China (Fig. 7c). Similar anthropogenic Hg, Ag, and Au signals during the late Kushan period (c. 450 CE) provide correlation with mentioned global signals (Fig. 7e).

One of the interesting findings of the Medieval period (c. 9th Century CE) is the very high Pb accumulation rates (Pb_{AR}) in peat bogs in Puścizna Mała, Poland (Fig. 7c). This peak can also be found in the other sites used in this correlation (Fig. 7a–d). This was the time of Mississippi Valley Type Pb–Zn type deposits mining and processing (Fiałkiewicz-Kozieł et al., 2018) with significant deposits in Europe located in the Silesia–Cracow Uplands, Poland. Galena mining and Ag extraction in the area dates back to Medieval period i.e., at least the first half of the 12th century CE (Molenda, 1963) with latest results even dating back the exploitation inception to the 9th century CE (Fiałkiewicz-Kozieł et al., 2018). A similar Pb rise in the Liangzhi Lake sediments, China occurred during the Holocene Optimum, Medieval Warm Period (c. 9th century CE to

12th Century CE) (Jin et al., 2013). This Pb Peak correlates very well to the most prominent Cu, Pb, Hg, Ag, and Au peaks observed in the Peshawar Basin during the Hindu Shahid period (Figs. 2 and 3; 7a–e).

7.3.4. Industrial period

Since the start of the 19th century, the Arctic ice core section at Devon Island, Canada, Pb accumulation rates (Pb_{AR}) in peat bogs in Puścizna Mała, Poland and Pb and Cu in the sediments of Liangzhi Lake China yield clear anthropogenic signals of Pb distribution worldwide (Fig. 7a, c and d). The entire post-Durrani period history of Gor Khuttree displayed higher anthropogenic contribution ($C_{deg} > 35$) of all the discussed elements especially Hg, Ag, and Au (Fig. 7e). Thus the Peshawar Basin provided clear anthropogenic signals for regional and global correlation during the industrial revolution. This is explained in detail in the following section.

7.4. The Great Acceleration

The Great Acceleration, the extreme growth of human influence after the Second World War (Steffen et al., 2018) is archived in the rising anthropogenic signals since the mid-20th and the associated impact on the Earth's System as a whole (1950 CE) is suggested as the chronostratigraphic base of a formalised Anthropocene (Waters et al., 2016). The anthropogenic input of the Spheroidal Carbonaceous Fly Ash Particles (SCP) became noticeable in the second half of 19th century and intensified globally following the 1950 CE (Fig. 7h) (Rose, 2015; Waters et al., 2016). Pb (Fig. 7f) and Cd (Fig. 7g) concentrations in Greenland ACT2 ice core (McConnell and Edwards, 2008) yield inception in the rise of the two species around the same time when the SCP started to do so (c. 1860 CE). At around 1950 CE Pb signals started to gain significant strength while Cd started to drop (Fig. 7f and g). Therefore, 1950 CE is globally considered as the start of the "Great Acceleration" and is presented as a potential start date of the Anthropocene (e.g., Waters et al., 2018).

Gor Khuttree provides a noticeable gain in the anthropogenic signals since the start of the 19th century (Fig. 7i). This indicates the beginning of the Industrial Revolution in the Peshawar Basin and correlates well with the global signals (Fig. 7g-i). During the latest part of the British period and the early post-partition time (c. 1950 CE) the basin yields extremely high anthropogenic contamination signals for various geochemical species and especially the Hg, Ag, and Au data have been used for global correlation (Fig. 7i). The Peshawar Basin verifies the onset of the "Great Acceleration" during 1950 CE that continued until the later part of the 20th century, followed by a drop during around 2000 CE. Thus the geochemical data from Peshawar Basin favour the base of the "Great Acceleration" as the base of the Anthropocene.

8. Conclusions

Archaeological records from the Peshawar Basin indicate the existence of a well-developed human civilization at least since 500 BCE. Geochemical proxies provide multifold evidences supporting anthropogenic control on the distribution of trace metals. As and Zn do not indicate significant anthropogenic dispersion of these elements while Cu indicates large scale anthropogenic input only during the Hindu Shahi period. Pb and Mo data support anthropogenic influence in their distribution throughout the history of Hund while Gor Khuttree supports extensive anthropogenic dispersion of Pb during the Hindu Shahi period. Hg, Ag, and Au data present Hund as a major site for panning of precious metals while Gor Khuttree supports processing of Ag and Au. Thus, the Peshawar Basin provides Ag-Au panning as evidence for the human impact on local environment since at least 500 BCE.

Geochemical signals from the Peshawar Basin can be considered as conservative for correlation with the Late Bronze Age anthropogenic activity because the present archaeological record is too young and deeper excavation at Gor Khuttree may provide further evidences in this regard. Geochemical proxies from sediments of the Mauryan period to Scytho-Parthians period at Gor Khuttree provide early anthropogenic signals from the basin for correlation with the classical Greek and Roman mining and smelting periods. The prominent anthropogenic signals from the Peshawar Basin during the Hindu Shahi period correlate to the signals from peat bogs for Medieval period mining and smelting in Europe and Medieval warming period observed in lake sediments of China. Gor Khuttree provides strong evidences for anthropogenic signals during the Industrial period that can be correlated globally and presents the mid-20th century time (the base of the Great Acceleration) as a potential base of the Anthropocene based on geochemistry markers.

Declaration of Competing Interest

It is hereby stated that there is no conflict of any kind in the present research work. All the data sources have been clearly mentioned and the funding sources have been acknowledged.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.134926.

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