








# The geochemical study of fluvio-lacustrine aquifers in the Kathmandu Basin (Nepal) and the implications for the mobilization of arsenic

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**Abstract** The geochemical analyses of fluvio-lacustrine aquifer sediments of the Kathmandu Valley have been made as a step in assessing the environment for the mobilization of arsenic in groundwater. Elements measured by X-ray fluorescence (XRF) include 4 major oxides ( $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$ ,  $\text{CaO}$ ,  $\text{P}_2\text{O}_5$ ) and 14 trace elements (As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, Sc and TS). Elution tests of 15 selected core samples were also carried out to determine the potential leaching of arsenic from the sediments. The XRF results show that average bulk concentrations of the major oxides and trace elements are similar to modern unconsolidated sediments and average upper continental crust. However, the abundance of elements varies with grain size, with higher concentrations in finer-size grades. Variations in elemental abundances within the basin are strongly controlled by sediment facies. The elution tests show that greater amounts of arsenic are generally eluted from the fine sediments, although the rates are variable. The results overall suggest that As concentrations in the bulk sediments are not a controlling factor for elevated As in the

Kathmandu Basin groundwater, and the roles of other factors such as redox conditions and organic matter contents are likely to be more significant.


**Keywords** Kathmandu Basin  Arsenic   
Groundwater  Sediment geochemistry   
Reducing condition

## Introduction

Arsenic contamination of drinking water is a worldwide problem due to its detrimental effects on health. These effects range from skin ailments through to serious diseases such as cancer, and to death. Arsenic contamination of groundwater has been reported from many countries including Bangladesh, West Bengal, India, Vietnam, Argentina, China, parts of the USA (Smedley and Kinniburgh 2002; Hossain 2006) and now Nepal. The provisional limit for arsenic (As) in drinking water as recommended by the World Health Organization (WHO) is 10  $\mu\text{g/L}$ . Nepal has set its interim guideline at the higher value of 50  $\mu\text{g/L}$ , the level also adopted in Bangladesh.

Arsenic contamination of the groundwater in the Bengal delta (Bangladesh and West Bengal, India) is the most severe in the world. The health of more than 85 million people is under threat in Bangladesh alone (Hossain 2006). The study of arsenic concentrations in the groundwater in Nepal began only after the severity of the arsenic contamination problem in the Bengal delta was recognized in 1998. The first report of arsenic contamination in groundwater above toxic levels in Nepal was made from the Terai Basin (Sharma 1999). Twenty-four percent of samples analyzed ( $n = 18,635$ )

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from the Terai Basin exceeded the WHO limit of 10 l g/L (Shrestha and Shrestha 2004).

The Terai Basin is the southern plain area of Nepal. The Terai Plain is remarkably flat, with an average altitude of 115 m above mean sea level, and stretches from east to west along the southern border with India. Groundwater is the main source of water for drinking and irrigation, with extraction primarily from streams, ponds, hand-dug wells and more recently from tube wells. Geologically, the Terai Plain is an active foreland basin filled with Quaternary sediments including molasse sediments, gravels, sands, silts and clays. The Terai Basin lies in the proximal part of the Ganges watershed, whereas Bangladesh is in the distal part.

The Kathmandu Valley of Nepal is an intermontane basin located in the central Nepal Himalaya (Fig. 1). This valley is elongated east to west, and has an area of about 650 km<sup>2</sup>. The central part of the valley has more or less flat topography, with an elevation of 1,300–1,400 m, and is surrounded by mountains with elevations exceeding 2,000 m. Drainage systems flow toward the valley center, and collect in the Bagmati River. The Bagmati River exits the basin through the Mahabharat Range to the south.

About 40% of the 1.5 million population of the Kathmandu Valley relies on groundwater for drinking, industrial use and irrigation. However, the deterioration in groundwater quality is now a major concern. Earlier studies have indicated that groundwater quality in the valley is degrading. The major contaminants of the hand-dug wells, hand pumps and spouts are elevated nitrate (>50 mg/L of N), ammonia (>15 mg/L) and high *E. coli* counts (2,500 mL<sup>-1</sup>). Deep wells are contaminated with ammonia (up to 119 mg/L), iron (>10 mg/L), manganese (1.17 mg/L) and phosphate (up to 22 mg/L) (Jha et al. 1997). Dissolved organic carbon concentrations are generally high in the deep wells (up to 63 mg/L), particularly so in the central part of the valley (Khatiwada et al. 2002).

Recent reports of high arsenic concentrations in Kathmandu groundwater (JICA/ENPHO 2005) have created considerable anxiety among users. Little attention was paid to the arsenic issue in the Kathmandu Valley until elevated arsenic levels were identified in the groundwater in the Terai Basin in 1999. Earlier studies of groundwater quality in the Kathmandu Valley excluded arsenic from both analysis and discussion. The Kathmandu Valley contains Quaternary sediments similar to those occurring in the Terai Basin, and thus the presence of arsenic in the groundwater can be suspected. A study of Kathmandu groundwater quality by Jha et al. (1997) did examine arsenic concentrations, and found that the contents

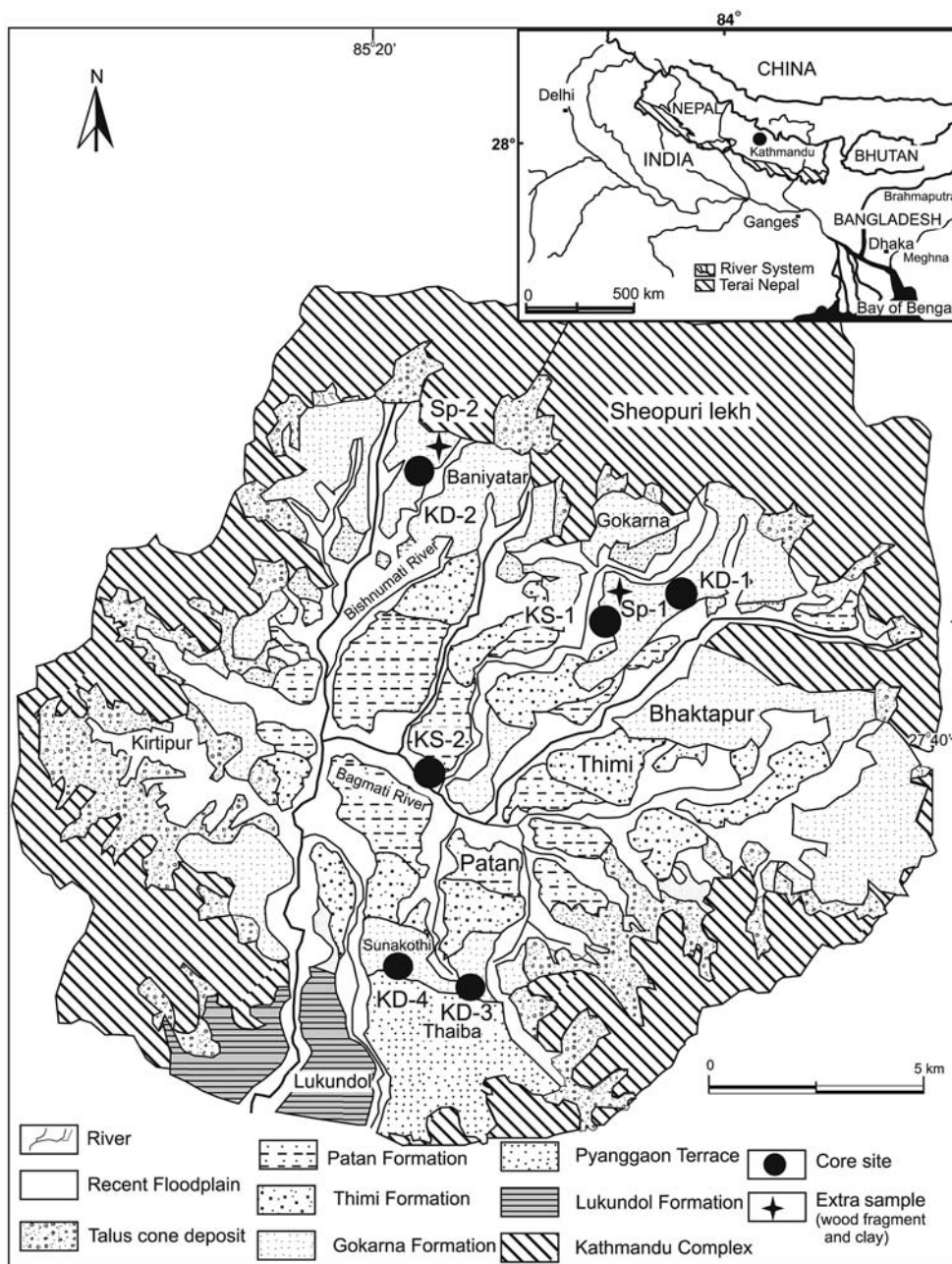
were within the Nepal interim national guideline value (50 l g/L). However, Jha et al. (1997) explicitly pointed out the need for the study of sediment geochemistry, and for regular monitoring for As concentrations in the Kathmandu Basin groundwaters. A later study by Amaya (2002) found that the As contents of a few samples exceeded the national guideline value. JICA/ENPHO (2005) reported that the Kathmandu groundwater is highly vulnerable to arsenic contamination, particularly in the deeper aquifers (>200 m) of the central part of the valley, where As contents of several samples exceeded 200 g/L. The studies made to date are based on the quantitative analyses of As concentrations in the groundwaters themselves, and do not provide any information on the release mechanisms involved or their controlling factors.

This study examines the bulk geochemistry of the Kathmandu aquifer sediments, the elution behavior of As in representative samples, and discusses the potential mechanisms causing mobilization of arsenic in groundwater. Bulk concentrations of elements including arsenic are not exceptional, and are similar to the general levels seen in modern unconsolidated sediments. The elution tests show that the amount eluted is not strictly related to the bulk concentration, although generally greater amounts of arsenic are leached from clay-rich samples compared to coarser core samples.

## Geology and hydrogeology

The Kathmandu Valley is a tectonically controlled basin filled with Quaternary fluvio-lacustrine sediments >600-m thick. The basal part of the basin-fill sediments consists of lower Pleistocene lacustrine clays and gravels of the Lukundol Formation (Yoshida and Igarashi 1984), which is stratigraphically equivalent to the Bagmati Formation (Sakai 2001a Fig. 2). The Lukundol Formation is overlain by a lignite member (Dangol 1985), which is in turn succeeded by thick black lacustrine clay unit locally known as the Kalimati Clay (Sakai 2001a). The Kalimati Clay is rich in organic matter, diatoms, plant fossils and natural gases (Fujii and Sakai 2001; JICA 1990). The uppermost basin-fill sediments are covered by extensive fluvio-lacustrine terraces and fan deposits (dominantly sands and gravels) of the Itahiti, Gokarna, Patan and Thimi Formations (Yoshida and Igarashi 1984; Sakai 2001a). The ages of these terrace deposits range from 40,000 to 11,000 years BP (Igarashi et al. 1988; Yoshida and Gautam 1988). The coarse sediments in the northern part of the valley represent delta deposits and facies that are influenced by the processes of delta progra-

Fig. 1 Geological map of Kathmandu Basin, Nepal (after Yoshida and Igarashi 1984), and core sample locations



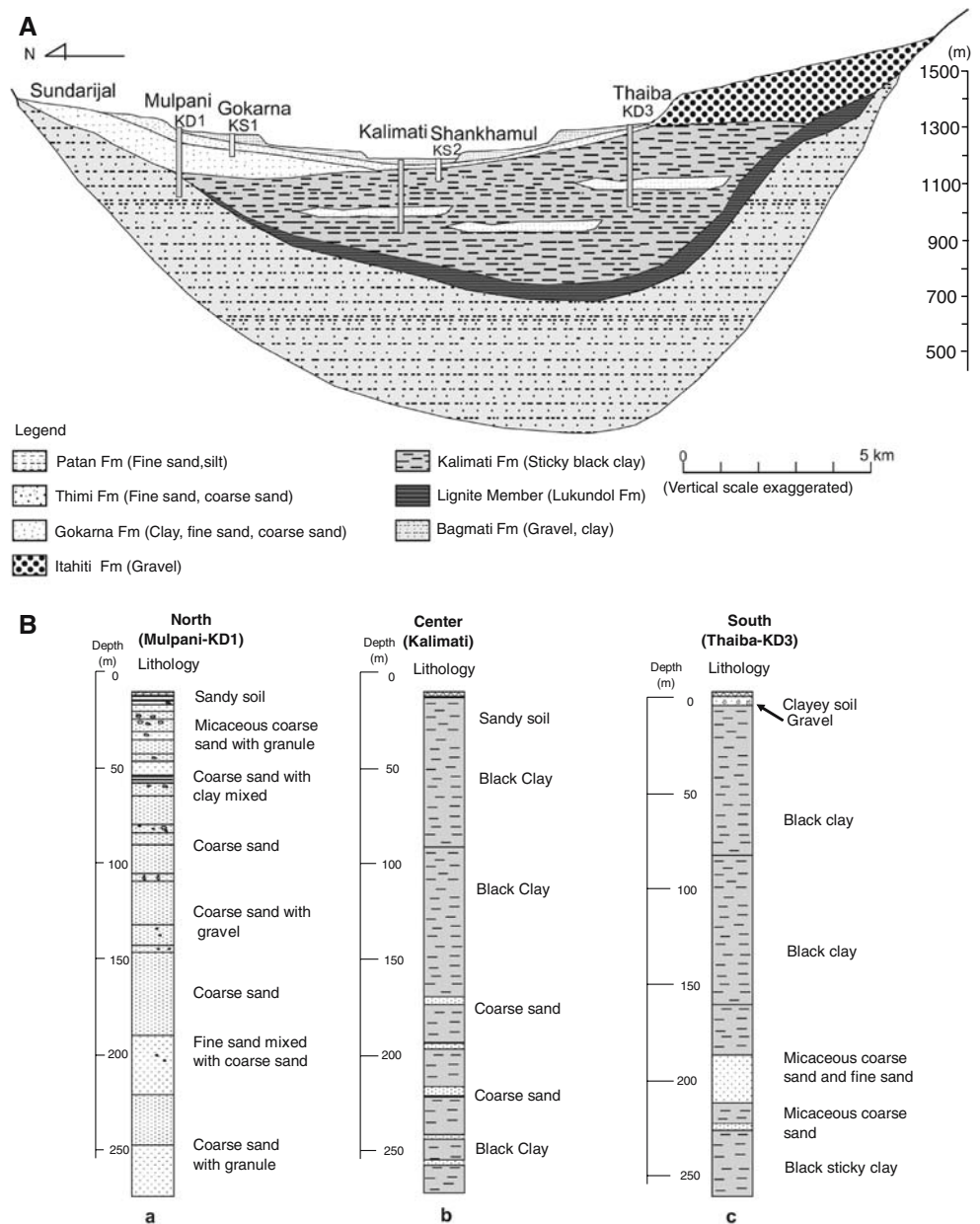
ation and paleo-lake fluctuation (Sakai 2001b). A geological map of the Kathmandu Valley and a schematic cross-section are shown in Figs. 1 and 2a, respectively.

Average annual rainfall in the Kathmandu Basin is around 2,000 mm, about 80% of which falls in the monsoon period during June and July. Surface runoff is high during the monsoon, with the estimated monthly discharge of the Bagmati River being  $15.81 \text{ m}^3 \text{ s}^{-1}$  at the Chobhar gorge; the rate of evaporation is 3.70 mm/day (JICA 1990). Recharge to the shallow aquifers (<60 m) occurs mostly along the basin margins, directly

from precipitation and by supply from a number of small rivers. However, recharge to the deeper aquifers is considered to be limited, due to the presence of clay beds that significantly restrict downward percolation. Because the Kathmandu Valley is a closed basin with gentle slopes toward the center, groundwater flow is assumed to be slow, particularly in the deeper aquifers.

By convention, the aquifers in the Kathmandu Basin can be divided into shallow and deep systems. Shallow aquifers typically extend from less than 5 to 60 m, and deeper aquifers lie below 60 m. Groundwater from the shallow aquifers is drawn from hand-dug wells, hand

Fig. 2 A Schematic cross-section of the Kathmandu Valley (after Sakai et al. 2001). B Columnar sections of the boreholes. a Mulpani (North), b Kalimati (Center) and c Thaiba (South), Kathmandu Basin, Nepal



pumps or roar pumps, whereas the deeper aquifers are exploited from deep wells. Traditional stone spouts (locally known as Dhungedhara) are also common, drawing water from shallow aquifers. Groundwater from both the shallow and deeper aquifers has been used extensively for drinking and industrial purposes. The static water table generally lies within a meter of the ground surface, but in some areas in the northern part of the valley it may lie as deep as 60 m (Kharel et al. 1998). Hand-dug wells and stone spouts in the central and southern parts of the valley are mostly dry in the dry season (April–May) due to the lowering of the water table.

## Materials and methods

### Core samples

Core samples were collected from the northern, central and southern parts of the Kathmandu Valley. In the northern part, samples were collected at Mulpani (KD1) and Baniyatar (KD2) from two machine-drilled boreholes (deep-well construction sites), both of which penetrated 250 m below surface (Fig. 1). The samples consist of poorly sorted, fine- to coarse-grained sands and gravels of the Gokarna Formation (Igarashi et al. 1988). The gravel clasts and sand grains are angular to

subangular in shape. Thin layers of micaceous clays are occasionally interbedded within the sands and gravels. Detailed lithology of borehole KD1 is shown in Fig. 2a. Core samples from shallow depth (8 m) were also collected from a site near Gokarna (KS1, Figs. 1, 3) by the manual auger method. These samples consisted of fine to coarse sands and silty clays. Two additional samples were also collected from the surface outcrop. These were a silty clay (Sp1) from a sand quarry near the Gokarna Forest, and a wood fragment (Sp2) from a fine sand bed at Baniyatar (Fig. 1).

The central valley samples were collected at Shankhamul (KS2, Figs 1, 3), by the same manual auger method. This 8-m core penetrated 5-m thick sandy beds at the top, and 3 m of underlying massive clay (Patan Formation, Igarashi et al. 1988). The core samples were collected from the southern part of the valley at Thaiba (KD3) and Sunakothi (KD4) (Fig. 1), and also from deep-well drilling sites. The lithology at both locations (KD3 and KD4) comprised thin sand and gravel beds of the Gokarna Formation at the top, underlain by thick underlying lacustrine clay beds of the Kalimati Clay (Sakai 2001a). The gravel beds are

muddy and matrix-supported. The sand beds are micaceous, similar to those in the northern part of the valley. The lithology of a borehole KD3 at Thaiba is given in Fig. 2c.

#### Laboratory analysis

##### X-ray fluorescence analysis

Fifty grams of each sample were dried in an oven at 110°C for 24 h and then crushed in an agate mortar and pestle for 20 min to ensure particle size of < 63 µm. The powdered samples were compressed into briquettes using a force of 200 kN for 60 s. The briquettes were then analyzed for total iron (Fe<sub>2</sub>O<sub>3</sub>), TiO<sub>2</sub>, CaO, P<sub>2</sub>O<sub>5</sub>, total sulfur (TS) and selected trace elements (As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th and Sc) using a RIX-2000 XRF spectrometer (Rigaku Denki Co. Ltd) at Shimane University. The concentrations were determined using the pressed powder method of Ogasawara (1987). The average error for these elements is less than ± 10%.

##### Elution analysis

Elution analysis is a useful method for determining the potential leaching of arsenic from the aquifer sediments. The pH of the Kathmandu Valley groundwater is almost neutral, commonly in the range 6.5–7.4 (Amaya 2002, JICA/ENPHO 2005). Consequently, elution tests were carried out at neutral pH on 15 representative samples (both fine and coarse grained) to determine the potential leaching of bulk arsenic. Because there is no standard elution technique for arsenic in Nepal, the procedure of the Japanese Industrial Standard (JIS K0102 61) was adopted for the evaluation of sediment dissolution and its potential effect on the potable groundwater. Fifty grams of each sample were treated with 500 mL of deionized water (pH 7), and agitated for about 6 h in an electric shaker (200 rpm) at room temperature (20°C). The samples were centrifuged at 3,000 rpm for 30 min, and then the supernatant liquid thus obtained was passed through a 0.45 µm millipore filter. The arsenic contents of the solutions were then determined at Shimane University, using an atomic absorption spectrophotometer (AAS, Shimadzu AA-660G) equipped with a graphite furnace atomizer (GFA-4B). Calibration of the AAS was made using a blank solution (1 N HNO<sub>3</sub>, 0.01 ml/L) and standard arsenic solution (1,000 µg/L). The detection limit of the AAS used was < 1 µg/L.

The amount eluted was corrected for the volume of solution to derive rock mass equivalent (rme) values

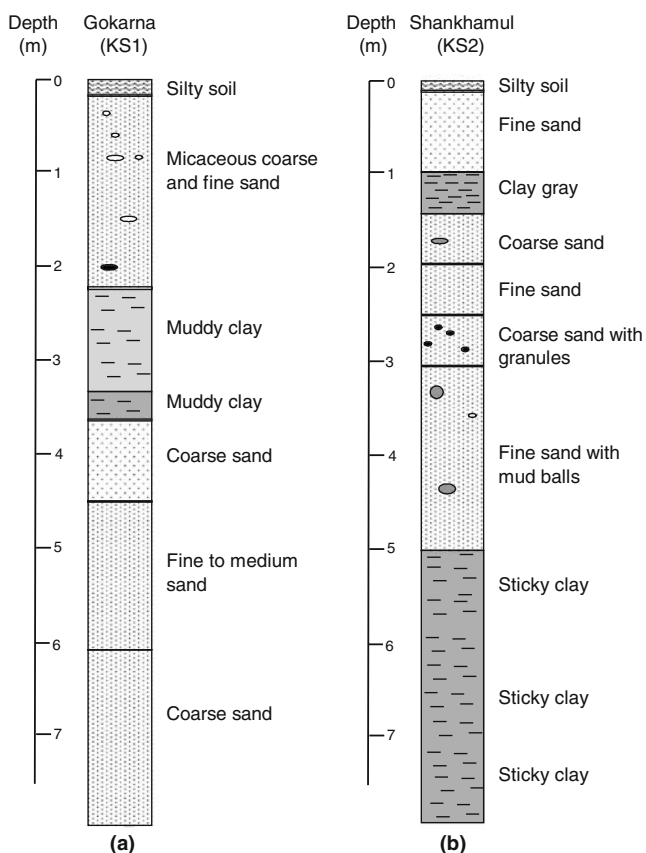


Fig. 3 Columnar sections of shallow boreholes: a Gokarna (KS1); b Shankhamul (KS2), Kathmandu Basin, Nepal

(eluted vol.  $\times$  50 mL/500 mL). Elution rate (ER) was calculated using the formula given by the Association for Environmental Measurement and Analysis in Japan (1995):  $ER = (C_{eluted}/C_{bulk}) \times 100\%$ , where  $C_{eluted}$  is the eluted concentration, and  $C_{bulk}$  is the bulk concentration in the sediments.

## Results

### Sediment chemistry

Data for the major oxides analyzed ( $Fe_2O_3$ ,  $TiO_2$ , CaO and  $P_2O_5$ ) and trace elements (As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, Sc and TS) are listed (Tables 1, 2, 3; northern, central and southern sectors, respectively). The grain size of the core samples ranges from silty clay to gravels, and the bulk concentrations of the oxides and trace elements vary considerably. The overall concentrations of most elements increase toward the center from the northern margin and diminish slightly in the southern margin of the valley, except for Pb, Sr and Zr. Pb and Sr, which decrease southward, whereas Zr increases slightly from the north to the south. The variations of median concentrations and ranges of  $Fe_2O_3$ , As, TS,  $P_2O_5$ , Zn and V are illustrated in Fig. 4. However, the distributions of elements do not vary systematically with depth.

Sample locations in the northern part of the valley (KD1, KD2 and KS1) lies at the basin margin, and thus consist mostly of coarse fluvial sediments. Bulk concentrations of the elements are not uniform among the samples (Table 1). In coarser samples such as gravel and coarse sand, abundances of major oxides and trace elements are less than the detection limit. The total iron content ( $Fe_2O_3$ ) of the sand and gravel is low ( $< 2$  wt%), whereas it is high in silty clay ( $>6$  wt%).  $TiO_2$  abundances in coarse sand and gravel seldom exceed the detection limit of 0.05 wt%, but are greater in finer samples such as silty clay ( $>0.5$  wt%). The CaO content is generally low ( $<1$  wt%), except for one silty clay sample (KD2-3, 12 wt%).  $P_2O_5$  abundances in four samples from Mulpani (KD1D9 to KD1D12) and three from Baniyatar (KD2D7, KD2D9 and KD2D10) are anomalously high ( $>0.5$  wt%). In all the other samples, the contents are more normal ( $<0.30$  wt%), comparable with upper continental crust (UCC) values (0.17 wt%). Arsenic averaged 6 mg/kg, ranging from 4 to 12 mg/kg, whereas Pb averaged 42 mg/kg, ranging from 19 to 69 mg/kg. The Zn content varies widely with grain size. In most coarse sand and gravel, Zn abundances are  $< 10$  mg/kg. In the finer-grained samples such as silty clay, the contents are considerably

greater (up to 94 mg/kg); except for one sample (KD2D17), which contained less than 5 mg/kg. Similarly, the V content in most coarse sand and gravel is low ( $<2$  mg/kg) and is greater in the finer samples ( $>100$  mg/kg). Other trace elements including Cu, Ni, Nb, Sr, Th and Sc follow similar patterns, with low concentrations in the coarser samples. Cr and Y values are more uniform relative to the other trace elements, thus indicating less effect of grain size. The Zr content in two silty clay samples (KS1D3 and Sp1) are considerably greater (274 and 345 mg/kg, respectively) than the average value (102 mg/kg). Concentrations of TS reach 0.28 wt% in the silty clay, but seldom exceeds 0.02 wt% in the coarse sand and gravel. The wood fragment analyzed (Sp2) had an exceptional As content (218 mg/kg), along with elevated TS (2.6 wt%), V (1,211 mg/kg) and Cr (160 mg/kg), as shown in Table 1.

The shallow core samples from the central part of the Kathmandu Valley penetrated lacustrine clay and sand. The overall bulk elemental concentrations are greater relative to the samples from the northern part; however, the concentration variations among the samples are more uniform. The total iron content in the black sticky clay is high ( $>7$  wt%), and average values of  $TiO_2$  (0.6 wt%) and TS (0.06 wt%) increase from those in the northern part of the valley. CaO values remain about the same (1.1 wt%); whereas average  $P_2O_5$  (0.17 wt%) decrease slightly from the north (0.21 wt%). Arsenic content also increases from the north (Fig. 4b), averaging 11 mg/kg, ranging from 4 to 25 mg/kg. The Zr content of the two samples (KS2D4 and KS2D7) are also unusually high, 227 and 246 mg/kg, respectively, compared to the average value of 156 mg/kg for this element.

Elemental distributions in the samples from the southern part of the valley (KD3 and KD4) follow a similar pattern to those in the northern and central parts. However, the average bulk concentrations of most elements decrease slightly with respect to the central part, as shown by the examples in Fig. 4; an exception is Zr, which averages 263 mg/kg in the south, compared to 102 mg/kg and 156 mg/kg in the northern and central parts of the valley, respectively.

The overall abundances of Fe, Cu, Zn, Ni, Cr, V in both the marginal and central parts are clay-controlled. High Fe concentrations in the finer-grained sediments ( $>7$  wt%) could also result from authigenic phases or enrichment during early diagenetic processes (Dill and Melcher 2004). The decreasing concentration of Pb and Sr from the valley margin in the north toward the center suggests their association with feldspar minerals that undergo dissolution easily. The higher Zr content

Table 1 Major and trace elements analyses, northern Kathmandu Basin, Nepal

Sample ID	Lithology	Depth (m)	Major oxides (wt%)				TS (wt%)	Trace element (mg/kg)												
			Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	CaO	P <sub>2</sub> O <sub>5</sub>		As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc
Gokarna (KS1)																				
KS1D1	CS, FS	1	0.85	0.12	0.73	0.08	0.02	6	41	12	<2	8	38	<2	83	23	4	80	4	5
KS1D2	C	3	7.45	0.91	0.84	0.06	0.05	7	31	89	26	50	90	158	75	44	23	158	13	15
KS1D3	C	3.6	7.03	0.88	0.99	0.06	0.06	7	31	90	23	44	83	151	84	48	24	274	17	14
KS1D4	CS	7.5	1.94	0.28	0.78	0.06	0.02	5	41	23	3	11	51	25	89	29	8	95	6	8
Sp1	SC	20	6.43	0.78	1.07	0.07	0.07	8	35	84	21	37	71	121	96	53	21	345	24	13
Mulpani (KD1)																				
KD1D1	SC	0.5	3.90	0.51	1.01	0.09	0.03	5	35	44	10	24	49	63	104	35	12	173	12	9
KD1D2	CS	2	1.12	0.16	0.72	0.06	0.02	6	42	13	4	9	28	2	90	26	5	85	5	3
KD1D3	CS	4	1.14	0.15	0.72	0.06	0.02	5	40	12	2	10	36	4	85	25	6	90	6	3
KD1D4	CS	7	0.43	0.06	0.70	0.06	0.02	6	46	7	<2	7	32	<2	94	22	5	77	3	2
KD1D5	CS	11	<0.5	0.06	0.74	0.05	0.02	6	46	8	2	6	33	<2	95	23	3	73	3	1
KD1D6	CS	18	<0.5	<0.05	0.68	0.09	0.02	8	69	<5	2	3	25	<2	136	29	2	48	2	<1
KD1D7	CS	22	<0.5	0.05	0.72	0.06	0.02	7	49	<5	2	7	37	<2	102	22	3	58	2	1
KD1D8	CS	26	<0.5	0.02	0.69	0.05	0.02	6	53	<5	<2	6	32	<2	104	23	3	63	3	1
KD1D9	CS	32	1.48	0.03	0.79	0.81	0.04	7	44	7	2	7	31	<2	86	21	2	66	2	1
KD1D10	CS	36	1.30	0.04	0.82	0.74	0.03	6	42	6	<2	8	29	<2	87	23	3	67	4	<1
KD1D11	CS, C	42	4.04	0.39	0.87	0.49	0.05	8	39	40	11	26	49	56	86	30	8	113	8	7
KD1D12	CS	47	1.04	0.01	0.70	0.69	0.03	6	41	<5	3	5	39	<2	74	20	2	60	2	<1
KD1D13	CS, G	67	<0.5	<0.05	0.69	0.22	0.04	8	55	<5	<2	7	34	<2	105	24	2	59	1	<1
KD1D14	CS	122	<0.5	<0.05	0.68	0.07	0.02	5	45	<5	4	5	36	<2	83	20	2	62	2	<1
KD1D15	FS, CS	132	<0.5	0.05	0.82	0.10	0.02	5	39	8	<2	7	35	<2	91	25	4	102	6	1
KD1D16	FS	167	1.08	0.16	0.92	0.08	0.02	4	38	12	3	9	40	8	93	27	6	130	9	5
KD1D17	CS, G	236	<0.5	0.05	0.76	0.15	0.02	5	41	5	3	7	40	<2	79	23	3	79	4	<1
KD1D18	CS	247	<0.5	0.05	0.74	0.09	0.02	5	37	5	2	8	33	<2	74	22	3	76	4	1
Baniyatar (KD2)																				
KD2D1	CS	5	1.55	0.18	2.26	0.06	0.07	6	46	8	6	7	48	<2	119	28	7	83	4	2
KD2D2	SC	12	7.41	0.78	1.43	0.22	0.13	11	27	88	29	54	93	141	86	41	17	212	19	14
KD2D3	SC	22	5.52	0.58	11.95	0.25	0.28	9	19	73	23	38	67	101	252	31	13	131	14	22
KD2D4	G	31	<0.5	<0.05	1.16	0.23	0.03	9	63	<5	2	5	27	<2	140	25	2	52	2	<1
KD2D5	SC	37	7.50	0.77	1.28	0.23	0.11	12	28	90	34	58	94	152	91	40	18	186	16	16
KD2D6	G	49	<0.5	<0.05	0.84	0.28	0.03	8	69	<5	2	7	28	<2	141	26	1	49	<1	<1
KD2D7	CS, G	54	0.90	<0.05	0.90	0.58	0.03	6	46	<5	<2	6	38	<2	98	22	2	63	1	<1
KD2D8	SC	57	7.89	0.74	1.06	0.22	0.07	12	32	94	27	49	81	138	60	48	20	176	21	13
KD2D9	CS, G	78	1.21	0.02	0.77	0.69	0.03	4	39	<5	2	4	36	<2	77	19	2	102	3	<1
KD2D10	CS, G	87	0.92	<0.05	0.81	0.66	0.03	6	46	<5	2	4	37	<2	86	19	2	67	1	<1
KD2D11	CS	95	<0.5	<0.05	0.67	0.18	0.02	5	42	<5	<2	5	32	<2	69	20	2	62	2	<1
KD2D12	CS	102	<0.5	<0.05	0.74	0.19	0.02	5	40	<5	<2	5	30	<2	76	19	1	61	3	<1
KD2D13	SC	108	6.90	0.79	1.20	0.10	0.06	6	27	70	21	42	80	137	96	44	17	226	18	15
KD2D14	CS, G	116	<0.5	<0.05	0.73	0.21	0.02	7	51	<5	<2	6	28	<2	97	26	2	56	1	<1
KD2D15	CS, G	132	<0.5	<0.05	0.73	0.21	0.02	6	42	<5	<2	7	29	<2	75	22	2	59	2	<1
KD2D16	CS	162	6.14	0.69	1.20	0.15	0.08	7	31	67	18	38	66	112	83	41	16	180	15	12
KD2D17	SC	176	0.66	<0.05	0.72	0.08	0.02	5	43	<5	<2	5	28	<2	60	24	2	64	3	<1
KD2D18	CS, G	185	<0.5	<0.05	0.68	0.05	0.02	5	41	<5	2	7	16	<2	56	21	2	59	2	<1
KD2D19	CS	208	<0.5	<0.05	0.68	0.06	0.02	4	38	<5	2	6	19	<2	50	19	1	59	2	<1
KD2D20	CS	225	<0.5	<0.05	0.69	0.10	0.02	4	38	<5	2	4	18	<2	51	23	2	53	2	<1
KD2D21	CS	245	<0.5	<0.05	0.67	0.07	0.02	6	51	<5	3	5	21	<2	64	24	1	46	<1	<1
Sp2	WF	4.5	3.78	0.33	2.49	0.28	2.60	218	31	40	45	77	160	1,211	32	31	20	92	8	7
UCC	Đ	Đ	4.50	0.50	4.20	0.17	Đ	1.5	20	71	25	20	35	60	350	22	25	190	11	11

Lithological code: SC silty clay, C clay, G gravel, CS coarse sand, FS fine sand, WF wood fragments, UCC upper continental crust (adapted from Taylor and McLennan 1985)

in the central part of the valley indicates the presence of higher contents of silt-size zircons in the finer-grained sediments. Phosphorus mineralization is abundant in the Kathmandu Valley sediments (Dill and Melcher 2004) and the occasional high P values

(up to 0.8 wt%) observed in this study could also be due to the presence of phosphate minerals.

To further examine the effect of grain size on the elemental concentrations, the samples were divided into three categories based on lithotype, i.e., Gravel

Table 2 Major and trace elements analyses, central Kathmandu Basin, Nepal

Sample ID	Lithology	Depth (m)	Major oxides (wt%)				TS (wt%)	Trace element (mg/kg)												
			Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	CaO	P <sub>2</sub> O <sub>5</sub>		As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc
Sankhamul (KS2)																				
KS2D1	C	1.10	6.57	0.88	0.86	0.07	0.04	6	28	86	26	48	90	144	72	47	22	204	17	14
KS2D2	CS	1.76	0.81	0.14	0.81	0.13	0.03	5	41	20	2	10	46	<2	80	83	11	128	40	1
KS2D3	FS	2.48	0.56	0.09	0.73	0.07	0.03	5	47	25	2	6	53	<2	84	26	5	72	6	4
KS2D4	FS	4.25	3.29	0.49	1.1	0.11	0.03	4	36	49	3	17	50	57	101	52	13	227	29	10
KS2D5	BC	5.25	11.59	0.81	1.06	0.12	0.07	15	25	92	36	60	108	195	65	44	18	131	19	16
KS2D6	BC	5.78	8.89	0.79	1.02	0.21	0.05	14	28	94	35	64	110	184	70	42	18	124	20	18
KS2D7	BC	6.26	7.54	0.74	0.98	0.16	0.05	10	21	67	22	45	82	141	59	40	17	246	19	13
KS2D8	BC	6.53	6.98	0.77	0.99	0.19	0.09	25	26	74	36	68	101	167	60	44	18	192	20	15
KS2D9	BC	7.00	14.92	0.63	1.33	0.33	0.08	12	19	74	27	43	92	196	65	34	14	110	15	12
KS2D10	BC	7.53	9.31	0.69	1.98	0.27	0.12	16	24	89	35	60	91	173	75	35	15	122	16	16
UCC	Đ	Đ	4.50	0.50	4.20	0.17	Đ	1.5	20	71	25	20	35	60	350	22	25	190	11	11

Lithological code: C clay, G gravel, CS coarse sand, FS fine sand, BC black clay, UCC upper continental crust (adapted from Taylor and McLennan 1985)

Table 3 Major and trace elements analyses, southern Kathmandu Basin, Nepal

Sample ID	Lithology	Depth (m)	Major oxides (wt%)				TS (wt%)	Trace element (mg/kg)												
			Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	CaO	P <sub>2</sub> O <sub>5</sub>		As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc
Thaiba (KD3)																				
KD3D1	C	2	4.94	0.70	0.76	0.11	0.06	9	20	54	17	38	75	115	45	37	17	368	19	11
KD3D2	G	5	5.33	0.44	0.68	0.10	0.03	11	17	24	8	18	68	87	22	25	10	327	12	5
KD3D3	C	35	5.12	0.71	0.86	0.13	0.08	8	22	60	19	43	92	119	51	40	17	319	19	11
KD3D4	C	185	1.65	0.21	1.24	0.15	0.11	7	27	17	5	9	70	21	67	21	6	115	4	10
KD3D5	BC	225	9.38	0.69	1.23	0.35	0.17	17	26	89	29	51	91	170	58	39	16	137	17	15
KD3D6	CS	300	1.62	0.12	0.91	0.30	0.07	7	39	15	3	7	39	2	76	23	7	61	2	8
Sunakhothi (KD4)																				
KD4D4	CS	11	3.10	0.54	0.68	0.13	0.03	4	11	19	5	20	58	58	43	28	13	297	15	6
KD4D5	BC	15	5.31	0.76	0.88	0.06	0.03	9	25	86	27	72	103	156	67	46	19	227	20	18
KD4D6	CS	18	2.99	0.52	0.70	0.16	0.02	4	10	16	6	20	62	64	44	27	13	313	15	7
KD4D7	BC	57	3.98	0.63	0.71	0.13	0.05	6	16	37	12	29	66	87	44	34	15	361	17	10
KD4D8	CS, G	65	3.81	0.57	0.94	0.14	0.03	3	11	31	7	27	68	76	56	32	14	290	16	9
KD4D9	G	70	3.89	0.59	1.05	0.15	0.05	8	14	35	12	29	66	84	58	32	14	306	16	9
KD4D10	BC	85	5.12	0.69	1.15	0.14	0.04	7	19	52	16	39	75	114	64	36	16	315	18	11
KD4D11	BC	105	7.66	0.69	1.58	0.21	0.07	11	23	77	26	49	85	145	76	38	16	224	17	14
KD4D12	G, CS	175	6.38	0.56	1.40	0.31	0.04	6	14	34	8	25	151	87	63	30	13	287	14	9
UCC	Đ	Đ	4.50	0.50	4.20	0.17	Đ	1.5	20	71	25	20	35	60	350	22	25	190	11	11

Lithological code: C clay, G gravel, CS coarse sand, BC black clay, UCC upper continental crust (adapted from Taylor and McLennan 1985)

including gravel (G) and sandy gravel (SG); Sand, fine sand (FS) to coarse sand (CS); and Clay, including silty clay (SC) and black clay (BC). The abundance of elements in all the three categories ranges considerably. However, average concentrations in each group do not differ greatly from the average composition of UCC (Fig. 5). The Clay average generally shows higher concentrations of most elements, and is slightly depleted in Nb, CaO and Sr with respect to UCC. In contrast, the average concentrations in Gravel and Sand are lower relative to UCC (Fig. 5), except for As, which is 3–5 times higher mostly.

Selected variation diagrams with the samples distinguished by the three lithological groups are given in Fig. 6. Significant positive correlations exist between As and Fe<sub>2</sub>O<sub>3</sub> (R = 0.6, n = 67), and between As and TS (R = 0.5, n = 67) (Fig. 6a–c). High correlations also occur between other element pairs, including Zn and Fe<sub>2</sub>O<sub>3</sub> (R = 0.9, n = 64), Cu and Ni (R = 0.9, n = 68) and Nb and TiO<sub>2</sub> (R = 0.9, n = 53) (Fig. 6d–f). These significant correlations show the effect of grain-size variation (Singh et al. 1999; Filipek and Owen 1979), with generally lower concentrations in the coarser-size grades, and higher contents in the clay (Fig. 6).

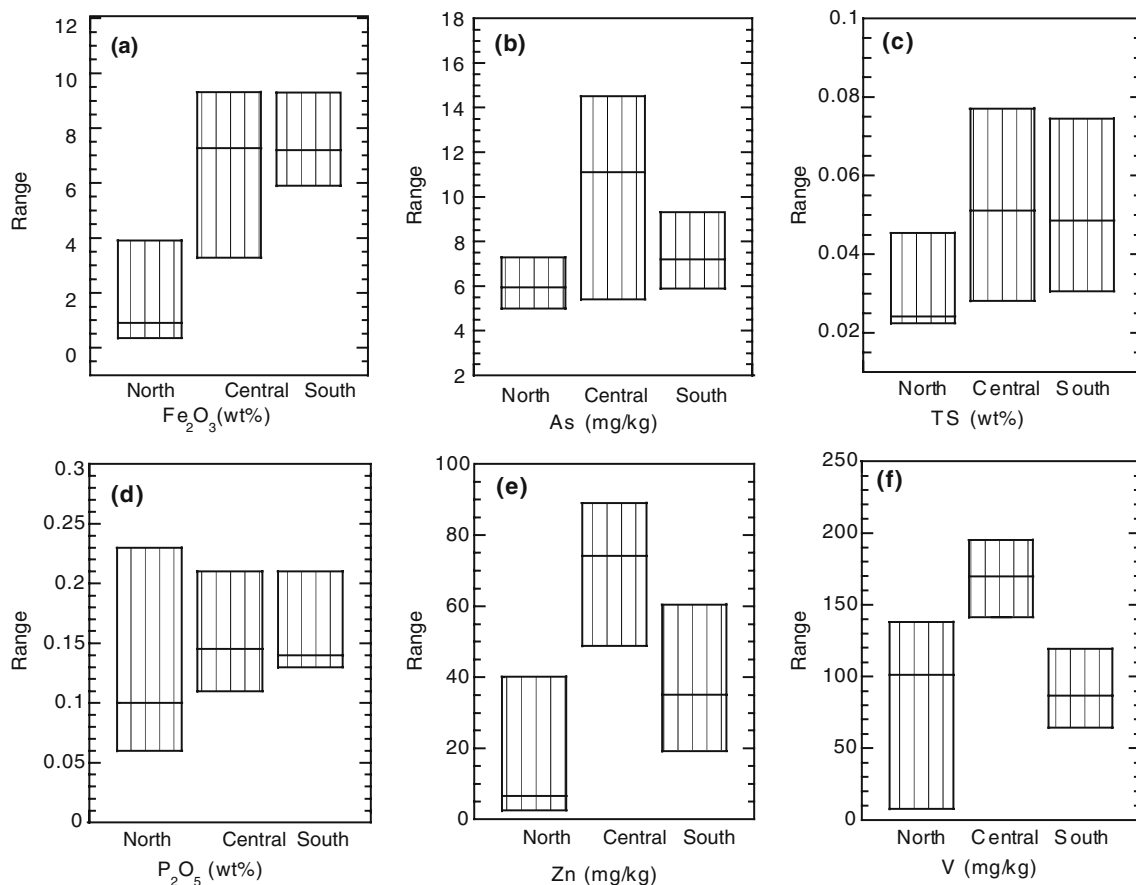
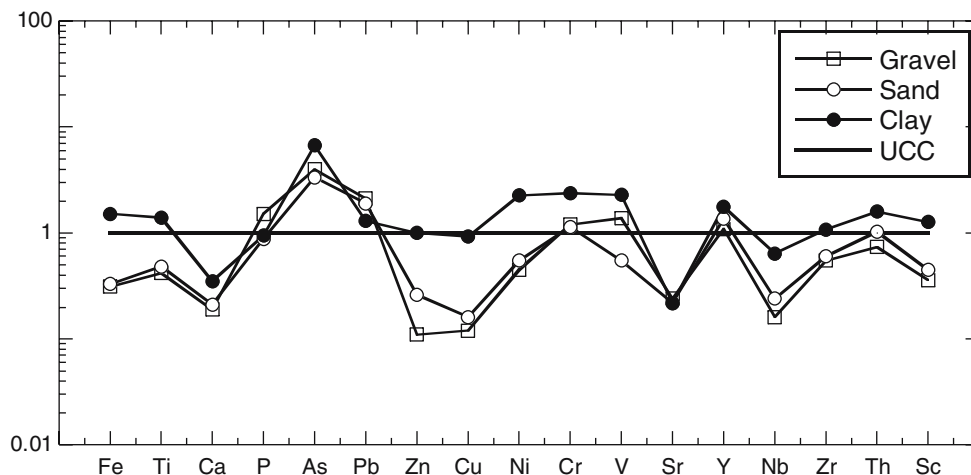


Fig. 4 Average values and ranges of selected elements from north to south, Kathmandu Basin, Nepal

Fig. 5 Average bulk concentrations of major oxides and trace elements by lithology normalized against average UCC composition (adapted from Taylor and McLennan 1985)

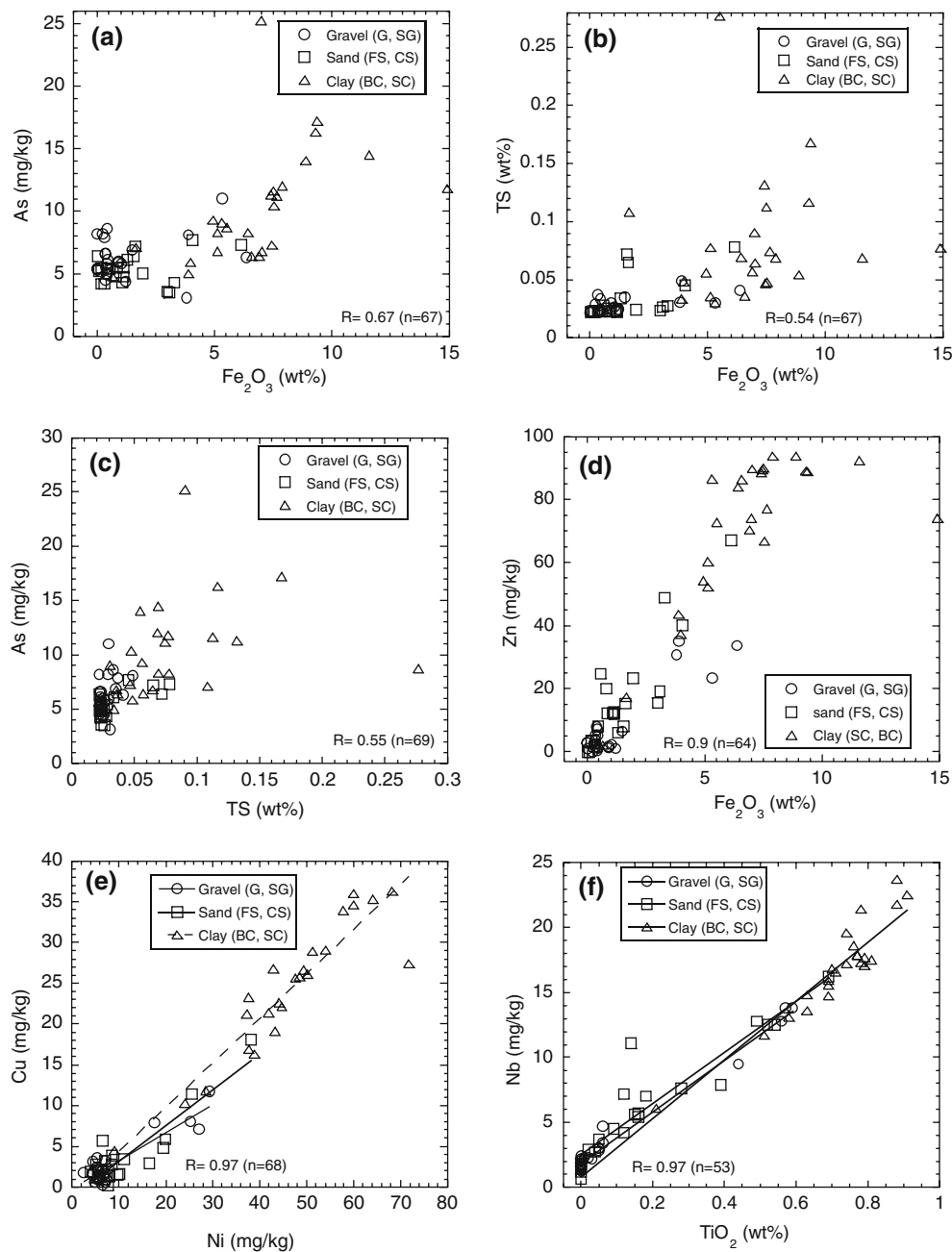


Elution analysis

The amount of As eluted from the core samples varied considerably, ranging from 0.1 to 511 g/L (Table 4). Sandy samples from the northern part of the valley (KD10, KD16 and KD212) yielded the least arsenic (0.1-0.4 g/L, Table 4). However, significant

amounts of As (25 and 261 g/L, respectively) were eluted from a muddy clay (KS12) and a silty clay (Sp1) from the same area. Low amounts of As (0.2-1.21 g/L) were also eluted from three shallow samples (Pne sand, KS24, clay, KS26 and KS210) from the central valley. Of the five samples analyzed from the southern margin of the valley, two clay samples (KD3D

Fig. 6 Selected elemental variations by grain size, Kathmandu Basin, Nepal. G gravel, SG sandy gravel, FS fine sand, CS coarse sand, BC black clay, SC silty clay



3 and KD4D11) yielded higher As (11 and 51 g/L), whereas two other clays (KD3D5 and KD4D20) and a sand (KD3D6) yielded lower As of 0.7, 0.1 and 0.2 g/L, respectively.

Although the number of samples subjected to elution analysis in this study was limited ( $n = 15$ ), the results show that only small amounts of arsenic are eluted from the coarser-grained samples by neutral water, and the maximum amount of arsenic eluted increases with decreasing grain size (Fig. 7). ER of the clayey samples ranges from low to high (from  $0.1 \times 10^{-2}$  to  $46.3 \times 10^{-2}$ ). However, the ER is not strictly

related to the bulk As content of the sediments (Table 4). The average bulk As content of the samples eluted is 9 mg/kg, with a range from 4 to 17 mg/kg. Among the samples eluted, the clay sample (KD3D5) with the highest bulk content (17 mg/kg) had one of the lower As ERs ( $0.4 \times 10^{-2}$ , Table 4).

## Discussion

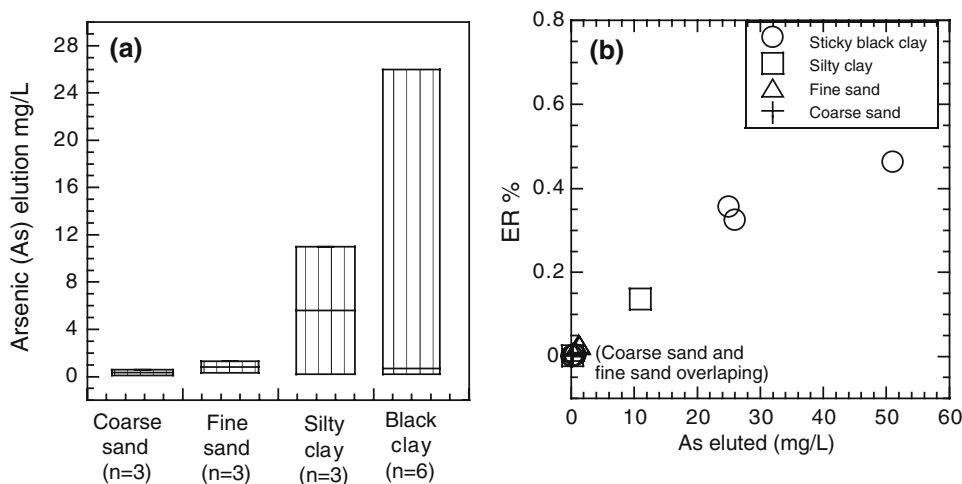
The overall bulk concentrations of the major oxides and trace elements determined for the sediments of

Table 4 Arsenic (As) elution from sediments, Kathmandu Basin, Nepal

S. No.	ID	Sediment type	Depth	As (1 g/kg); eluted (rme)	As in sediments (mg/kg)	% Elution ( $\times 10^3$ )
1	KD1D10	Coarse sand	35	0.1	6	0.2
2	KD1D16	Fine sand	170	0.4	4	1.0
3	KD2D8	Silty clay	60	0.2	12	0.2
4	KD2D12	Coarse sand	105	0.3	5	0.6
5	KD3D3	Gray clay	35	11.0	8	13.7
6	KD3D5	Sticky clay	210	0.7	17	0.4
7	KD3D6	Coarse sand	215	0.2	7	0.3
8	KD4D11	Black clay	37	51.0	11	46.3
9	KD4D20	Sticky clay	132	0.1	11	0.1
10	KS1D2	Sticky clay	3	25.0	7	35.7
11	KS1D4	Fine to coarse sand	7.5	1.3	5	2.6
12	KS2D4	Fine sand	4.5	1.2	4	3.0
13	KS2D6	Sticky clay	5.8	0.5	14	0.4
14	KS2D10	Sticky clay	7.5	0.2	16	0.1
15	Sp1	Silty clay	12	26.0	8	32.5

Rock mass equivalent (rme) = eluted vol.  $\times$  50 mL/500 mL; % Elution = eluted amount of As/As in the sediments

Fig. 7 Amount of arsenic eluted from the core sediments in deionized water, Kathmandu Basin, Nepal. ER = elution rate (%)



the Kathmandu Valley are not exceptional, and are comparable to the levels seen in average UCC. The Kathmandu Valley BII consists of coarser fluvial sediments at the margins, and grain size decreases toward the basin center, which is mostly filled with thick lacustrine clay. Elemental concentrations including arsenic increase from the margin of the valley toward the center (Fig. 4) in response to this decrease in grain size, as in general, concentrations of heavy metals in sediments tend to increase as the size fractions become finer (Singh et al. 1999; Filippek and Owen 1979). This is well illustrated by the variation diagrams in Fig. 6.

Arsenic concentrations in the sediments of the Kathmandu Valley average 8 mg/kg (ranging 3–25 mg/kg), similar to the general level seen in modern unconsolidated sediments, typically 5–10 mg/kg (Smedley and Kinniburgh 2002). The Kathmandu values are also similar to those reported from the Terai Basin of Nepal (Nawalparasi, average 9 mg/kg,

Gurung et al. 2005) and in the Holocene sediments in Bangladesh (7.5 mg/kg, Ali et al. 2003).

The highest value of arsenic in groundwater reported in the central valley (>200  $\mu$ g/L, JICA 2005) coincides with the higher arsenic contents of the sediments there. However, the elution tests of samples from the basin center and margins showed that the amount of As leached is not strictly congruent with the bulk concentration in the sediments (Table 4). Although ERs among the coarser samples were generally low (<3  $\times 10^3$ %), in the fine-grained samples the amount of arsenic eluted ranges from low to high (from  $0.1 \times 10^3$  to  $46.3 \times 10^3$ %).

These above features show that the bulk As contents of the aquifer sediments are not the controlling factor for high As concentrations in the groundwater. A more important aspect could be the process by which elevated concentrations in the groundwater can be generated from the relatively low bulk As concentrations in the aquifer sediments.

The literature on As contamination of groundwater all over the world is now vast, and many studies have stated and explained the nature of contamination and the potential mechanisms. In the Bengal Delta, three main mechanisms for As release have been proposed based on the chemical behavior of arsenic. These are: (1) mobilization of arsenic by oxidation of arsenic-bearing pyrite (Mallick and Rajgopal 1996; Mandal et al. 1996); (2) release of naturally occurring solid forms of arsenic due to reducing conditions (Nickson et al. 2000; McArthur et al. 2001; Smedley and Kinniburgh 2002); and (3) arsenic anions sorbed to aquifer minerals are displaced into solution by the competitive exchange of phosphate anions (Acharya et al. 2000).

The geochemical and hydrochemical characteristics of the Kathmandu Valley aquifers show some similarities with those of the Ganges Delta, in both the proximal part (Terai Basin) and the distal part (Bangladesh). High arsenic levels in groundwater in the Terai Basin are mainly related to the reductive dissolution of iron oxy-hydroxides, and the wide spatial variation is controlled by the lithofacies of the sediments and the varying organic matter content (Gurung et al. 2005). Generally, reducing conditions (low ORP), neutral pH, correlation of As and Fe oxides and negative correlation of As and  $\text{SO}_4$  are the typical characteristics associated with arsenic-rich groundwater in Bangladesh (Ishiga et al. 2000).

The mobilization of arsenic in the Kathmandu Valley is probably mainly related to the change in the redox conditions, as a redox gradient exists between waters in the shallow and deep aquifers. The shallow aquifers are relatively oxic, as indicated by high nitrate contents (>50 mg/L of N) and high ORP values (>100 mV, Jha et al. 1997). The widespread thick lacustrine clay probably restricts the downward diffusion of oxidants such as  $\text{O}_2$ ,  $\text{NO}_3^-$ . Earlier studies (e.g., Binnie and Partners 1973; JICA 1990; Jha et al. 1997) have reported high ammonium ( $\text{NH}_4^+$ ) and bicarbonate ( $\text{HCO}_3^-$ ) in deep-well groundwater. Khatiwada et al. (2002) indicated negative ORP values (–195 mV) in waters of the deeper aquifers, more common in the central part of the valley. All these features demonstrate that the deeper aquifers are under reduced conditions. Under such conditions, Fe oxy-hydroxides are the common host matter for arsenic, either adsorbed into the surface or co-precipitated (Pierce and Moore 1982), and under dissolution or desorption As could be released into groundwater (Bose and Sharma 2002; McArthur et al. 2001). In this study, the iron oxide content of the sediments are generally high (ranges < 0.5–15 wt%), and are uniformly higher in the fine sediments of the central valley (average

7 wt%). The correlation between Fe oxides and As is significant ( $R = 0.6$ ,  $n = 67$ , Fig. 6a), indicating their close association. A similar correlation has been observed between As and Fe in Bangladesh (Nickson et al. 2000; McArthur et al. 2001). Moreover, in the Kathmandu Valley the relatively low As contents found in the shallow well waters compared to deep wells is coupled with low dissolved Fe (JICA/ENPHO 2005; Amaya 2002). The low As and Fe in the shallow aquifer water could therefore be due to in-situ iron mineralization that significantly reduces the dissolved form (McArthur et al. 2004).

Phosphate ( $\text{PO}_4$ ) is another species of concern to the groundwater environment. Phosphate content is high (up to 22 mg/L) in the waters from some shallow and deep tubewells in the Kathmandu Valley (Jha et al. 1997; Amaya 2002). There are several possible non-point sources for such phosphate in the groundwater. Based on detailed mineralogical studies, Dill and Melcher (2004) reported that vivianite [ $\text{Fe}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$ ] was abundant in the Kathmandu Valley sediments. Dissolution of phosphate minerals can contribute to the high phosphate content in groundwater (Murphy et al. 2001; Rao and Prasad 1997). Anomalously high values of  $\text{P}_2\text{O}_5$  (up to 0.8 wt%) were observed in some samples in this study. Phosphate sourced from organic matter in the lacustrine clay, phosphate fertilizers and domestic sources such as septic tanks also cannot be dismissed. The lacustrine clay in the basin is rich in organic matter (Fujii and Sakai 2001), and the septic system is poorly managed throughout the Kathmandu Valley. Dissolved phosphate in the groundwater could compete for As sorption sites (Acharya et al. 2000), and may also affect As mobilization. Yamazaki et al. (2003a, b) eluted arsenic with phosphate elutant from core samples (peat) from a highly As-contaminated aquifer in Deuli village, southwest of Bangladesh, and reported that As elution was 20 times greater than in distilled water. The dissolved phosphate in the groundwater in the Kathmandu Basin could also have enhanced As release in a similar manner.

In the Terai Basin, arsenic contamination is greater in the shallow wells (<20 m) than in the deep wells. However, the opposite is the case in the Kathmandu Basin, with the deep tubewell water having the higher content (up to 200 µg/L). The cause of the higher As in the deep wells is not understood. However, conditions in the deep aquifers in the Kathmandu Basin are more reducing, as shown by lower ORP (–195 mV, Khatiwada et al. 2002) compared to the Terai Basin (ORP > –45 mV, Gurung et al. 2005). The deep water of the Kathmandu Valley also contain dissolved gases

including methane and hydrogen sulfide (JICA 1990); whereas no such gases have been reported from deep tubewells in the Terai Basin. Moreover, the Kathmandu Valley is a closed basin and groundwater movement is assumed to be relatively low, particularly in the deep aquifer. In contrast, in the Terai Basin groundwater generally flows toward the south, flow-gradient is relatively high (e.g., 3 m/km in Rupandehi, UNDP 1989), possibly attenuating the dissolved arsenic, resulting in its low concentrations.

It is widely accepted that organic matter may play an important role in the mobilization of arsenic, as shown by many case studies from West Bengal (India) and Bangladesh (Nickson et al. 2000; McArthur et al. 2001; Akai et al. 2004). Organic matter analyses were not attempted in this study, higher concentrations of solid organic matter can be expected to occur in the fine-grained lacustrine sediments than in the coarser alluvial fan deposits. Previous studies (e.g., Fujii and Sakai 2001; Dill et al. 2003) have indicated that the fine-lacustrine sediments in the valley are rich in organic matter, and this organic matter may play a significant role in As mobilization. The higher arsenic ERs from the clayey sediments (up to  $46.3 \times 10^{-2}\%$ , Table 4) in this study could have been influenced by the organic matter.

The wood fragment (Sp2) analyzed in this study was rich in As (218 mg/kg). Earlier studies (e.g., Igarashi et al. 1988) have reported that wood fragments are abundant in the Kathmandu Valley sediments. When buried, plant material is changed into peat, which is subsequently transformed to coal or lignite. Lignite beds are distributed in the central and southern parts of the valley (Dangol 1985; Dill et al. 2003). Unfortunately, the core samples in this study did not penetrate lignite. Buried plants such as mangrove forests are a potential source of arsenic in Bangladesh (Ishiga et al. 2000), and peat from shallow aquifers yielding arsenic-rich groundwater (>200 µg/L) in Bangladesh have high bulk As concentrations (111 mg/kg) (Yamazaki et al. 2003a, b). Therefore, buried wood fragments and peat beds in the Kathmandu Valley sediments could also be a potential source of arsenic in the groundwater. This possibility requires further study.

As in the Terai Basin, release of As from pyritic phases is unlikely to be significant in the Kathmandu Valley, as the pH of the aquifer waters is almost neutral (Amaya 2002; JICA/ENPHO 2005). Although pH could be lowered by  $\text{SO}_4^{2-}$  from sulfides (range 2.3–5.9, Lengke and Tempel 2005), measured  $\text{SO}_4^{2-}$  contents of the deep wells are low (<2 mg/L, Amaya 2002), further suggesting that As release from sulfides is not the main process controlling the composition of groundwater in the Kathmandu Valley.

## Conclusions

The overall concentrations of major oxides ( $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$ ,  $\text{CaO}$ ,  $\text{P}_2\text{O}_5$ ) and trace elements (As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, Sc and TS) of the sediments of the Kathmandu Valley are not exceptional, and are similar to modern unconsolidated sediments and average composition of UCC. Overall concentrations increase toward the center from the northern marginal parts of the Kathmandu Valley. The variations of elemental concentration are mainly clay-controlled in both the margin and central parts of the valley. Arsenic concentration in the sediments of Kathmandu Valley average 8 mg/kg (ranging 3–25 mg/kg), similar to typical modern sediments (5–10 mg/kg). The elution analyses have shown that the amount of arsenic eluted is not strictly corresponding with the bulk concentration in the sediments. The coarser-grained sediments eluted low amount of arsenic and the maximum amount of arsenic eluted increases with decreasing grain size, which indicates that the widespread lacustrine clay of the Kathmandu Valley could have greater potentiality for arsenic release. The mobilization of arsenic in the Kathmandu Valley is probably mainly related to change in the redox conditions; resembles the mechanism as reported from Terai Basin (Nepal) and Bengal Delta (Bangladesh). Iron oxides are abundant in the Kathmandu Valley sediments and show close association with arsenic, suggesting that iron oxy-hydroxides could be the potential source of arsenic. Also, the role of organic matter could be important for the arsenic release.

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