

Arsenic and other heavy metals in the Sunkoshi and Saptakoshi Rivers, eastern Nepal

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ABSTRACT

Since previous research had shown that elevated fluvial As occurs throughout Nepal outside of the Higher Himalayan Zone, the objective of this study was to determine whether elevated fluvial As occurs throughout the Ganges River system or, if not, where in the river system As adsorbs onto sediments. Samples were collected during an 11-day 288-km rafting trip down the Sunkoshi River to the Saptakoshi River from Dolalghat to Koshi Barrage. Water samples were collected from the Sunkoshi and Saptakoshi rivers twice daily and from five major tributaries, while sediment samples were collected from sand bars daily. Dissolved concentrations of As, Fe, Cu, Ni, Co, Mn, Zn and Cr were measured with a spectrophotometer. Mobile sediment As was measured by extraction with 1 M HCl. Dissolved transition elements were elevated both upstream of and within the Terai Zone (Indo-Gangetic Plain), while dissolved As was elevated upstream of the Indo-Gangetic Plain, but became uniformly undetectable within the Indo-Gangetic Plain. There was a moderate correlation ($R^2 = 0.44$) between mobile sediment As and downstream distance. The fluvial behavior of As can be related to the large mass, volume and valency of the As oxyanion so that, when river velocity is high, the As oxyanion does not have sufficient residence time in the vicinity of sorption sites to attach to those sites. The implication of this study is that it may be possible to address As contamination of groundwater by addressing deforestation only in the tectonic valleys upstream of the Indo-Gangetic Plain.

Keywords: Arsenic, fluvial, deforestation, Saptakoshi River, Sunkoshi River, Nepal

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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 eight years have documented that the region of As contamination extends into the Terai Zone, the Indo-Gangetic Plain of southern Nepal (Neku and Tandukar 2002; Bhattacharya et al. 2003; Shrestha et al. 2003; ENPHO and USGS 2004; Kanel et al. 2005; Tandukar et al. 2005; Brikowski et al. 2006; Neku et al. 2006; Panthi et al. 2006; Neku and Brikowski 2009; Pokhrel et al. 2009) and even into Kathmandu Valley, a heavily populated tectonic valley well upstream of the Indo-Gangetic Plain (Khatiwada et al. 2002; Gurung et al. 2006; Bajracharya et al. 2007; Warner et al. 2008; Maharjan et al. 2009; Chapagain et al. 2009; Emerman et al. 2010) (Fig. 1). According to the most recent published count, 25,058 tubewells in the Terai Zone have been tested for As, of which 5,686 tubewells (22.7%) exceed the WHO (World Health Organization) As Standard (As = 0.01 mg/L) and 1,916 tubewells (7.6%) exceed the Nepal Interim As Standard (As = 0.05 mg/L) (Panthi et al. 2006). Pokhrel et al. (2009) estimated that there were perhaps 200,000 tubewells in the Terai Zone and that 3.5

million Nepalis had no access to drinking water that did not exceed the WHO As Standard. Most recently, 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. 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 impact 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).

Possible responses to As contamination of groundwater could be placed into the following five categories:

- 1) Treatment could be provided to those members of the local population who show symptoms of arsenicosis,
- 2) The health of the local population could be improved, especially with regard to nutrition, so that people who drink As-contaminated water do not develop symptoms of arsenicosis,
- 3) The water from tubewells that are contaminated with As could be treated so as to remove the As,

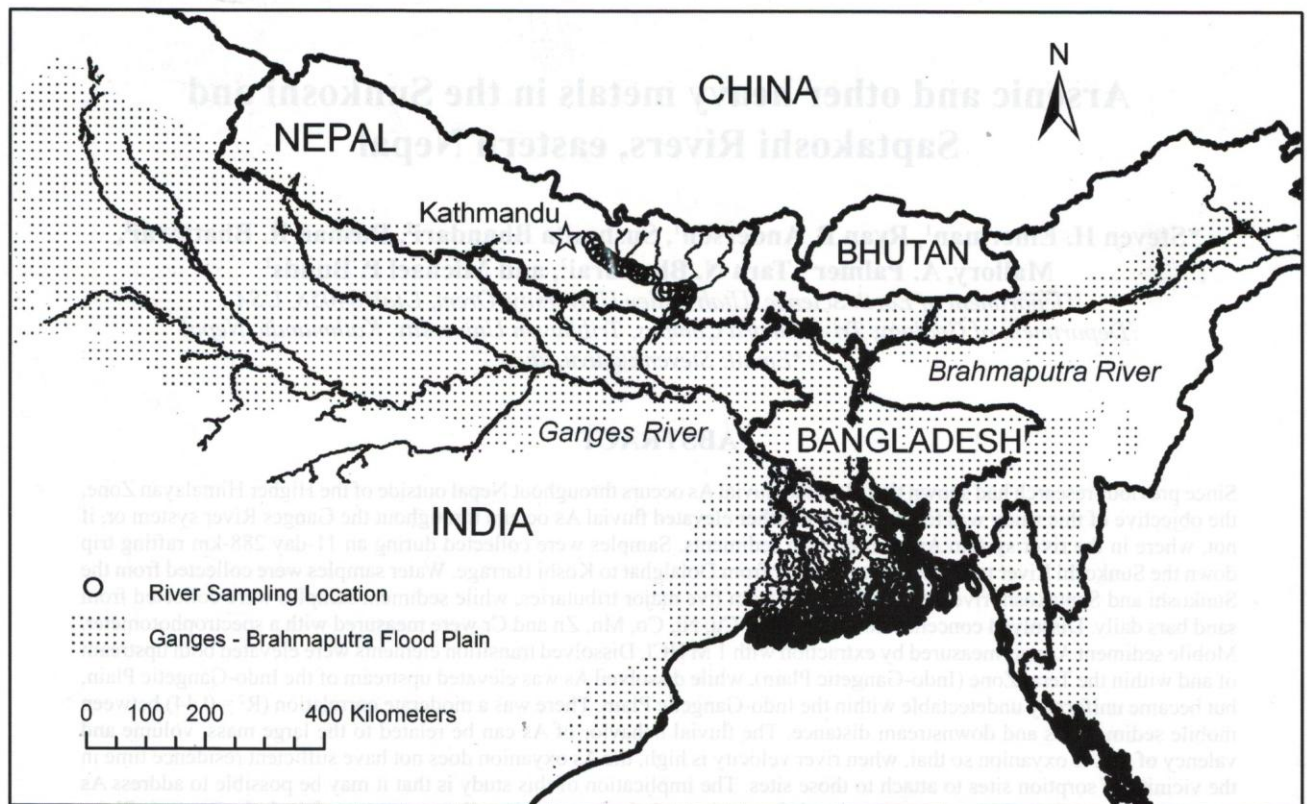


Fig. 1: River sampling locations in the context of the watershed of the Ganges River (Hearn et al. 2001). Not all tributaries of the Ganges River in Nepal are shown in order to emphasize the Sunkoshi – Saptakoshi River system.

4) Tubewells could be drilled into aquifers that are not contaminated with As (or in other ways the local population could be provided with water that is not contaminated with As),

5) Steps could be taken to prevent further contamination of groundwater with As.

Most current work is in Categories #3 and #4, either developing more effective and less costly means of removing As from groundwater (Neku and Tandukar 2003) or searching for the aquifers that are not contaminated with As (Brikowski et al. 2004, 2005; Winkel et al. 2008). Category #5 cannot be addressed without knowledge of the cause of As contamination.

It has been generally agreed for a long time that the As contamination of groundwater is too widespread to be due to direct human activities such as mining, smelting or the use of As-based pesticides (Aswathanarayana 1997). The following seven models have been proposed for the release of As into groundwater in the Indo-Gangetic Plain, some of which involve indirect human activity:

1) 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).

2) Excessive use of phosphate fertilizers has resulted in displacement of As from sediment sorption sites by phosphate (Acharyya et al. 1999, 2000).

3) Arsenic that is co-precipitated with diagenetic carbonate concretions is released into groundwater upon dissolution of the carbonate concretions under acidic conditions (Shanker et al. 2001).

4) Arsenic is displaced from sorption sites by carbonate after sediments deposited in surface waters with low carbonate concentration are later exposed to groundwater with high carbonate concentration (Appelo et al. 2002).

5) The loss of healthy forests and grasslands causes the loss of the microbial population that sequesters As in soil so that As is leached into groundwater (Emerman 2004; Emerman et al. unpub.).

6) In the strongly reducing conditions of the thick sedimentary package of the Indo-Gangetic Plain, As is released 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).

7) Groundwater is recharged by losing streams with elevated As due to rapid erosion caused by monsoon climate, tectonic uplift and deforestation (fluvial recharge model) (Emerman et al. 2007, 2010).

The reductive-dissolution model (Model #6) has gradually become the dominant paradigm in As studies in South Asia. 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. It is, of course, necessary to continually re-evaluate all of the above models in light of every new set of data that is collected. For example, Williams et al. (2004, 2005) has argued that As contamination in the Terai Zone could result from sulfide oxidation (Model #1) 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. Emerman et al. (2010) have shown that groundwater As in Kathmandu Valley is uncorrelated with Fe, Mn (indicative of reducing conditions), or any combination of the sulfide-forming transition elements, which is inconsistent with both the reductive-dissolution and sulfide-oxidation models.

What the first six 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) 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. By contrast, the global average fluvial As is in the range 0.00013 - 0.0021 mg/L (Smedley and Kinniburgh 2002). Emerman et al. (2007) 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 the first six models could be thermodynamically plausible, the kinetics could be sufficiently slow that the groundwater chemistry simply reflects the surface water chemistry. Although several studies have examined the effects of groundwater recharge on As-enriched aquifers (Van Geen et al. 2006; Harvey et al. 2006; Mukherjee et al. 2007; Saha et al. 2011), we are aware of only one other study (Yamasaki and Hata 2000) that has related the dissolved As in a losing stream to the dissolved groundwater As in the aquifer that is recharged by the losing stream.

The basis of the fluvial recharge model (Model #7) is that 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 aqueous form predominantly as the oxyanion AsO_4^{3-} . 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 the other common oxyanions nitrate and phosphate. Since all ions in overland flow are traveling 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 trivalent As oxyanion cannot attach to three positively-charged sorption sites unless it has sufficient residence time in the vicinity of the sorption sites for all three sites to become simultaneously vacant, for example, by diffusion of three monovalent chloride ions from sorption sites into water. Moreover, the large volume of the As oxyanion implies that the region of the three sorption sites must be sufficiently large for the As oxyanion to attach. When large regions of positively-charged sorption sites are scarce, an As oxyanion that is carried along a sediment surface downstream from the vicinity of one large region will not likely be carried into the vicinity of another large region. 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 be adsorbed onto sediment that is deposited within the watershed. Naturally occurring elevated levels of fluvial As have been reported elsewhere in the world, although they have been attributed to input of geothermal waters (McLaren and Kim 1995; Robinson et al. 1995; Nimick et al. 1998; Wilkie and Hering 1998) or evaporative concentration of baseflow-dominated rivers in arid areas (Cáceres et al. 1992; Lerda and Proserpi 1996) (see discussion in Smedley and Kinniburgh (2002)). It is possible that elevated fluvial As in Nepal results from a "perfect storm" of monsoon climate, tectonic uplift and deforestation.

Despite the importance of As to public health in south Asia, measurements of fluvial As or As adsorbed onto fluvial sediment have been remarkably scant. Smedley and Kinniburgh (2002) reported on seven fluvial samples from Bangladesh with As concentrations in the range <0.0005 –

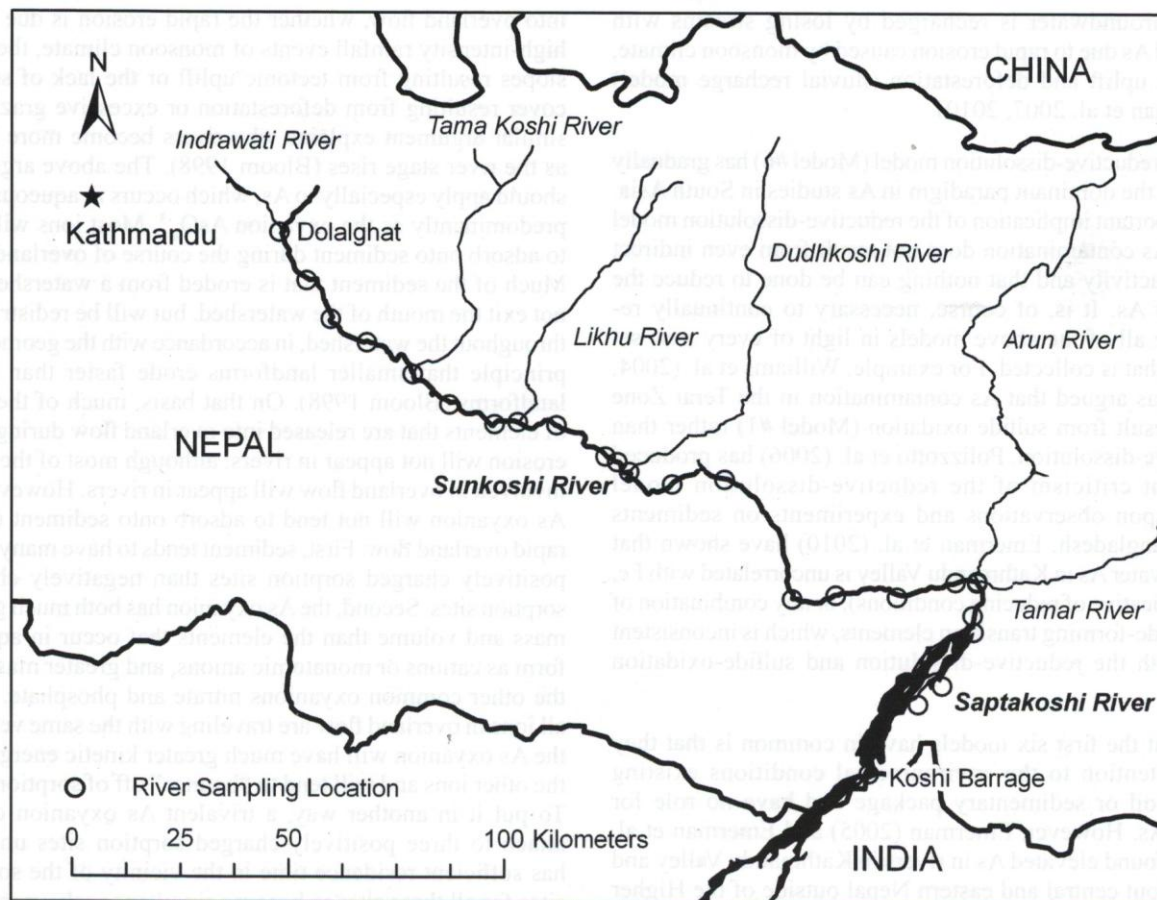


Fig. 2: Close-up of river sampling locations from Dolalghat to Koshi Barrage. The Terai Zone (Indo-Gangetic Plain) occurs downstream of the confluence of the Sunkoshi, Arun and Tamar rivers to create the Saptakoshi River.

0.0027 mg/L with one sample having a high concentration of 0.029 mg/L. Pandey et al. (2002) investigated high levels of groundwater As in Rajnandgaon District, Chhattisgarh State, India, outside the floodplain of the Ganges River, and measured fluvial As in the Shivnath River whose catchment lies in the region with high groundwater As. Pandey et al. (2002) found that fluvial As was undetectable until it began to rise with the onset of the monsoon in June, reached a maximum in July at 0.060 mg/L, and returned to undetectable by October. Elevated fluvial As during the monsoon may be related to elevated levels of fluvial organic matter and organic complexation of As (Emerman 2005). Aside from studies of As in river bed sediments in Bangladesh (Datta and Subramanian 1997), India (Subramanian et al. 1987; Chakravarti 1997; Pal et al. 2008) and Pakistan (Halfpenny and Mazzucchelli 1999), we are not aware of any other studies of fluvial As in south Asia outside of Nepal. Bhatt and Gardner (2009) have also measured fluvial As in rivers in Kathmandu Valley. Other studies of fluvial chemistry in Nepal that did not measure As include Collins and Jenkins (1996), Sharma (1997), Karn and Harada (2001), Sharma et al. (2005), and Bhatt and McDowell (2007).

The objective of this study was to determine whether elevated fluvial As occurs throughout the Ganges River system or, if not, where As either adsorbs onto sediment or co-precipitates with sulfides. The objective was carried out by collecting water samples while whitewater rafting down the Sunkoshi River to the Saptakoshi River from Dolalghat to Koshi Barrage (Figs. 1 and 2) and analyzing samples for As and the transition elements commonly associated with As (Boyle and Jonasson 1973) (English names and spellings follow DS (2002)). This route was chosen because it is the longest commercial whitewater route in Nepal and crosses the major geological zones of the Higher Himalayan Zone, Lesser Himalayan Zone, Siwalik Group and Terai Zone (Indo-Gangetic Plain) (Dahal 2006; Emerman et al. 2007). The most important geological and topographical boundary in Nepal occurs between the Siwalik Group (foothills of the Himalaya) and the Indo-Gangetic Plain. Along the rafting route this boundary occurred at the confluence of the Sunkoshi, Arun and Tamar rivers to create the Saptakoshi River. Although we wished to continue collecting water samples along the Saptakoshi River as it entered India, this was not a legal border crossing.

MATERIALS AND METHODS

Water samples were collected twice daily from the Sunkoshi and Saptakoshi rivers during the 11-day 288-km rafting trip from Dolalghat to Koshi Barrage (Fig. 2). Water samples were also collected from the major tributaries Tama Koshi River, Likhu River, Dudhkoshi River, Arun River and Tamar River (Fig. 2), just upstream of their confluence with the Sunkoshi River, for a total of 27 water samples. Electrical conductivity (EC), pH and temperature were measured on site with the Oakton PCSTestr 35. Water was forced through a 0.45- μ m syringe filter into two 250-mL polyethylene containers with the contents of one container used for measurement of As and the other used for all other heavy metals. Trace metal grade concentrated nitric acid was added to the container for arsenic analysis to reduce pH < 2 (Saunders 1998). Concentrations of Fe, Cu, Ni, Co, Mn, Zn and Cr were measured the same day using the Hach DR-2700 Spectrophotometer. Water samples were returned to Utah Valley University for measurement of As using the silver diethyldithiocarbamate method (U.S. Environmental Protection Agency Standard Method 3500-As) with the Hach DR-2700 Spectrophotometer. The spectrophotometer was re-calibrated after every 15 samples using three standard solutions with As concentrations of 0.020 mg/L, 0.040 mg/L and 0.200 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, and 0.1 mg/L for Mn.

About 500 g of sediment was collected from sand bars daily for a total of 11 samples. Samples were returned to Utah Valley University for analysis. It was desired to compare mobile As in the same size fraction from each sediment sample. We chose to measure mobile As in the 125–250 μ m size fraction as seven samples had abundant sand in that size range. The remaining four sediment samples were not analyzed as insufficient sand was present in the 125–250 μ m size range. Mobile sediment As was measured by adding 10 g of air-dried sediment to 250 mL of 1 M HCl and stirring for 60 minutes (Emerman 2004; Emerman et al. unpub.). The mixture was filtered and the filtrate was measured for As using the same procedure as for water samples. There is presently no consensus in the soil or sediment chemistry literature as to the procedure for measuring mobile soil or sediment As (Alam et al. 2001; Cai et al. 2002). The most elegant study was carried out by Keon et al. (2001) who tested a sequential As extraction procedure by synthesizing the various pools of solid-phase As. Keon et al. (2001) showed that 1 M MgCl₂ (2 hours, two repetitions) extracted the ionically bound As and 1 M NaH₂PO₄ (16 and 24 hours, one repetition of each time duration) extracted the strongly adsorbed As. The next step of 1 M HCl (1 hour, one repetition) extracted the As co-precipitated with acid-volatile sulfides, carbonates, Mn oxides, and very amorphous Fe oxyhydroxides. Successively more aggressive extractants were used until hot 16 M HNO₃ and 30% H₂O₂ were used to extract As from orpiment and other recalcitrant As minerals. Langston (1980) and Reuther (1992) also used extraction with 1 M HCl as a measure of bioavailable soil As.

RESULTS

It is crucial to test for the existence of normal distributions prior to using any statistical tests that assume normal distributions. For each element, the mean and standard deviation were calculated and the cumulative percentage for each concentration (percentage of concentrations equal to or less than that concentration) were compared with the cumulative percentage for a normal distribution with the same mean and standard deviation. None of the elements followed a normal distribution (Fig. 3a). The concentrations tended to be very positively skewed so that most of the concentrations were low, but with a long tail of higher concentrations. On the other hand, comparing the cumulative percentage of each concentration of each element with the cumulative percentage of the equivalent lognormal distribution (same mean and standard deviation as the set of logarithms of concentrations) showed that for all elements a lognormal distribution was a better fit than a normal distribution (Fig. 3b). The use of a lognormal distribution requires an assumption about the zero concentrations (concentrations below the detection limit). We set all zero concentrations equal to the detection limit. There were ten measured zero concentrations for As, four for Mn and Ni, three for Cu and Cr, and one for Co. On the above basis, we decided to carry out all statistical comparisons of elemental concentrations on the logarithms of concentrations with concentrations below the detection limit set equal to the detection limit.

The non-elemental measurements (pH, electrical conductivity, water temperature) showed a different statistical behavior. Electrical conductivity and water temperature followed a normal distribution (Fig. 3c), which was not improved by plotting the logarithms of measurements (Fig. 3d). Measurements of pH were positively skewed (Fig. 3c) and were still positively skewed when the logarithms of pH were plotted. All statistical comparisons of the non-elemental measurements were carried out assuming the measurements followed a normal distribution. The results of the statistical comparisons were sufficiently unambiguous that there was no point in considering statistical methods that did not assume any underlying distribution. All statistical comparisons were made using the Student's t-test on either the measurements or their logarithms.

The major comparison was between measurements made in the Saptakoshi River (Indo-Gangetic Plain) and those made in the tributaries of the Saptakoshi River (upstream of the Indo-Gangetic Plain). Measurements of pH, EC and temperature were remarkably similar in both the Indo-Gangetic Plain and upstream from the Indo-Gangetic Plain (Table 1). Arsenic was considerably lower in the Saptakoshi River than its tributaries with the difference between the geometric mean concentrations being statistically significant at the 99.9% confidence level (Fig. 4). No differences between the geometric mean concentrations of any transition element in the Saptakoshi River and its tributaries were statistically significant at the 95% confidence level (Fig. 4). Except for As in the Saptakoshi River, concentrations of As

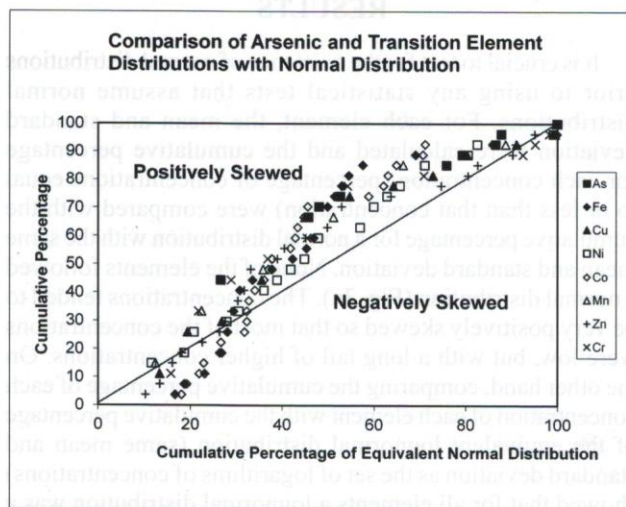


Fig. 3a: For each measured concentration of As, Fe, Cu, Ni, Co, Mn, Zn and Cr, the percentage of measured concentrations less than or equal to that value is plotted against the percentage that would result from a normal distribution with the same mean and standard deviation as the set of measured concentrations. None of the elements follow a normal distribution.

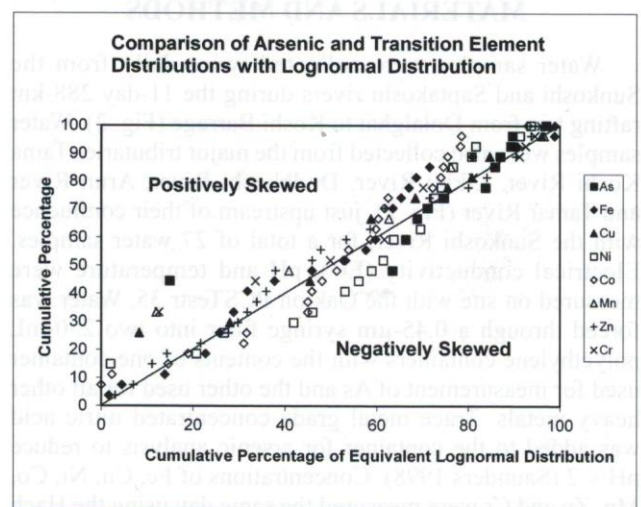


Fig. 3b: For each measured concentration of As, Fe, Cu, Ni, Co, Mn, Zn and Cr, the percentage of measured concentrations less than or equal to that value is plotted against the percentage that would result from a lognormal distribution with the same mean and standard deviation as the set of logarithms of measured concentrations. Values below the detection limit are set equal to the detection threshold. For all elements, a lognormal distribution is a better fit than a normal distribution.

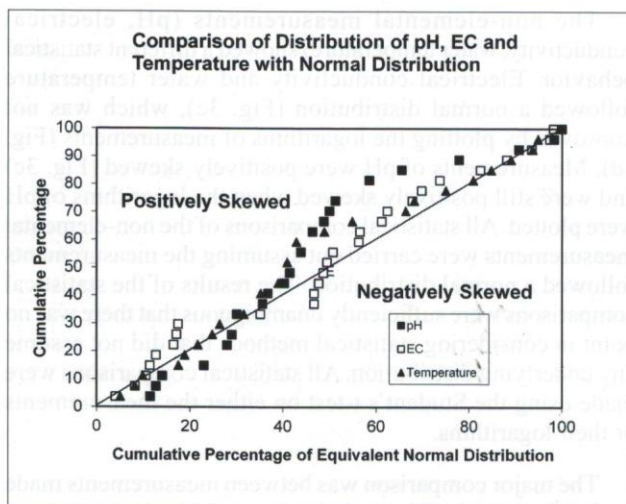


Fig. 3c: For each measurement of pH, electrical conductivity (EC) and temperature, the percentage of measurements less than or equal to that value is plotted against the percentage that would result from a normal distribution with the same mean and standard deviation as the set of measurements. Electrical conductivity and temperature follow a normal distribution, while pH is positively skewed.

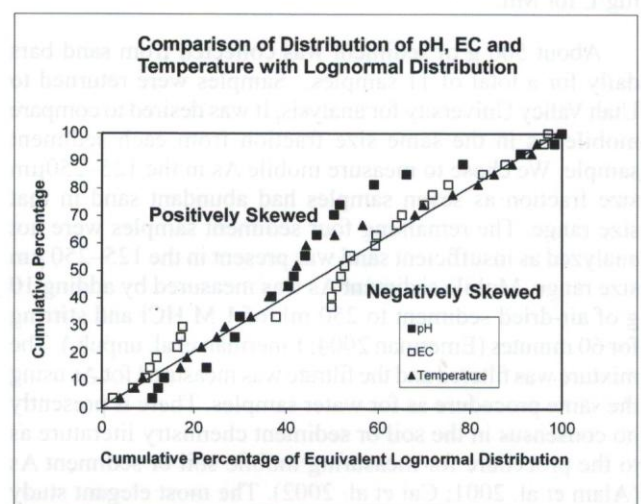


Fig. 3d: For each measurement of pH, electrical conductivity (EC) and temperature, the percentage of measurements less than or equal to that value is plotted against the percentage that would result from a lognormal distribution with the same mean and standard deviation as the set of logarithms of measurements. Electrical conductivity and temperature also follow a lognormal distribution, while pH is still positively skewed.

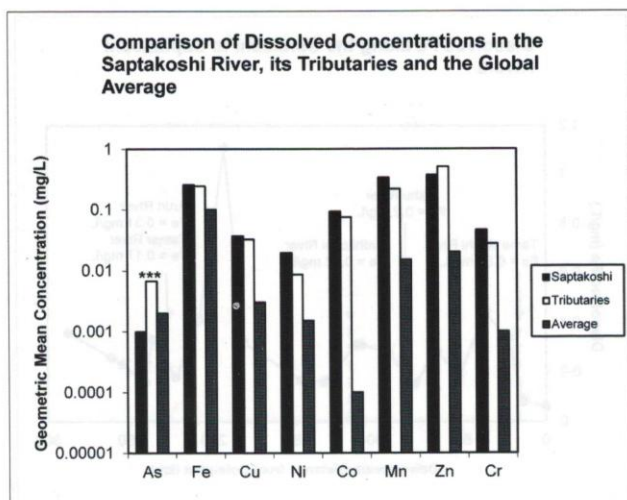


Fig. 4: The difference between dissolved As in the Saptakoshi River (within the Indo-Gangetic Plain) and its tributaries (upstream of the Indo-Gangetic Plain) is statistically significant at the 99.9% confidence level (indicated by ***), according to a comparison of logarithms of concentrations using the Student's t-test, with concentrations below the detection limit set equal to the detection limit. Differences between the Saptakoshi River and its tributaries are not statistically significant for any other element. Except for dissolved As in the Saptakoshi River, all geometric mean concentrations are elevated above the global average (Langmuir 1997).

and the transition elements were elevated above their global average concentrations (Langmuir 1997), especially Co, which was elevated nearly three orders of magnitude above the global average, while the transition elements beside Fe were about one order of magnitude greater than the global average (Fig. 4).

The most dramatic result was that dissolved As became uniformly undetectable as soon the Sunkoshi – Saptakoshi River system entered the Indo-Gangetic Plain (downstream of the confluence of the Arun and Tamar rivers) (Fig. 5a). Such a systematic change was not seen in any other element, pH, EC, or temperature (Figs. 5b-i). No systematic change was seen in any parameter when any other major tributary was crossed or any geological zone was crossed aside from the boundary between the Siwalik Group and the Terai Zone. Mobile As adsorbed onto the 125 – 250 µm sand size fraction did increase downstream, although the correlation with distance was moderate ($R^2 = 0.44$) and was statistically significant only at the 90% confidence level (Fig. 6). It is important to keep in mind that the methodology of this study did not track individual particles of water or sand as they moved downstream, so that spatial and temporal variation could easily be confounded.

The chief results are summarized as follows:

¹ Arithmetic mean ± standard error

Table 1: Comparison of pH, EC and temperature for the Saptakoshi River (Indo-Gangetic Plain) and its tributaries (upstream of the Indo-Gangetic Plain)

	Tributaries	Saptakoshi River	P-value ²
pH	8.22 ± 0.04 ¹	8.25 ± 0.03	0.51
EC (µS/cm)	93 ± 4	100 ± 4	0.19
Temperature (°C)	22.8 ± 0.4	24 ± 1	0.42

² Probability that differences between means are not statistically significant according to the student's test

- 1) Fluvial dissolved arsenic was elevated upstream of the Indo-Gangetic Plain, but became undetectable within the Indo-Gangetic Plain.
- 2) Fluvial dissolved transition elements were elevated both upstream of and within the Indo-Gangetic Plain.

DISCUSSION AND CONCLUSIONS

The results of this study are fully consistent with the fluvial recharge model proposed by Emerman et al. (2007, 2010) and outlined in the introduction to this paper. Rivers in the Himalayan region (upstream of the Indo-Gangetic Plain) have sufficiently high velocity and turbulence so that the large As oxyanion has too high kinetic energy to attach to sediment sorption sites and does not have sufficient residence time in the vicinity of those sorption sites to attach to the sorption sites. Therefore, most of the As remains in dissolved form. When the river enters the Indo-Gangetic Plain, the river velocity and turbulence drop so that the kinetic energy of the As oxyanion drops and its residence time in the vicinity of sediment sorption sites increases. Therefore, the majority of fluvial As migrates from the dissolved form to the adsorbed form, where it can sometimes be transported adsorbed onto suspended sediment and sometimes be deposited on sand bars or floodplains. The transition elements, which occur in aqueous form as small cations, do not mimic the behavior of As, but establish an equilibrium between the dissolved and adsorbed state whether the river velocity is high or low. Since their mass is much smaller than the mass of the As oxyanion, the kinetic energy of the cations is not so high in the Himalayan rivers that they bounce off the sediment sorption sites. Moreover, the smaller volume, lower valency and positive charge of the cations implies that the cations have many more options for sorption sites so that they do not require sufficient residence time in the vicinity of limited sorption sites for attachment to occur.

Aside from Emerman et al. (2007, 2010), we are not aware of any other discussion of the above hypothetical behavior of fluvial As. There are at least four directions for testing the above ideas:

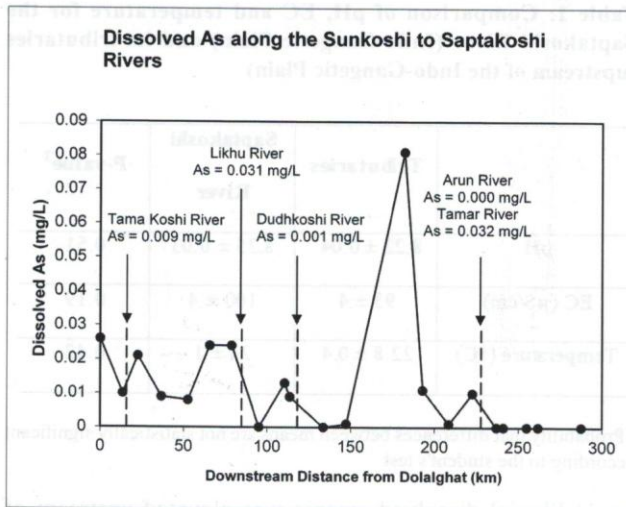


Fig. 5a: Dissolved As becomes undetectable downstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (Indo-Gangetic Plain).

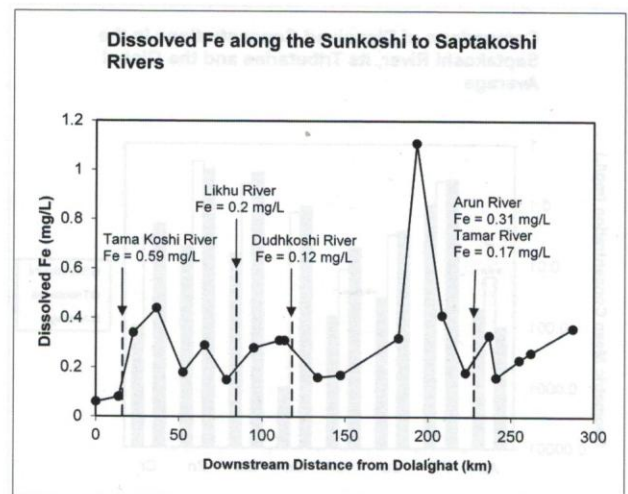


Fig. 5b: There is no systematic difference between dissolved Fe upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

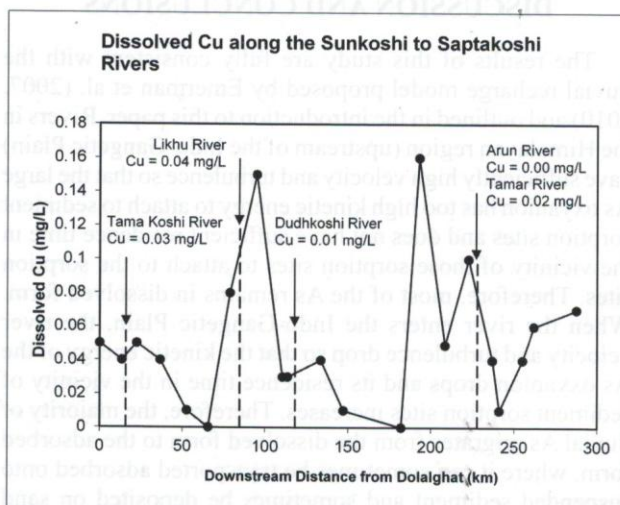


Fig. 5c: There is no systematic difference between dissolved Cu upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

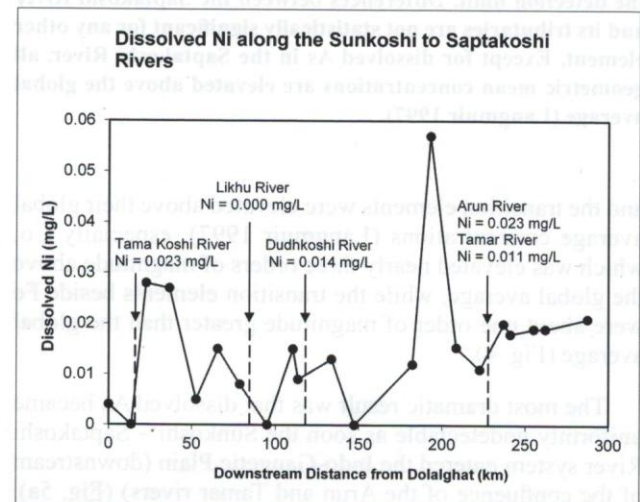


Fig. 5d: There is no systematic difference between dissolved Ni upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

1) A mixture of sediment and water could be stirred sufficiently fast in the laboratory that As cannot adsorb onto sediment, in contrast to other elements, especially the smaller cations, which will adsorb onto sediment.

2) Other oxyanions, such as nitrate and phosphate, should show behavior similar to that of the As oxyanion. That is, for rivers descending from the Himalayan region onto the Indo-Gangetic Plain, or for any other rivers descending from a mountain range onto a flat plain, concentrations of

dissolved oxyanions should drop as soon as the river enters the plain.

3) As mentioned in the introduction, the As oxyanion will tend to remain in overland flow and eventually enter rivers, while the cations will be adsorbed onto sediment and deposited before entering a river. Therefore the concentrations of dissolved As, as well as nitrate and phosphate, should be much stronger functions of the discharge of overland flow than the concentrations of cations.

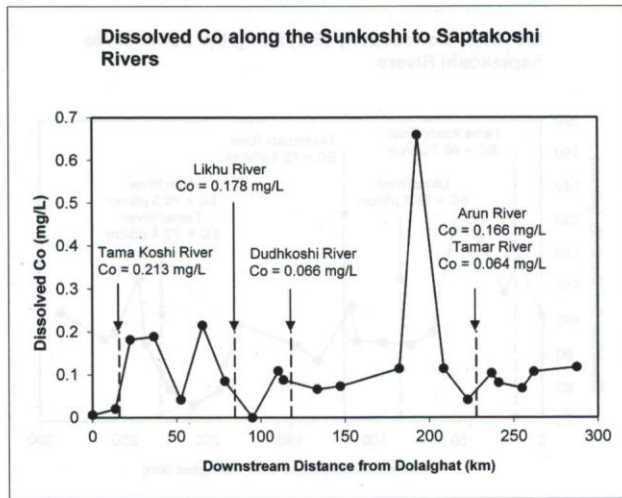


Fig. 5e: There is no systematic difference between dissolved Co upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

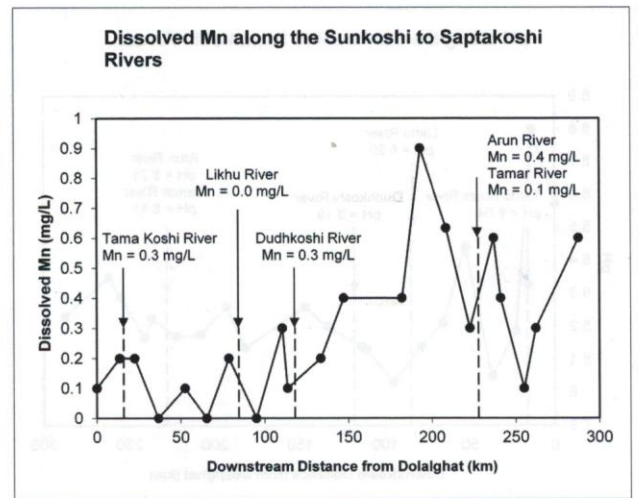


Fig. 5f: There is no systematic difference between dissolved Mn upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

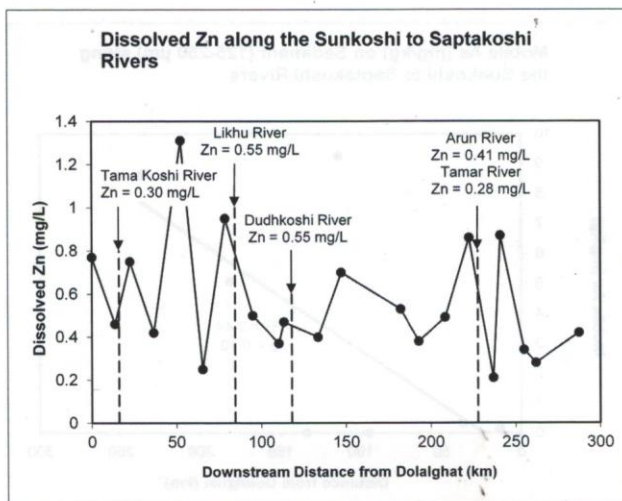


Fig. 5g: There is no systematic difference between dissolved Zn upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

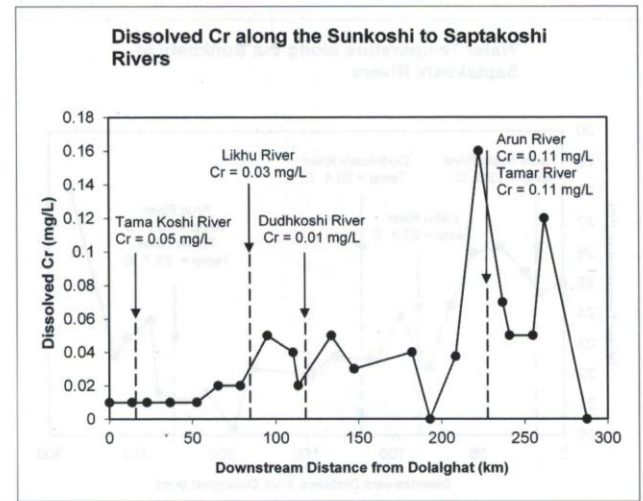


Fig. 5h: There is no systematic difference between dissolved Cr upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

4) It should be possible to sample the suspended sediment from the Sunkoshi – Saptakoshi River system, and verify that the mix of adsorbed anions changes from monovalent monatomic ions such as chloride to oxyanions such as the As oxyanion as the river enters the Indo-Gangetic Plain.

We now consider the implications of this study for As contamination of groundwater in south Asia. The dominant paradigm for arsenic studies in south Asia, called the

reductive-dissolution model, implies that As contamination of groundwater in south Asia is unrelated to any human activity and that solutions must be sought in low-cost methods of water treatment and the avoidance of aquifers contaminated with As. Emerman et al. (2010) found elevated groundwater As in Kathmandu Valley and showed groundwater geochemistry inconsistent with the reductive-dissolution model. Emerman (2005) and Emerman et al. (2010) showed that the concentration of surface water As was statistically indistinguishable from that of groundwater As

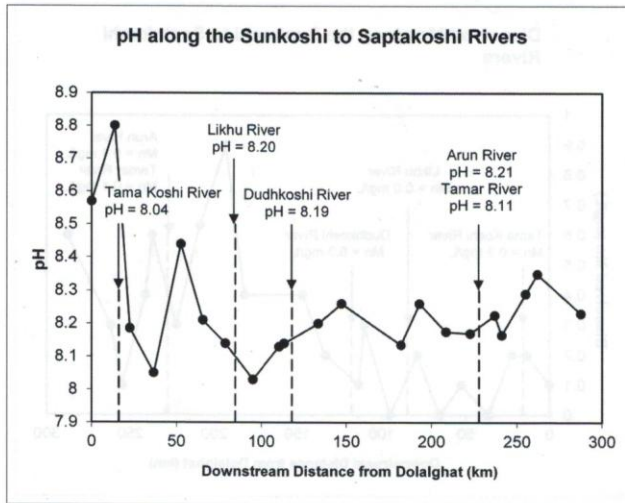


Fig. 5i: There is no systematic difference between fluvial pH upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

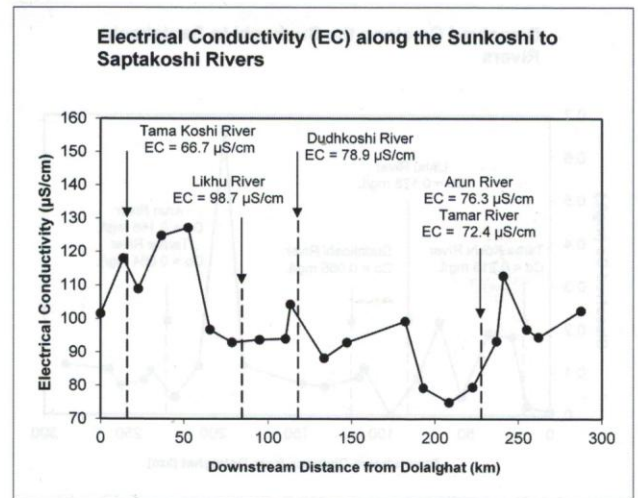


Fig. 5j: There is no systematic difference between fluvial electrical conductivity (EC) upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

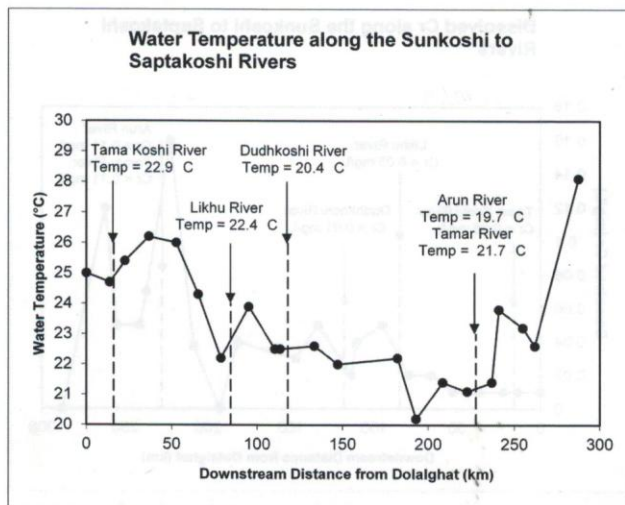


Fig. 5k: There is no systematic difference between water temperature upstream of the confluence of the Arun and Tamar Rivers with the Sunkoshi River to create the Saptakoshi River (upstream of the Indo-Gangetic Plain) and downstream of the confluence (within the Indo-Gangetic Plain).

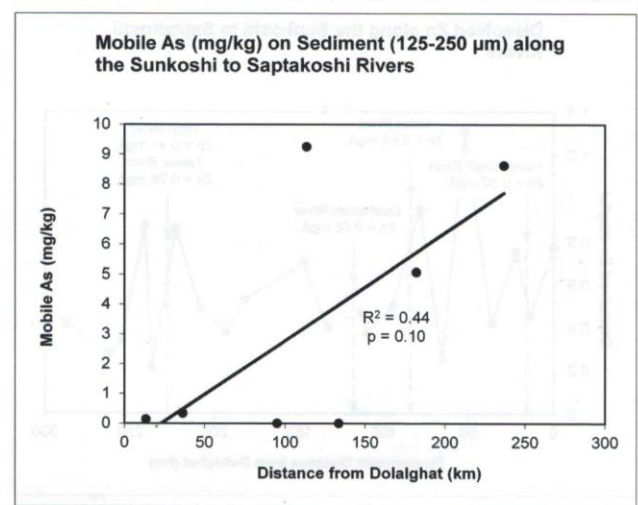


Fig. 6: There is a moderate correlation ($R^2 = 0.44$) significant at the 90% confidence level between mobile As adsorbed to sediment (125-250 μm) and downstream distance along the Sunkoshi to Saptakoshi Rivers.

in Kathmandu Valley and argued that elevated groundwater As was simply due to recharge by rivers with elevated As. Although high fluvial As could result from tectonic uplift and the monsoon climate, deforestation could also be a major contributing factor. Although the headmost waters of the Bagmati River watershed (equivalent to Kathmandu Valley) are in the protected forest of the Shivapuri Watershed and Wildlife Reserve, the vast majority of the watershed has been intensively deforested (Shrestha et al. 1998).

Although neither Emerman (2005) nor Bhatt and Gardner (2009) sampled the Bagmati River or its tributaries within the protected area, comparing water quality within and downstream of the protected area could yield insight into the impact of deforestation. It could be argued that the occurrence of elevated groundwater As in deep tubewells that tap the confined aquifer in Kathmandu Valley (Khatriwada et al. 2002; Gurung et al. 2006; Warner et al. 2008; Chapagain et al. 2009; Maharjan et al. 2009; Emerman et al. 2010) is

inconsistent with a possible impact of deforestation during historic time. Based on a limited number of measurements of ³⁶Cl, Cresswell et al. (2001) estimated the age of groundwater in the confined aquifer to be 200,000 to 400,000 years with groundwater flow velocities from the recharge area of only 45 mm/year. On the other hand, Maharjan et al. (2009) tested 122 deep tubewells for As, both pre-monsoon and monsoon, and found that the number exceeding the WHO As Standard dropped from 71.3% to 57.4% from the pre-monsoon to the monsoon periods. The findings of Maharjan et al. (2009) on the dramatic effect of monsoonal recharge on the chemistry of the confined aquifer draws into question the interpretation of Cresswell et al. (2001). Therefore, based on the current evidence, it is possible that As contamination of groundwater in the tectonic valleys upstream from the floodplain of the Ganges River cannot be fully addressed without also addressing the contributing factor of deforestation.

This study, as well as the results cited by Smedley and Kinniburgh (2002), shows that elevated fluvial As occurs only upstream of the floodplain of the Ganges River. Therefore, the fluvial recharge model could be valid only in the upstream tectonic valleys, while the reductive-dissolution model could be valid in the Ganges floodplain. This spatial separation of models could imply a different set of solutions that should be sought in the different tectonic and depositional environments. Obviously, much more data are needed with regard to both surface water and groundwater As. Both groundwater and surface water As concentrations are needed from the upstream tectonic valleys as Kathmandu Valley is the only tectonic valley studied thus far. Pokhara Valley and Mustang Valley of Nepal, Dehra Dun Valley and Har Ki Dun Valley of India, and Puna Tsang Chhu Valley of Bhutan are additional possibilities. It is hoped that this study will stimulate the collection of far more data on surface water As in India and Bangladesh.

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