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Support for the fluvial recharge model for arsenic contamination of groundwater in Pokhara Valley, Nepal Himalaya

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ABSTRACT

Elevated levels of As in groundwater in the floodplain of the Ganges River have been well-documented over the past 15 years. Recent studies have shown that elevated groundwater As occurs even in Kathmandu Valley in Nepal, a tectonic valley well upstream of the floodplain of the Ganges River. Moreover, studies in Kathmandu Valley showed surface water As to be statistically indistinguishable from groundwater As, which led to the fluvial recharge model in which elevated groundwater As results from losing streams (streams that recharge groundwater) with elevated As, which is a consequence of rapid erosion caused by a combination of monsoon climate, tectonic uplift and deforestation. The objective of this study was to further test the fluvial recharge model and other existing models in Pokhara Valley, another tectonic valley in Nepal Himalaya far upstream from the floodplain of the Ganges River. In November 2010 water samples were collected from 20 hand-dug wells (depths 2-18 m), 12 borings (depths 34-220 m), four springs, 16 streams, three lakes and two caves. Arsenic concentrations in all but one of the 57 samples (a stream) exceeded the WHO As Standard (As = 0.01 mg/L). The As concentration of all surface water (geometric mean As = 0.067 mg/L) was statistically indistinguishable (P = 0.43) from that of all groundwater (geometric mean As = 0.086 mg/L), which is consistent with the fluvial recharge model. Groundwater As was uncorrelated with either sulfate or any combination of the transition elements, which is inconsistent with both the reductive-dissolution and sulfide-oxidation models.

Key words: Arsenic, fluvial, groundwater, Nepal, Pokhara.

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INTRODUCTION

The widespread contamination of groundwater with As in the floodplain of the Ganges River in Bangladesh and West Bengal (India) has been well-documented over the past 15 years (Bhattacharaya et al. 1997; Dhar et al. 1997; Nickson et al. 1998). Studies over the past ten years have documented that the region of As contamination extends even into the Terai Zone, the Indo-Gangetic Plain of southern Nepal (Fig. 1) (Neku and Tandukar 2002; Bhattacharya et al. 2003; Shrestha et al. 2003; Brikowski et al. 2004, 2006; Emerman 2004; ENPHO and USGS 2004; Kanel et al. 2005; Tandukar et al. 2005; Neku et al. 2006; Panthi et al. 2006; Neku and Brikowski 2009; Pokhrel et al. 2009; Emerman et al. 2012).

According to the most recent count in a scholarly journal, 737,009 groundwater samples in Nepal have been tested for As, of which 7.9% exceeded the WHO (World Health Organization) As Standard (As = 0.01 mg/L) and 2.3% exceeded the Nepal Interim As Standard (As = 0.05 mg/L) (WHO 2008; Thakur et al. 2011). IRIN (2010) reported that UNICEF had tested over 1.1 million wells in the Terai and found that 1.8% exceeded the Nepal Interim As Standard, while another 5.6% exceeded only the WHO As Standard. It is estimated that there are perhaps 200,000 tubewells in the Terai Zone and that 3.5 million Nepalis have no access to drinking water that does not exceed the WHO As Standard (Pokhrel et al. 2009). Other studies in the Terai have addressed the geologic setting of As contamination (Shrestha et al. 2004; Williams et al. 2004, 2005; Brikowski et al. 2005; Gurung et al. 2005; Van Geen et al. 2008), the health impacts of As-contaminated groundwater upon the local population (Ahmad et al. 2004; Maharjan et al. 2005, 2006, 2007) and the As contamination of soils and crops (Dahal et al. 2008).

The dominant paradigm for As contamination of groundwater in south Asia is the reductive-dissolution model, according to which the strongly reducing conditions of the thick sedimentary package of the Indo-Gangetic Plain cause the release of As from sorption sites on Fe oxyhydroxides after dissolution of the Fe oxyhydroxides by micro-organisms (Nickson et al. 2000; McArthur et al. 2001; Bose and Sharma 2002; Harvey et al. 2002) or after reduction of adsorbed As from As^{+5} (arsenate) to As^{+3} (arsenite) (Bose and Sharma 2002). The important implication of the reductive-dissolution model is that As contamination does not result from even indirect human activity and that nothing can be done to reduce the input of As into aquifers. However, the input of organic-rich water from irrigated fields and constructed ponds may provide the carbon source for reductive dissolution (Charlet and Polya 2006). A competing model has been the sulfide-oxidation model, according to which overpumping of aquifers has caused oxidation of sulfide minerals and release of co-precipitated As into groundwater (Badal et al. 1996; Mallick and Rajgopal 1996). Williams et al. (2004, 2005) has argued that As contamination in the Terai Zone could result from sulfide oxidation rather than reductive dissolution. Polizzotto et al. (2006) has produced a cogent criticism of the reductive-dissolution model based upon observations and experiments on sediments from Bangladesh.

What both the reductive-dissolution and sulfide-oxidation models have in common is that they draw attention to the environmental conditions existing in the soil or sedimentary package and have no role for fluvial As. However, Emerman (2005) and Emerman et al. (2007, 2011) found elevated As in rivers in Kathmandu Valley and throughout central and eastern Nepal outside of the Higher Himalayan Zone. Emerman (2005) measured fluvial As six times monthly in eight rivers in Kathmandu Valley and found mean fluvial As exceeding the WHO As Standard (As = 0.01 mg/L) in all but one river. Emerman (2005) and Emerman et al. (2007) also collected 115 fluvial samples from 30 locations outside of Kathmandu Valley and the Higher Himalayan Zone and found that 53% of samples met or exceeded the WHO As Standard. Emerman et al. (2011) measured fluvial As from the Himalaya to the Ganges River floodplain along a 288-km traverse of the Sunkoshi River to the Saptakoshi River. They found that, upstream from the Ganges River floodplain, 45% of fluvial samples met or exceeded the WHO As Standard,

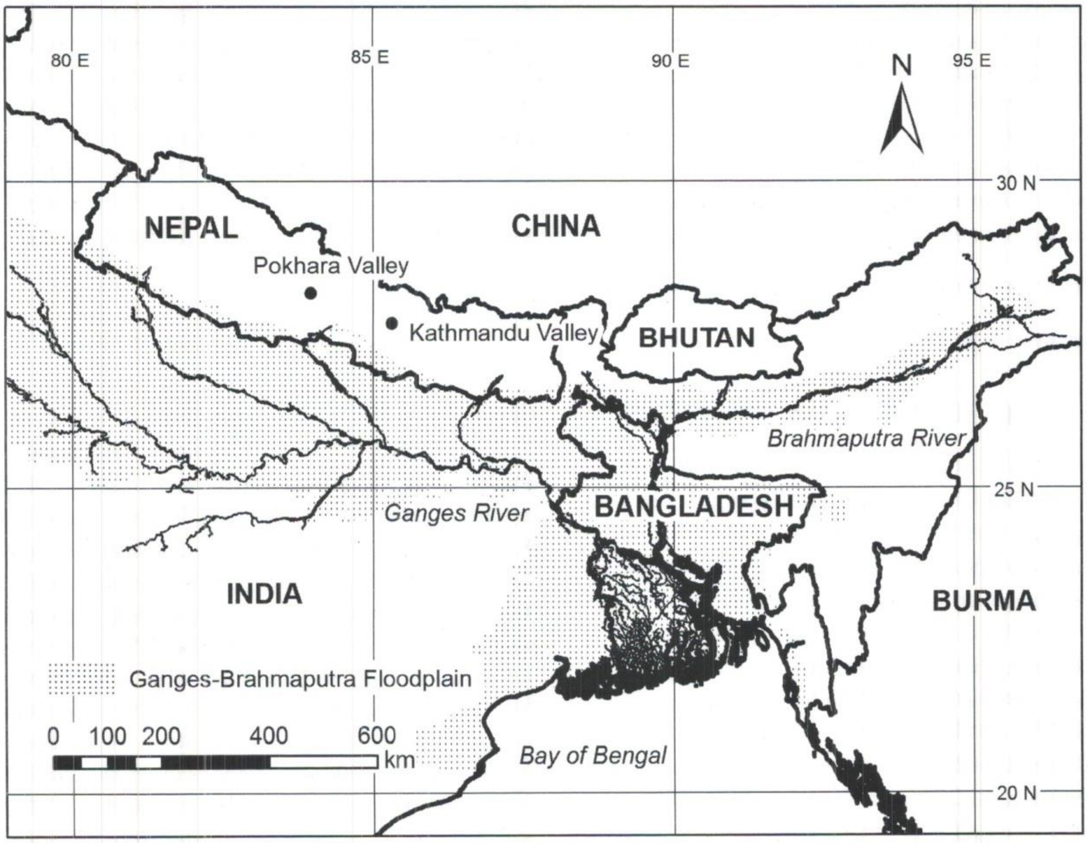

Fig. 1: Kathmandu and Pokhara Valleys are far upstream from the Ganges River floodplain in which As contamination of groundwater has been well-documented. Base map data are from Hearn et al. (2001).

while As concentrations fell to undetectable within the Ganges River floodplain. By contrast, the global background fluvial As is in the range 0.00013-0.0021 mg/L (Smedley and Kinniburgh 2002). Emerman et al. (2007, 2010, 2011) questioned whether it could be a coincidence that rivers with elevated As are draining into sedimentary basins in which the groundwater has elevated As. They suggested that while both the reductive-dissolution and sulfide-oxidation models could be thermodynamically plausible, the kinetics could be sufficiently slow that the groundwater chemistry simply reflects the surface water chemistry. This reasoning led to the fluvial recharge model for As contamination of groundwater, according to which groundwater is recharged by losing streams with elevated As due to rapid erosion caused by monsoon climate, tectonic uplift and deforestation.

Rapid erosion should lead to increased concentrations of all elements in rivers because, as the rate of erosion increases, a given volume of rainfall will result in the release of a larger mass of each element from rock, sediment or soil into overland flow, whether the rapid erosion is due to the high-intensity rainfall events of monsoon climate, the steep slopes resulting from tectonic uplift or the lack of surface cover resulting from deforestation or excessive grazing. A similar argument explains why rivers become more turbid as the river stage rises (Bloom 1998). The above argument should apply especially to As, which occurs in dissolved fluvial form predominantly as the arsenate oxyanion $HAsO_4^{-2}$ (Smedley and Kinniburgh 2002). Most ions will tend to adsorb onto sediment during the course of overland flow. Much of the sediment that is eroded from a watershed will not exit the mouth of the watershed, but will be redistributed throughout the watershed, in accordance with the geomorphic principle that smaller landforms erode faster than larger landforms (Bloom 1998). On that basis, much of the mass of elements that are released into overland flow during rapid erosion will not appear in rivers, although most of the water involved in overland flow will appear in rivers. However, the As oxyanion will not tend to adsorb onto sediment during rapid overland flow. First, sediment tends to have many fewer positively-charged sorption sites than negatively-charged sorption sites. Second, the As oxyanion has both much greater mass and volume than the elements that occur in aqueous form as cations or monatomic anions, and greater mass than other common oxyanions such as NO_3^- and PO_4^{-3}. Since all ions in overland flow are travelling with the same velocity, the As oxyanion will have much greater kinetic energy than the other ions and will tend to "bounce" off of sorption sites. To put it in another way, a divalent As oxyanion cannot attach to two positively-charged sorption sites unless it has sufficient residence time in the vicinity of the sorption sites for both sites to become simultaneously vacant, for example, by diffusion of two monovalent chloride ions from sorption sites into water. Moreover, the large volume of the As oxyanion implies that the region of the two sorption sites must be sufficiently large for the As oxyanion to attach. The result is that the As oxyanion cannot attach to sorption sites when the overland flow is rapid so that most of the As that is released into overland flow by rainfall will appear in rivers rather than be adsorbed onto sediment that is deposited within the watershed.

Recent studies have shown that elevated As in groundwater is not confined to the Terai Zone of Nepal, but occurs even in Kathmandu Valley, a tectonic valley well upstream from the Ganges River floodplain (Fig. 1) (Khatiwada et al. 2002; Gurung et al. 2006; Bajracharya et al. 2007; Warner et al. 2008; Chapagain et al. 2009; Maharjan et al. 2009; Emerman et al. 2010; Thakur et al. 2011). For example, Emerman et al. (2010) found As levels exceeding the Nepal Interim As Standard in 33% of 27 groundwater samples. Emerman et al. (2010) tested the fluvial recharge model by comparing the As concentrations of groundwater samples with 48 river samples obtained in a previous study in Kathmandu Valley (Emerman 2005). They found that the geometric mean As concentration of groundwater (As = 0.015 mg/L) was statistically indistinguishable from the geometric mean As concentration of surface water (As = 0.013 mg/L). Emerman et al. (2010) also tested the reductive-dissolution and sulfide-oxidation models by comparing the concentration of As in groundwater with concentrations of sulfide-forming transition elements (Fe, Cu, Mn, Ni, Co, Zn, Cr). Arsenic was uncorrelated with either Fe ($R^2 = 0.096$), Mn (R2 = 0.0004), or any combination of the transition elements ($R^2 < 0.083$), which is inconsistent with both models. The reductive-dissolution model predicts positive relations between As and both Fe and Mn, due to the breakdown of Fe oxyhydroxides and release of As under reducing conditions, and the lack of dissolved Mn ($Mn \leq 0.2$ mg/L) that should occur under oxidizing conditions (Van Geen et al. 2008).

The findings that elevated groundwater As occurred outside of the Terai Zone and that the geochemical data were inconsistent with the reductive-dissolution and sulfide-oxidation models were so startling that it was decided to repeat and extend the Kathmandu Valley study (Emerman et al. 2010) in Pokhara Valley, the second most heavily populated tectonic valley in Nepal after Kathmandu Valley (Fig. 1). Although Kathmandu Valley lies outside of the Ganges floodplain, the abundance of lacustrine clays makes a reducing subsurface environment likely (Gurung et al. 2006), so that Kathmandu Valley could act as a miniature version of the Ganges floodplain. On the other hand, the sediments of Pokhara Valley are dominated by coarse-grained debris flow deposits of limestone, schist, gneiss and granite (Fort and Gupta 1981; Fort and Freytet 1982; Koirala and Rimal 1996; Koirala et al. 1997, 1998) in which an oxidizing subsurface environment is highly likely so that Pokhara Valley is the area where elevated As due to reductive dissolution should be least expected. On the other hand, the abundance of carbonaceous sediments could promote As mobilization due to the reduction in number of positively-charged sorption sites that occur at higher pH. The specific goals were:

1. to determine whether elevated groundwater As occurs in Pokhara Valley,
2. to test the fluvial recharge model by comparing concentrations of As in groundwater and surface water,
3. to test the reductive-dissolution model by comparing As concentrations with Fe and Mn concentrations in groundwater,
4. to test the sulfide-oxidation model by comparing As concentrations with concentrations of sulfate and the transition elements that are normally associated with As, and
5. to determine whether there is clustering of As concentrations by watershed, depth or surface location.

Although geomorphology and natural hazards in Pokhara Valley have been extensively studied since the late 1970s (Fort and Freytet 1979, 1982; Fort 1981, 1984,1987; Freytet and Fort 1980; Fort and Gupta 1981; Kansarkar et al. 1982; Yamanaka 1982; Yamanaka et al. 1982; Merh 1985; Thapa et al. 1989; Koirala and Rimal 1995, 1996; Koirala et al. 1997, 1998; Sikrikar et al. 1998; Kaphle 2001, 2002; Upreti and Yoshida 2005), the only previous published study of hydrology or water quality in the region of Pokhara Valley has been an abstract by Warner et al. (2007). Warner et al. (2007) collected samples from springs and surface water along the trekking route from Pokhara to Muktinath in the Annapurna Conservation Area. Warner et al. (2007) did not report measurements of As, but found elevated levels of Hg, which they attributed to industrial pollution carried by winds from the southeast.

MATERIALS AND METHODS

In November 2010 water samples were collected from 20 hand-dug wells, 12 borings, four springs, 16 streams, three lakes and two caves over the whole of Pokhara Valley (about 25 km × 10 km) (Figs. 2 and 3). To the best of our knowledge, every dug well and boring in Pokhara Valley was sampled. Dug wells were found in four clusters in the villages of Karkiko Tara, Leknath and Mohoriya, and in the Lakeside neighborhood of Pokhara City (Fig. 2) with five

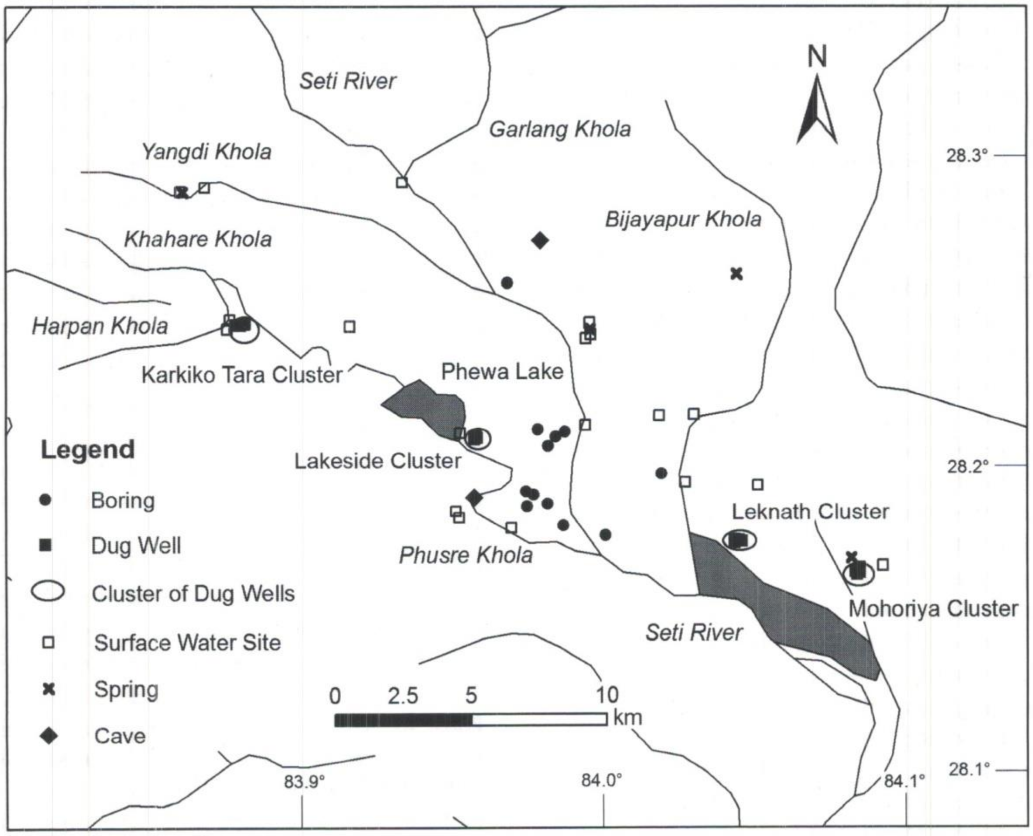

Fig. 2: Water samples were collected from 12 borings, 19 surface water sites, four springs, two caves, and 20 dug wells in four clusters. Base map data are from Hearn et al. (2001) and Shangri-La Maps (2008).

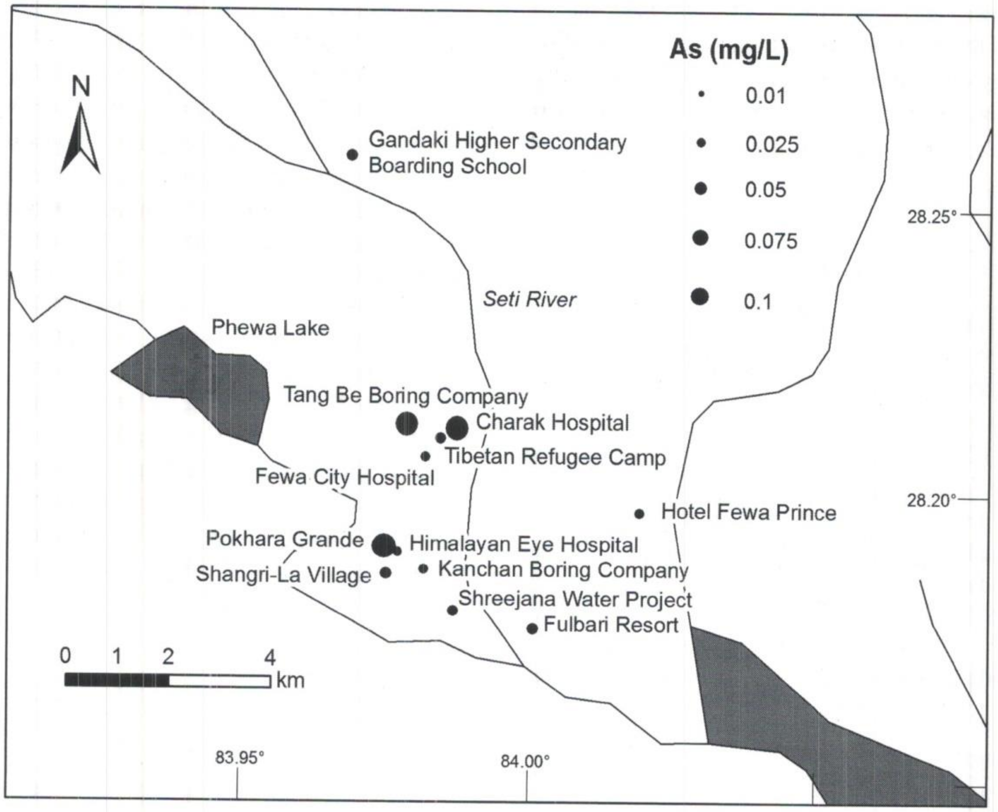

Fig. 3: No spatial pattern was found in As concentrations in borings. Arsenic concentrations exceeded the Nepal Interim As Standard (As = 0.050 mg/L) in borings owned by Pokhara Grande, Tang Be Boring Company, and Charak Hospital and Research Centre. Base map data are from Hearn et al. (2001) and Shangri-La Maps (2008).

wells in each cluster over length scales of 150-325 m. Depths of dug wells ranged from 1.7-18.3 m (Table 1a) with an average depth of 8.2 m, which is inconsistent with an earlier report that the depth to the water table in Pokhara exceeded 50 m (Kaphle 2002), although very few dug wells may have existed at the time of the earlier study. Borings were much deeper with depths ranging from 33.5-220 m (Table 2a), with an average depth of 110 m. Although it was not possible to purge dug wells before sampling, all borings were running continuously. Water temperature and pH were measured on site with the Hach EC-10 pH Meter. Electrical conductivity (EC) was measured on site with the Hanna HI 8033 Multi-Range Conductivity Meter. Locations were measured with the Trimble Juno SB GPS Receiver. Information collected about each dug well or boring included depth, diameter, screen interval, age, uses of water, and methods of water treatment. (Complete data on well diameters, uses of water and methods of water treatment are not reported in this paper, but are available from the authors.)

Water samples were collected in a 250-mL and a 125-mL polyethylene container with the contents of the larger container used for measurement of As and the other used for all other heavy metals and sulfate. Water samples were returned to Utah Valley University where water was forced through a 0.45-μm syringe filter after which trace metal grade concentrated nitric acid was added to reduce pH < 2 (Sanders 1998). Filtration and acidification were completed within one week of collection of all samples. Prior to analysis for all parameters beside As, concentrated NaOH was added to raise pH to the range 4-5. Concentrations of Fe, Cu, Ni, Co, Mn, Zn, Cr and sulfate were measured using the Hach DR-2700 Spectrophotometer. Water samples were analyzed for As using the silver diethyldithiocarbamate method (U.S. Environmental Protection Agency Standard Method 3500-As B) (Clesceri et al. 1998) with the same spectrophotometer. The spectrophotometer was re-calibrated after every 15 As measurements using three standard solutions with As concentrations of 0.02 mg/L, 0.04 mg/L and 0.2 mg/L. The coefficient of variation (ratio of standard deviation to mean) of the silver diethyldithiocarbamate method is 6.6% for 0.005 mg As according to Marshall (1978) and <10% for 0.010 mg As according to Clesceri et al. (1998), which may be regarded as measures of the precision or reproducibility of the method. Based on the four calibrations carried out for this study, the accuracy of the method is As = 0.007 mg/L. Whenever the As concentration of a sample exceeded As = 0.220 mg/L, the reacted As absorbing solution was diluted with unreacted As absorbing solution until the As concentration of the mixture was less than As = 0.220 mg/L. All analyses were completed within six months of sampling. The detection limits were 0.001 mg/L for As, Ni and Co, 0.01 mg/L for Fe, Cu, Zn and Cr, 0.1 mg/L for Mn and 1 mg/L for sulfate.

RESULTS

Out of 57 samples, As concentrations in all but one (a stream) exceeded the WHO As Standard (Tables 1a, 2a, 3a, 4a). Moreover, the Nepal Interim As Standard was exceeded in 60% of dug wells (Table 1a), 25% of borings (Table 2a), 100% of caves (Table 3a), 75% of springs (Table 3a) and 74% of surface water sites (Table 4a). None of the dug wells owned by the hotels in the Lakeside neighborhood of Pokhara City had As concentrations exceeding the Nepal Interim As Standard (Table 1a), although none of these dug wells were used for drinking water. On the other hand, As concentrations exceeded the Nepal Interim As Standard in borings owned by the Pokhara Grande (As = 0.182 mg/L), Tang Be Boring Company (As = 0.161 mg/L) and Charak Hospital and Research Centre (As = 0.160 mg/L) (Table 2a, Fig. 3), all of which were providing water for drinking. Extremely elevated As concentrations exceeding 1 mg/L were found in two dug wells in the Mohoriya cluster (As = 7.900 mg/L, 2.180 mg/L) and one well in the Leknath cluster (As = 1.220 mg/L) (Table 1a, Fig. 2). By contrast, WHO drinking water standards for Mn (0.4 mg/L) were exceeded in only one dug well (Table 1b), one surface water site (Table 4b) and no borings, springs or caves (Tables 2b, 3b), while other WHO drinking water standards (Cu = 2 mg/L, Ni = 0.07 mg/L, Cr = 0.05 mg/L) were met for all water samples (WHO 2008). The Nepal National Drinking Water Quality Standard for Fe (0.3 mg/L) (Warner et al. 2008) was exceeded in only two dug wells (10%) (Table 1b). (WHO (2008) does not have drinking water standards for Fe, Co, Zn or sulfate.)

Table 1a: Description and arsenic concentrations of dug well sampling sites.

As Rank[1]	As (mg/L)	Location	Age[2] (yrs)	Depth (m)	Latitude[3] (°N)	Longitude[3] (°E)
1	7.900	Mohoriya	21	4.0	28.1643092	84.0841647
2	2.180	Leknath	7	18.3	28.1749525	84.0450278
3	1.220	Mohoriya	9-10	4.0	28.1648206	84.0851347
4	0.810	Karkiko Tara	1	3.4	28.2452286	83.8786361
5	0.660	Leknath	7	14.6	28.1741456	84.0439272
6	0.630	Karkiko Tara	1.5	4.0	28.2451911	83.8801728
7	0.212	Mohoriya	0.5	1.8	28.1662139	84.0851236
8	0.158	Karkiko Tara	10	4.0	28.2455200	83.8816914
9	0.097	Mohoriya	8-9	2.4	28.1646547	84.0841719
10	0.067	Leknath	7	14.6	28.1747914	84.0437975
11	0.067	Karkiko Tara	1.5	3.0	28.2452872	83.8785717
12	0.053[4]	Leknath	7	18.3	28.1746522	84.0462261
13	0.044	Leknath	9	15.9	28.1751383	84.0437522
14	0.037	Hotel Mayur, Lakeside	0.75	11.6	28.2099514	83.9583300
15	0.034	Boardwalk Guest House, Lakeside	0.6	10.4	28.2091936	83.9579364
16	0.030	Hotel Snowland, Lakeside	3	7.6	28.2087019	83.9577253
17	0.027	Candle Inn, Lakeside	10-12	10.7	28.2093444	83.9587811
18	0.025	Hotel ABC, Lakeside	1	10.1	28.2091986	83.9576561
19	0.024	Mohoriya	15	1.7	28.1650528	84.0837356
20	0.021	Karkiko Tara	2	4.3	28.2457689	83.8818561

[1] As Rank refers to the ranking of all dug well samples from highest to lowest As concentration.

[2] Ages are based upon interviews conducted in November 2010.

[3] Latitude and longitude are based upon WGS 84 coordinate system.

[4] Arsenic values above the dashed line exceed the Nepal Interim As Standard (As = 0.05 mg/L).

Table 1b: Geochemistry of dug wells.

As Rank[1]	Mn (mg/L)	SO_4 (mg/L)	Fe (mg/L)	Cu (mg/L)	Ni (mg/L)	Co (mg/L)	Zn (mg/L)	Cr (mg/L)
1	0.2	1	0.90	0.06	0.003	0.012	0.06	0.01
2	0.1	20	0.00	0.08	0.011	0.137	2.18	0.01
3	0.2	3	0.03	0.07	0.000	0.030	0.10	0.01
4	0.3	0	0.20	0.24	0.005	0.015	2.35	0.01
5	0.9*	4	0.19	0.05	0.007	0.000	1.93	0.02
6	0.1	0	0.17	0.04	0.011	0.122	1.78	0.03
7	0.2	0	2.88	0.07	0.024	0.139	1.88	0.02
8	0.2	1	0.00	0.00	0.005	0.003	0.13	0.01
9	0.1	0	0.06	0.03	0.001	0.000	0.07	0.01
10	0.2	0	0.15	0.28	0.060	0.139	2.14	0.03
11	0.1	0	0.16	0.05	0.000	0.022	0.13	0.01
12	0.3	14	0.01	0.35	0.012	0.132	2.14	0.01
13	0.0	1	0.22	0.18	0.023	0.326	3.88	0.02
14	0.0	12	0.02	0.08	0.000	0.037	0.07	0.01
15	0.0	7	0.01	0.61	0.006	0.000	0.10	0.02
16	0.3	12	0.06	0.22	0.009	0.034	0.15	0.01
17	0.2	6	0.04	0.02	0.001	0.039	0.22	0.01
18	0.3	17	0.00	0.00	0.000	0.042	0.08	0.01
19	0.2	1	0.27	0.06	0.000	0.030	0.12	0.01
20	0.0	0	0.02	0.05	0.000	0.006	0.10	0.01

[1] See Table 1a.

*Concentration exceeds the WHO Standard (Mn = 0.4 mg/L, Cu = 2 mg/L, Ni = 0.07 mg/L, Cr = 0.05 mg/L).

It is crucial to test for the existence of normal distributions prior to using any statistical tests that assume normal distributions. For each parameter, the mean and standard deviation were calculated and the cumulative percentage for each value (percentage of values equal to or less than that value) were compared with the cumulative percentage for a normal distribution with the same mean and standard deviation. The cumulative percentage of each value of each parameter was also compared with the cumulative percentage of the equivalent lognormal distribution (same mean and standard deviation as the set of logarithms of values). The use of a lognormal distribution requires an assumption about the zero values (values below the detection limit). We set all zero values equal to the detection limit. Samples were below the detection limit for 40% of sulfate values, 39% of Ni values, 19% of Mn values, 12% of Co values, 11% of Fe and Cu values, 5% of Cr values, and 0% of As, Zn and EC values, which would make it impossible to reasonably fit a lognormal distribution in the case of sulfate and Ni. Samples were separated into surface water and groundwater and the root mean square error (RMSE) between the cumulative percentage and the cumulative percentage of the equivalent normal and lognormal distributions was calculated for each parameter. Mean values of RMSE averaged over all elements and compounds and over all on-site parameters (pH, EC, temperature) were also calculated. For groundwater, element and compound distributions were a better fit to a lognormal distribution (mean RMSE = 12.1%) than a normal distribution (mean RMSE = 19.9%) (Figs. 4a-b). For surface water, element and compound distributions were also a better fit to a lognormal distribution (mean RMSE = 13.9%) than a normal distribution (mean RMSE = 17.0%) (Figs. 4d and 4e). For the on-site parameters, normal distributions were a good fit for both groundwater (mean RMSE = 5.4%) (Fig. 4c) and surface water (mean RMSE = 7.0%) (Fig. 4f). Based on the above, it was decided to carry out all statistics on the values of pH, EC and temperature, and on the logarithms of the values of element and compound concentrations, with all zero values set equal to the detection limit.

The fluvial recharge model was tested by using the Student's t-test to compare the geometric mean

Table 1c: On-site measurements of dug wells. EC-Electrical conductivity.El

As Rank[1]	Temperature (°C)	pH	EC (µS/cm)
1	22.6	7.27	624
2	24.1	7.39	377
3	22.9	7.34	435
4	21.1	6.64	28.5
5	24.1	6.48	145.6
6	23.1	6.23	25.5
7	23.8	7.09	514
8	23.2	6.17	—[2]
9	23.3	7.36	401
10	23.3	6.29	245
11	20.8	6.71	26.0
12	23.2	7.34	450
13	22.7	5.62	64.7
14	23.0	7.44	610
15	24.3	7.28	636
16	24.3	7.10	710
17	24.3	7.24	435
18	23.6	7.07	721
19	23.9	7.44	333
20	23.5	6.26	27.9

[1] See Table 1a.

[2] Indicates unknown value.

concentrations for groundwater and surface water. The geometric mean As concentrations of surface water (As = 0.067 mg/L) and groundwater (As = 0.086 mg/L) were statistically indistinguishable (P = 0.43), which is consistent with the fluvial recharge model (Fig. 5). Differences in concentrations of all other elements and compounds were indistinguishable, except for the statistically significant increase in concentration of Cu in groundwater relative to surface water (Fig. 5). Differences between means of surface water pH (7.82) and groundwater pH (7.15), surface water EC (106.4 µS/cm) and groundwater EC (346.4 µS/cm), and surface water temperature (20.5 °C) and groundwater temperature (23.4 °C) were all statistically significant at the 99% confidence level (Tables 1c, 2c, 3b, 4b). In both surface water and groundwater, concentrations of As, Mn, Co, Zn and Cr were elevated relative to global averages (Langmuir 1997) (Fig. 5).

The fluvial recharge model was further tested by separating groundwater samples into dug wells and borings (leaving out springs and caves). The differences between the geometric mean As concentration in surface water and

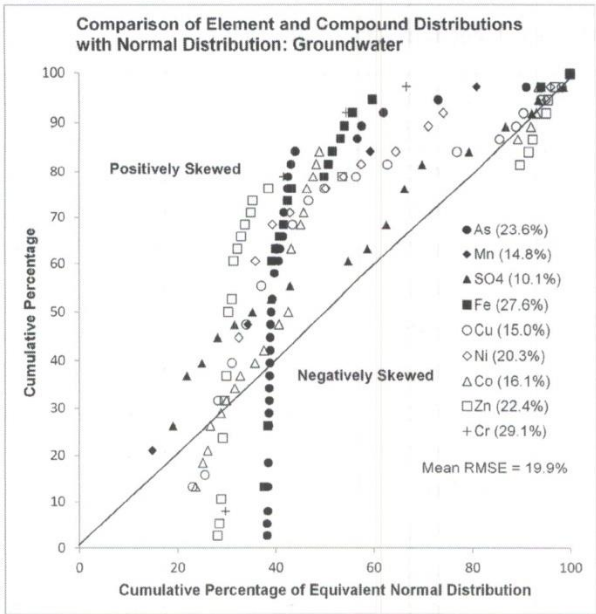

Fig. 4a: A comparison of the cumulative percentage of each element and compound in groundwater with the equivalent normal distribution with the same mean and standard deviation showed element and compound distributions to be a moderate fit to a normal distribution. The root mean square error (RMSE) averaged over all parameters was 19.9% (RMSE for each parameter shown in parentheses).

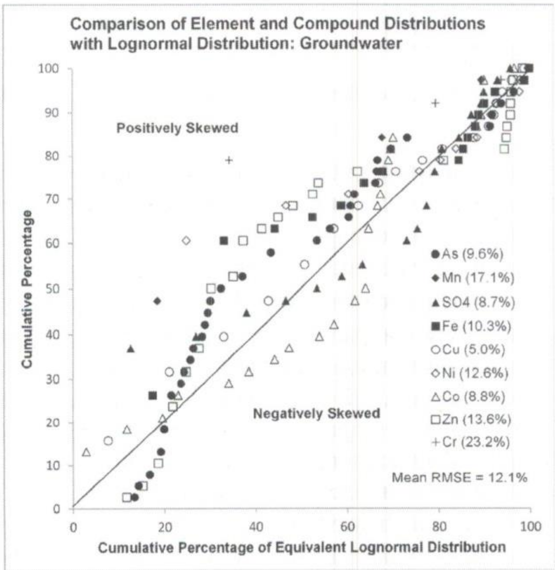

Fig. 4b: A comparison of the cumulative percentage of each element and compound in groundwater with the equivalent lognormal distribution with the same mean and standard deviation showed element and compound distributions to be a better fit to a lognormal distribution. The root mean square error (RMSE) averaged over all parameters was 12.1% (RMSE for each parameter shown in parentheses).

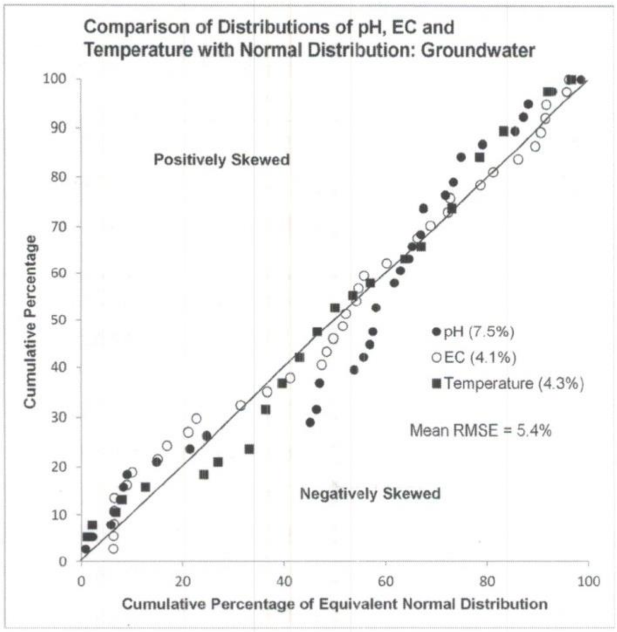

Fig. 4c: A comparison of the cumulative percentage of pH, electrical conductivity (EC), and temperature in groundwater with the equivalent normal distribution with the same mean and standard deviation showed distributions to be a good fit to a normal distribution. The root mean square error (RMSE) averaged over all parameters was 5.4% (RMSE for each parameter shown in parentheses).

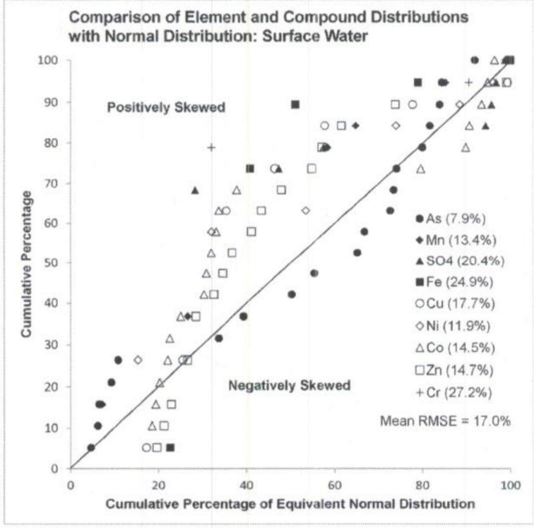

Fig. 4d: A comparison of the cumulative percentage of each element and compound in surface water with the equivalent normal distribution with the same mean and standard deviation showed element and compound distributions to be a moderate fit to a normal distribution. The root mean square error (RMSE) averaged over all parameters was 17.0% (RMSE for each parameter shown in parentheses).

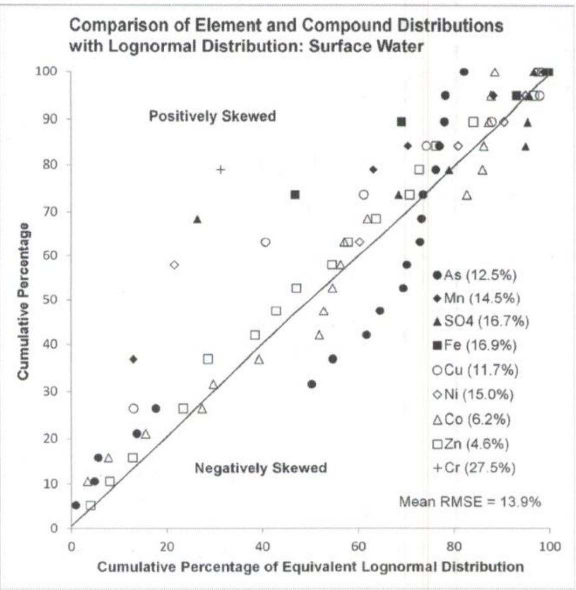

Fig. 4e: A comparison of the cumulative percentage of each element and compound in surface water with the equivalent lognormal distribution with the same mean and standard deviation showed element and compound distributions to be a better fit to a lognormal distribution. The root mean square error (RMSE) averaged over all parameters was 13.9% (RMSE for each parameter shown in parentheses).

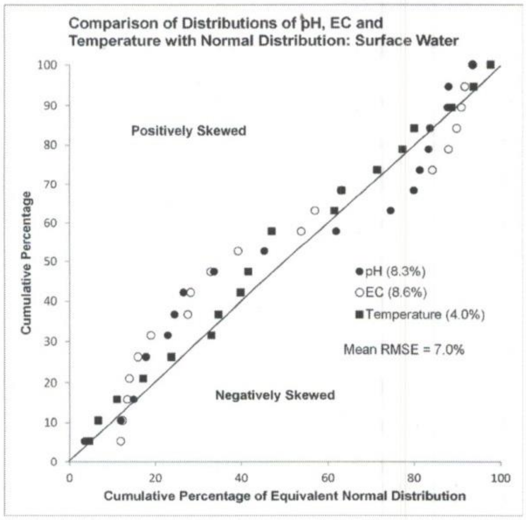

Fig. 4f: A comparison of the cumulative percentage of pH, electrical conductivity (EC), and temperature in surface water with the equivalent normal distribution with the same mean and standard deviation showed distributions to be a good fit to a normal distribution. The root mean square error (RMSE) averaged over all parameters was 7.0% (RMSE for each parameter shown in parentheses).

Table 2a: Description and arsenic concentrations of boring sampling sites.

As Rank [1]	As (mg/L)	Location	Age[2] (yrs)	Depth (m)	Screeen (m)	Latitude[3] (°N)	Longitude[3] (°E)
1	0.182	Pokhara Grande	15	162.5	120-141	28.1911850	83.9748725
2	0.161	Tang Be Boring Co.	5	100	—[4]	28.2119156	83.9787861
3	0.160[5]	Charak Hospital and Research Centre	—	—	—	28.2113458	83.9876594
4	0.040	Shangri-La Village	—	—	—	28.1862719	83.9752514
5	0.039	Gandaki Higher Secondary Boarding School	8	72	—	28.2584933	83.9688231
6	0.038	Fulbari Resort	14	210	145-200	28.1765258	84.0011444
7	0.033	Tibetan Refugee Camp	2	91.5	84.1-91.5	28.2097042	83.9847075
8	0.031	Shreejana Water Project	2	85	30-85	28.1797031	83.9870889
9	0.027	Hotel Fewa Prince	15	73.2	72.9-73.2	28.1971553	84.0196992
10	0.027	Kanchan Boring Co.	2	33.5	—	28.1870972	83.9819275
11	0.025	Fewa City Hospital and Research Centre	5	50	5-50	28.2065997	83.9820578
12	0.024	Himalayan Eye Hospital	11	220	—	28.1902289	83.9773714

[1]As Rank refers to the ranking of all boring samples from highest to lowest As concentration.[2]Ages are based upon interviews conducted in November 2010.[3]Latitude and longitude are based upon WGS 84 coordinate system.[4]Indicates unknown value.
[5]Arsenic values above the dashed line exceed the Nepal Interim As Standard (As = 0.05 mg/L).

Table 2b: Geochemistry of borings.

As Rank [1]	Mn (mg/L)	SO_4 (mg/L)	Fe (mg/L)	Cu (mg/L)	Ni (mg/L)	Co (mg/L)	Zn (mg/L)	Cr (mg/L)
1	0.0	13	0.02	0.03	0.002	0.043	0.13	0.00
2	0.1	11	0.00	0.01	0.000	0.010	0.06	0.01
3	0.0	20	0.02	0.00	0.000	0.038	0.05	0.01
4	0.1	5	0.05	0.02	0.001	0.033	0.07	0.01
5	0.1	31	0.01	0.25	0.002	0.025	0.10	0.01
6	0.1	43	0.00	0.02	0.002	0.034	0.08	0.01
7	0.0	13	0.02	0.11	0.001	0.034	0.31	0.01
8	0.1	10	0.02	0.04	0.001	0.034	0.67	0.01
9	0.2	23	0.07	0.02	0.000	0.039	0.08	0.01
10	0.0	24	0.01	0.03	0.001	0.034	0.07	0.01
11	0.2	13	0.02	0.06	0.000	0.003	0.09	0.01
12	0.2	14	0.02	0.02	0.000	0.037	0.10	0.00

[1]See Table 2a.

the geometric mean As concentration in either dug wells (As = 0.133 mg/L) or borings (As = 0.047 mg/L) were not statistically significant, indicating that no sedimentary geochemical processes are required to explain the levels of As in shallow groundwater (Fig. 6). On the other hand, Fe, Cu, Ni, and Zn were much higher in shallow groundwater compared with surface water, so that asking what sedimentary geochemical processes are causing release of Fe, Cu, Ni, and Zn from sediment into shallow groundwater are reasonable questions (Fig. 6). The increases in concentrations in As, Mn, Fe, Cu, Ni, Zn and Cr in dug wells relative to borings were statistically significant. The increase in sulfate concentration in borings relative to dug wells was also statistically significant (Fig. 6). The difference

Table 2c: On-site measurements of borings. EC-Electrical conductivity.

As Rank[1]	Temperature (°C)	pH	EC (µS/cm)
1	24.3	7.83	369
2	25.0	7.40	345
3	22.1	7.26	575
4	24.5	7.91	300
5	23.4	7.52	474
6	25.5	7.55	355
7	25.0	7.28	533
8	21.7	8.10	275
9	23.0	7.58	471
10	24.5	7.67	338
11	23.8	7.21	639
12	25.0	7.58	358

[1] See Table 2a.

between mean pH in dug wells (pH = 6.89) and borings (pH = 7.57) was statistically significant at the 99.9% confidence level, although differences between EC and temperature were statistically indistinguishable. The difference between surface water pH (7.82) and dug well pH (6.89) was also statistically significant at the 99.9% confidence level.

The reductive-dissolution and sulfide-oxidation models were tested by comparing the concentration of As in groundwater with concentrations of Fe, Mn and sulfate. Arsenic was uncorrelated with Fe ($R^2 = 0.15$), Mn ($R^2 = 0.02$) and sulfate ($R^2 = 0.06$), which is inconsistent with both models (Fig. 7). The reductive-dissolution model predicts a negative relation between As and sulfate due to the lack of sulfate under reducing conditions, while the sulfide-oxidation model predicts a positive relation between As and sulfate due to the abundance of sulfate that should result from the oxidation of sulfides and release of co-precipitated As. The sulfide-oxidation model predicts moreover a positive relation between As and between Fe or Mn or some other sulfide-forming transition element. Consideration

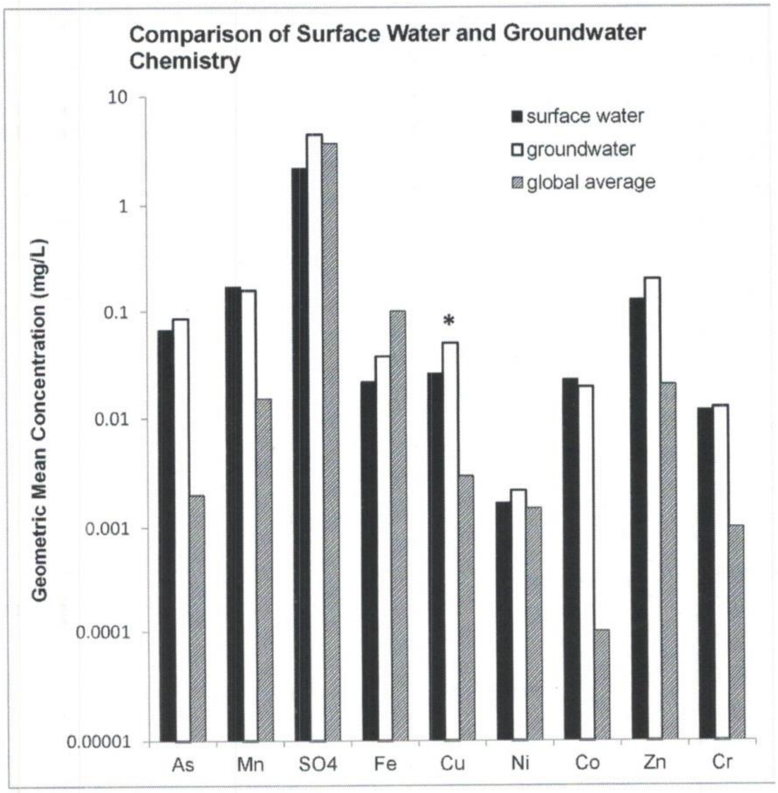

Fig. 5: The symbol * indicates differences between geometric mean concentrations in surface water and groundwater are statistically significant at the 95% confidence level according to the Student's t-test. The difference between concentration of As in surface water and groundwater is not statistically significant. The increase in concentration of Cu in groundwater relative to surface water is statistically significant. In both surface water and groundwater, concentrations of As, Mn, Co, Zn and Cr are elevated relative to global averages (Langmuir 1997). The global average for sulfate applies only to surface water as the groundwater global average is considerably higher (SO_4^{-2} = 30 mg/L) (Langmuir 1997).

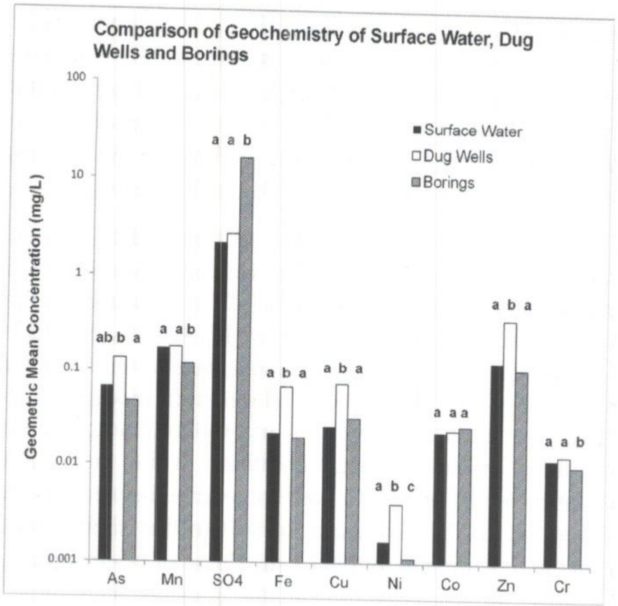

Fig. 6: For each parameter, differences between columns with the same letter are not statistically significant at the 95% confidence level according to the Student's t-test. The differences between As concentration in surface water and As concentration in either dug wells or borings are not statistically significant. The increases in concentrations in As, Mn, Fe, Cu, Ni, Zn and Cr in dug wells relative to borings are statistically significant. The increase in sulfate concentration in borings relative to dug wells is statistically significant.

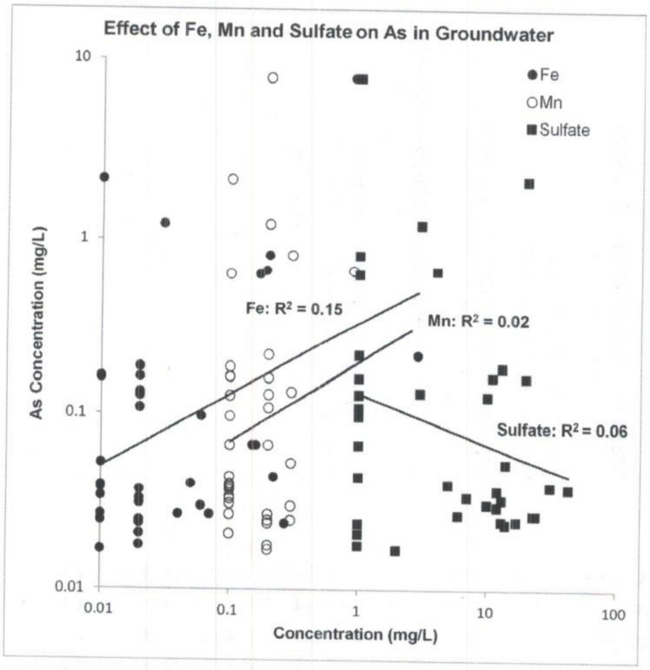

Fig. 7: Arsenic in groundwater is uncorrelated with Fe, Mn and sulfate, which is inconsistent with both the reductive-dissolution and sulfide-oxidation models.

of other transition elements or various molar sums of the five transition elements most commonly associated with As (Fe, Cu, Ni, Co, Zn) (Boyle and Jonasson 1973) did not improve the correlations, nor were correlations improved by separating groundwater samples into dug wells and borings. Although an increase in pH should release As by decreasing the number of positively-charged sorption sites, As was uncorrelated with pH, whether all groundwater samples were considered (R^2 = 0.005) or whether groundwater samples were separated into dug wells (R^2 = 0.003) and borings (R^2 = 0.01). Arsenic was similarly uncorrelated with pH (R^2 = 0.07) in surface water samples. The lack of correlation between pH and As makes it unlikely that elevated As results from the relatively alkaline environment of Pokhara Valley.

The spatial distribution of As concentrations was addressed by looking separately at dug wells, borings and surface water sites. Aside from the Lakeside cluster, there was wide variation within clusters (Fig. 8), which was consistent with the lack of spatial autocorrelation of As commonly seen in the floodplain of the Ganges River (Van Geen et al. 2003). Differences between geometric mean As concentrations of clusters were not statistically significant

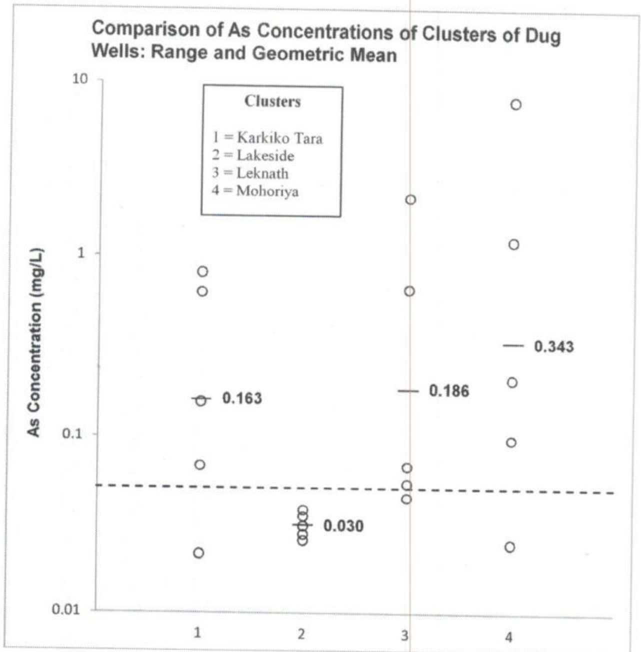

Fig. 8: Aside from the Lakeside cluster, there is wide variation in As concentrations within clusters of dug wells. The differences between geometric mean As concentrations of each cluster (indicated by labeled horizontal lines) are not statistically significant at the 95% confidence level according to the Student's t-test. The dashed line is the Nepal Interim As Standard (As = 0.05 mg/L).

Table 3a: Description, arsenic concentrations and on-site measurements of other groundwater sampling sites.

As Rank[1]	As (mg/L)	Location	Temperature (°C)	pH	EC (μS/cm)	Latitude[2] (°N)	Longitude[2] (°E)
1	0.152	Mahendra Gupha[3]	21.8	7.88	189.4	28.2720119	83.9797089
2	0.134	Guptesowr Mahadev Cave	23.4	7.10	178.0	28.1892672	83.9577153
3	0.133	Unnamed spring	20.8	8.56	130.0	28.2612189	84.0444953
4	0.129	Unnamed spring	23.1	6.14	78.7	28.1692081	84.0825511
5	0.126[4]	Unnamed spring	23.0	5.86	27.5	28.2881869	83.8613439
6	0.008	Unnamed spring	24.1	7.43	371	28.2440019	83.9961069

[1] As Rank refers to the ranking of all cave and spring samples from highest to lowest As concentration.

[2] Latitude and longitude are based upon WGS 84 coordinate system.

[3] Gupha is the Nepali word for cave. Names of caves follow Shangri-La Maps (2008).

[4] Arsenic values above the dashed line exceed the Nepal Interim As Standard (As = 0.05 mg/L).

at the 95% confidence level, so that there are no As "hot spots" in Pokhara Valley (Fig. 8). There did not appear to be any spatial pattern in As concentration of borings (Fig. 3), nor was depth dependence evident (Table 2a), aside from the greater As concentration in dug wells than in borings (Fig. 6). Surface water sites were separated into three watersheds: Phusre Khola upstream of the confluence with the Seti River, Seti River upstream of the confluence with Phusre Khola, and Bijayapur Khola upstream of the confluence with the Seti River, which accounted for all but the easternmost surface water site (Fig. 2). The increase in As concentration in both Phusre Khola and Bijayapur Khola over the Seti River was statistically significant at the 99% confidence level, while the difference between Phusre Khola and Bijayapur Khola was not statistically significant (Fig. 9). The differences in As concentrations are consistent with the origins of the rivers. Phusre Khola and Bijayapur Khola originate within Pokhara Valley in the low-grade metamorphic rocks (phyllites) of the Kuncha Formation (Lesser Himalayan Zone), while the Seti River originates from outside Pokhara Valley in the high-grade metamorphic rocks of the Himalayan Gneiss (Higher Himalayan Zone) (Koirala et al. 1998). Although there have been no measurements of As concentrations in rocks from these formations, As concentrations tend to be much greater in slates and phyllites (mean As = 18 mg/kg) than in schists and gneisses (mean As = 1.1 mg/kg) (Smedley and Kinniburgh 2002; Emerman et al. 2007). High-grade metamorphic rocks have the lowest As concentrations of all common rock groups, even lower than carbonates (mean As = 2.6 mg/kg) (Smedley and Kinniburgh 2002; Emerman et al. 2007). There were no other statistically significant differences in chemistry among the three watersheds, aside from the increase in sulfate in the Seti River relative to Phusre Khola. It was not possible to compare shallow groundwater As concentrations among watersheds as there were no dug wells in the watershed of the Seti River.

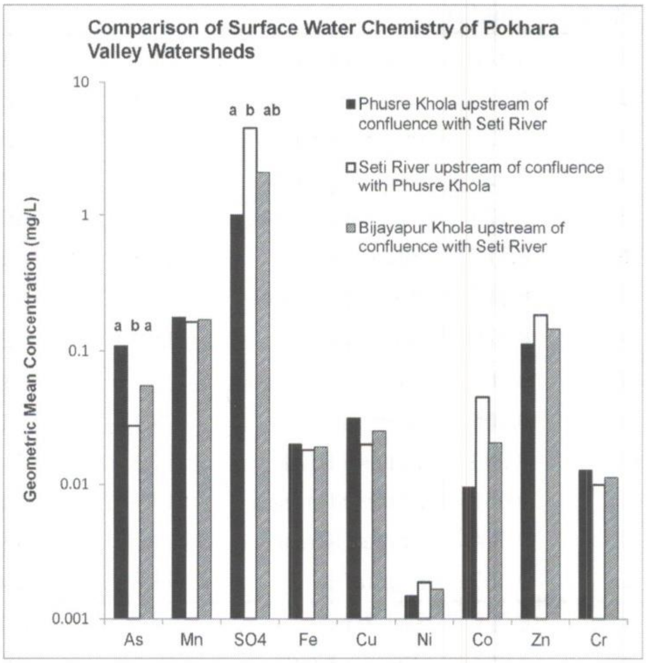

Fig. 9: For each parameter, differences between columns with the same letter are not statistically significant at the 95% confidence level according to the Student's t-test. The increase in As concentration in the Phusre Khola and Bijayapur Khola relative to the Seti River, upstream of the confluence with Phusre Khola, is statistically significant, which is consistent with the higher As concentrations in the low-grade metamorphic rocks of the Lesser Himalayan Zone drained by Phusre Khola and Bijayapur Khola, as opposed to the lower As concentrations in the high-grade metamorphic rocks of the Higher Himalayan Zone drained by the Seti River.

Table 3b: Geochemistry of other groundwater sites.

As Rank [1]	Mn (mg/L)	SO_4 (mg/L)	Fe (mg/L)	Cu (mg/L)	Ni (mg/L)	Co (mg/L)	Zn (mg/L)	Cr (mg/L)
1	0.3	3	0.02	0.02	0.000	0.041	0.04	0.01
2	0.2	0	0.02	0.09	0.000	0.017	0.07	0.02
3	0.1	10	0.02	0.13	0.000	0.005	0.23	0.00
4	0.2	0	0.02	0.04	0.000	0.000	0.09	0.20*
5	0.2	0	0.02	0.00	0.000	0.006	0.17	0.01
6	0.2	2	0.01	0.00	0.000	0.000	0.19	0.01

[1] See Table 3a.

*Concentration exceeds the WHO Standard (Mn = 0.4 mg/L, Cu = 2 mg/L, Ni = 0.07 mg/L, Cr = 0.05 mg/L).

Table 4a: Description, arsenic concentrations and on-site measurements of surface water sampling sites. EC-Electrical conductivity.

As Rank[1]	As (mg/L)	Location	Temp. (°C)	pH	EC (µS/cm)	Latitude[2] (°N)	Longitude[2] (°E)
1	0.152	Kahu Khola[3]	21.1	8.06	72.1	28.2164508	84.0189322
2	0.134	Ambote Khola	21.7	7.10	113.8	28.1824753	83.9525864
3	0.133	Phusre Khola	20.3	7.10	85.4	28.1845858	83.9516317
4	0.129	Khahare Khola	19.5	9.00	29.8	28.2469153	83.8768725
5	0.126	Bijayapur Khola	20.0	8.73	204.0	28.2168886	84.0303639
6	0.117	Begnas Tal	24.8	7.33	38.7	28.1668517	84.0927606
7	0.116	Bijayapur Khola	17.2	8.59	213.0	28.1941858	84.0276447
8	0.115	Seti River	18.9	8.34	183.6	28.2915914	83.9340014
9	0.107	Phewa Lake outlet	22.1	7.01	132.2	28.1790164	83.9700108
10	0.105	Phewa Lake	23.1	7.28	61.9	28.2108917	83.9529914
11	0.093	Guheko Khola	22.3	7.49	21.5	28.2449636	83.9165875
12	0.087	unnamed stream	18.4	7.24	17.3	28.2896961	83.8686217
13	0.074	Harpan Khola	21.2	6.90	15.8	28.2438953	83.8759331
14	0.067[4]	Khaste Tal	23.8	6.40	60.5	28.1932522	84.0515267
15	0.029	Seti River	17.8	8.74	223.0	28.2134181	83.9945611
16	0.025	Bhalam Khola	19.6	8.58	120.0	28.2423147	83.9963542
17	0.016	Seti River	16.8	8.48	209.0	28.2411586	83.9947214
18	0.015	Garlang Khola	20.3	8.52	196.5	28.2459761	83.9959761
19	0.008	Ghobang Khola	19.9	7.73	23.3	28.2882794	83.8605561

[1] As Rank refers to the ranking of all surface water samples from highest to lowest As concentration.

[2] Latitude and longitude are based upon WGS 84 coordinate system.

[3] Khola and tal are the Nepali words for river and lake, respectively. Names of surface water bodies follow Shangri-La Maps (2008).

[4] Arsenic values above the dashed line exceed the Nepal Interim As Standard (As = 0.05 mg/L).

DISCUSSION

Geometric mean As concentrations were higher for Pokhara Valley than for Kathmandu Valley both for surface water (As = 0.067 mg/L in Pokhara Valley vs. As = 0.013 mg/L in Kathmandu Valley) and groundwater (As = 0.086 mg/L in Pokhara Valley vs. As = 0.015 mg/L in Kathmandu Valley), which is consistent with the difference in bedrock

Table 4b: Geochemistry of surface water sites.

As Rank [1]	Mn (mg/L)	SO$_4$ (mg/L)	Fe (mg/L)	Cu (mg/L)	Ni (mg/L)	Co (mg/L)	Zn (mg/L)	Cr (mg/L)
1	0.0	0	0.06	0.02	0.000	0.000	0.07	0.02
2	0.2	0	0.01	0.02	0.001	0.008	0.08	0.01
3	0.1	0	0.03	0.03	0.005	0.014	0.29	0.01
4	0.1	0	0.03	0.04	0.000	0.002	0.07	0.03
5	0.0	4	0.03	0.02	0.001	0.030	0.04	0.01
6	0.5*	0	0.02	0.04	0.003	0.109	0.15	0.01
7	0.2	17	0.02	0.06	0.000	0.029	0.08	0.02
8	0.1	19	0.02	0.01	0.000	0.000	0.17	0.01
9	0.2	0	0.02	0.12	0.001	0.037	0.10	0.01
10	0.3	0	0.02	0.03	0.003	0.009	0.12	0.01
11	0.2	0	0.02	0.02	0.001	0.024	0.23	0.02
12	0.2	0	0.01	0.01	0.004	0.159	0.14	0.01
13	0.2	0	0.02	0.02	0.000	0.004	0.05	0.01
14	0.2	0	0.02	0.01	0.006	0.137	0.20	0.01
15	0.0	18	0.02	0.02	0.001	0.025	0.03	0.01
16	0.2	0	0.18	0.34	0.002	0.140	0.59	0.01
17	0.1	22	0.01	0.00	0.003	0.170	0.21	0.01
18	0.2	6	0.00	0.02	0.001	0.027	0.67	0.01
19	0.3	0	0.01	0.01	0.003	0.152	0.11	0.01

[1] See Table 4a.

*Concentration exceeds the WHO Standard (Mn = 0.4 mg/L, Cu = 2 mg/L, Ni = 0.07 mg/L, Cr = 0.05 mg/L).

geology between the two tectonic valleys. Of the seven rivers studied in Kathmandu Valley by Emerman (2005), four originate in the Sheopuri Gneiss, two in the Chandragiri Formation (limestones), and only one in the low-grade metamorphic rocks of the Tistung Formation (phyllites and slates) (Shrestha et al. 1998). In fact, the geometric mean As concentration for the Seti River watershed alone (As = 0.028 mg/L) was not much greater than the geometric mean As concentration for Kathmandu Valley and the difference was not statistically significant at the 95% confidence level. A lack of spatial clustering of groundwater As concentrations was found in Kathmandu Valley with regard to both surface location and depth (Emerman et al. 2010), just as in Pokhara Valley. The difference between mean surface water pH in Pokhara Valley (pH = 7.82) and Kathmandu Valley (pH = 7.31) (Emerman 2005) was statistically significant at the 99% confidence level, which is consistent with the abundance of carbonate sediments in Pokhara Valley (Koirala and Rimal 1996; Koirala et al. 1997).

The results of this study are consistent with the fluvial recharge model for As contamination of groundwater in the following ways:

1. There is elevated As in rivers. There is higher As in rivers that originate in bedrock that tends to have higher As concentrations.
2. Fluvial As is uncorrelated with pH, which is consistent with a lack of As sorption / desorption in overland flow or streambeds.
3. Fluvial As concentrations are statistically indistinuishable from groundwater As concentrations.
4. There are no spatial patterns in groundwater As concentrations, which is consistent with randomly shifting losing streams (streams that recharge groundwater).
5. Groundwater As is uncorrelated with pH, sulfate or any of the transition elements either individually or in combination, which is consistent with a lack of reductive dissolution, sulfide oxidation or any other groundwater As chemistry.

The central feature of the fluvial recharge model is that As acts almost as a passive tracer with almost no interaction with sediment. The question is then: Why is there no As chemistry in the particular sedimentary basins of Pokhara Valley and Kathmandu Valley? We suggested that fastmoving aqueous As does not sorb onto fluvial or hillslope sediment because there is insufficient residence time for sorption of the large, divalent arsenate oxyanion. However, elevated As was found at all four of the lake sampling sites (Table 4a). By contrast, there does seem to be cation-sediment interaction. The increase in shallow groundwater concentrations of Fe, Cu, Ni, and Zn relative to surface water (Fig. 6) is consistent with the decrease in negatively-charged sorption sites that would occur under the more acidic conditions of shallow groundwater. The real mystery is the discrepancy between As chemistry and the chemistry of sulfate, another large, multivalent oxyanion, although not as large as the arsenate oxyanion. Sulfate is moderately correlated with pH for both groundwater ($R^2 = 0.38$) and surface water ($R^2 = 0.38$) with nearly identical positive slopes with statistical significance at the 99.99% and 99% confidence levels, respectively (Fig. 10), which is consistent with the decrease in positively-charged sorption sites that would occur with an increase in pH. Hopefully, these observations can motivate further research into the thermodynamics and kinetics of As-sediment interactions.

The obvious question at this point is whether the residents of Pokhara Valley show detrimental health effects from elevated As. We are not aware of any reports of arsenicosis or any health surveys searching for arsenicosis in either Pokhara Valley or Kathmandu Valley or anywhere in Nepal outside of the Terai Zone. It is quite possible that there has not been enough time for detrimental health effects to develop as groundwater exploitation is very recent in Pokhara Valley. Detrimental health effects from As are, in fact, much more likely in Kathmandu Valley, where there has been widespread exploitation of groundwater for centuries. In Pokhara Valley we found only five dug wells constructed in 2000 or earlier, only two of which were used for drinking water (As Ranks #8 and #19 in Table 1a), only one of which exceeded the Nepal Interim As Standard. The oldest boring that supplied water to local residents (Tang Be Boring Co.) was constructed in 2005 (Table 2a). Any older borings, the oldest of which was constructed in 1995, supplied water to resorts, hospitals and a private school. (Even a boring for a school will not provide the majority of the drinking water of its consumers for an extended period.) We are not aware of any data on historical water usage in Pokhara Valley, but if the main historical source of water has been surface water from the watershed of the Seti River upstream from the confluence with Phusre Khola (Figs. 2 and 9) with only very recent exploitation of other water sources, we would not be expect to see detrimental health effects from As at the present time. The municipal water for Pokhara City, which is the main water source of the majority of the residents of Pokhara Valley, currently comes from Yangdi Khola (Fig. 2), which is within the Seti River watershed, although it originates in the low-grade metamorphic rocks of the Kuncha Formation (Koirala et al. 1998). Unfortunately, no samples of Pokhara municipal water were collected. We do not mean to cause alarm, but As levels in drinking water in Pokhara Valley need to be monitored. At a minimum, we recommend a repeat of this study in 2015.

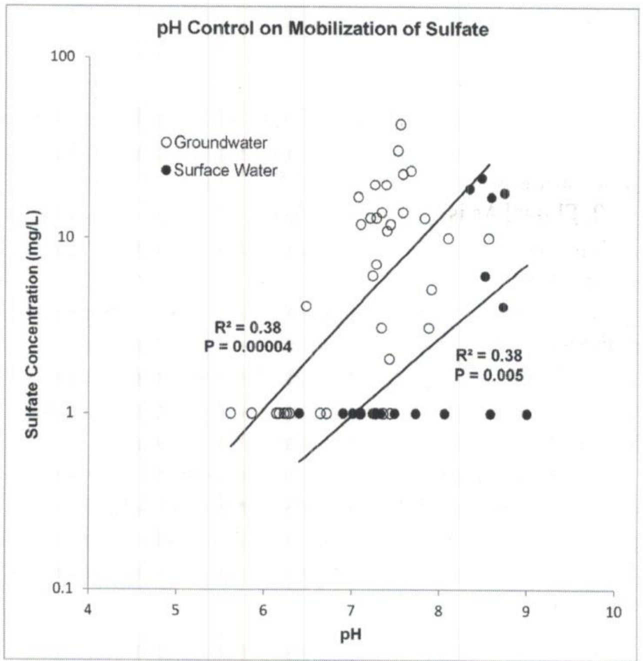

Fig. 10: Sulfate is moderately positively correlated with pH in both groundwater and surface water, which is consistent with a pH desorption model.

CONCLUSIONS

The main conclusions of this and related studies are:

1. Elevated As in groundwater does not occur only in the Terai Zone of Nepal, but also in the tectonic valleys of Pokhara and Kathmandu.

2. The reductive-dissolution model, the dominant paradigm for As contamination of groundwater in south Asia, which does not assign responsibility to any human activity, is not supported by the geochemical data from Pokhara Valley or Kathmandu Valley. The geochemical data do support the fluvial recharge model in which As contamination of groundwater results from accelerated erosion, which could be due to a combination of tectonic uplift, monsoon climate and deforestation.

Further research needs to be carried out on surface water and groundwater As in the tectonic valleys of the Himalaya with special attention to the relation between As contamination and deforestation or overgrazing. Extensive water sampling was conducted in Mustang Valley in May 2011 and will be reported in a follow-up paper. Other future study sites include Dehra Dun Valley and Har Ki Dun Valley of India Himalaya and Puna Tsang Chhu Valley of Bhutan Himalaya. The possibility has been raised that "safe" watersheds could be identified based on the bedrock geology of the river headwaters. This means that As concentrations in rocks of key formations in Nepal need to be measured, instead of relying solely on global averages for rock types.

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