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# Deforestation, arsenic, and the self-organizing jungle in the Terai region of Nepal

**Steven H. Emerman**
*Department of Biology and Environmental Science, Simpson College, Indianola, Iowa 50125, U.S.A.*
*(E-mail: emerman@simpson.edu)*

## ABSTRACT

The objectives of this study were (1) to determine whether As contamination of groundwater in the Terai region of Nepal could result from deforestation and (2) to understand the As cycle in terms of the theory of complexity. The hypothesis is that jungle includes a microbial population that sequesters As in immobile form during the dry season and converts As into mobile form during the monsoon season. When jungle is converted into pasture or agricultural land, the new microbial population keeps As in mobile form throughout the year. The hypothesis was tested near Hiraganj (Nawalparasi District) and near Bardibas (Mahottari District). At each site, soil was collected from the A and B horizons along five traverses across jungle and adjacent pasture in February (dry season) and June (beginning of monsoon season). Mobile soil As was measured by extraction with 1 M HCl. At the Hiraganj site, jungle mobile soil As rose from (260 ± 20) µg/kg in February to (350 ± 20) µg/kg in June, while the pasture mobile soil As was (370 ± 30) µg/kg in February and (360 ± 20) µg/kg in June. At the Bardibas site, jungle mobile soil As was (190 ± 10) µg/kg in February and (250 ± 40) µg/kg in June, while the pasture mobile soil As was (240 ± 20) µg/kg in February and (230 ± 10) µg/kg in June. Variation in mobile soil As could not be explained simply in terms of variation in soil pH or gravitational water content.

## INTRODUCTION

### Deforestation and arsenic contamination of groundwater

In the past several years a great deal of attention has focused on the problem of As contamination of groundwater in West Bengal and Bangladesh (Bhattacharaya et al. 1997; Dhar et al. 1997; Nickson et al. 1998). It is generally agreed that the As contamination is too widespread to be due to human activities such as smelting or use of As-based pesticides (Aswathanarayana 1997). However, some workers have suggested that human activities have promoted the transfer of naturally occurring As from sediment into groundwater. Badal et al. (1996) and Mallick and Rajgopal (1996) have argued that over-pumping of aquifers has caused oxidation of sulfide minerals and release of coprecipitated As into groundwater. Acharyya et al. (1999, 2000) have proposed that excessive use of phosphate fertilizers has resulted in displacement of As from sediment adsorption sites by phosphate. The most recent studies have argued that As contamination is unrelated to human activities. According to these studies, As contamination results from the release of As from adsorption sites on Fe oxyhydroxides after dissolution of the Fe oxyhydroxides (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). Both processes are likely under the strongly reducing conditions found in the thick package of alluvial sediments in West Bengal and Bangladesh (Bose and Sharma 2002). An alternative model is that As is displaced from adsorption 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). Still another alternative is that As is coprecipitated with diagenetic carbonate concretions and that As is released into groundwater upon dissolution of the carbonate concretions under acidic conditions (Shanker et al. 2001). Smedley and Kinniburgh (2002) have contributed a comprehensive review of the occurrence of As in natural waters.xxxxxx

The World Health Organization (WHO) maximum permissible concentration for As in drinking water is 10 µg/l. By contrast, the average As concentration in unconsolidated sediment is 3 mg/kg with range 0.6 – 50 mg/kg. Alluvial sediments in Bangladesh do not have unusually high As concentration. Alluvial sands have average As concentration of 2.9 mg/kg with range 1.0–6.2 mg/kg. Alluvial muds have average As concentration of 6.5 mg/kg with range 2.7–14.7 mg/kg (Smedley and Kinniburgh 2002). Smedley and Kinniburgh (2002) concluded that insight into naturally occurring As contamination in groundwater should be sought not in determining the ultimate source of As in bedrock, but in determining why As is released into groundwater, rather than being retained in sediment. Drinkable groundwater results from the fact that, normally, the vast majority of As is adsorbed on sediment or coprecipitated with sediment or exists in crystalline form in sediment and is not released into groundwater. The same discussion could be applied to soil. The average As concentration in soil is 7.2 mg/kg with range 0.1– 55 mg/kg (Smedley and Kinniburgh 2002). If even a small fraction of soil As were mobilized and leached into groundwater, undrinkable groundwater could result. Therefore, it is difficult to ignore changes in land use, such as deforestation or overgrazing, in terms of understanding the As cycle in south Asia.

As contamination in groundwater is also known to occur in the Terai region (Indo-Gangetic plain) of Nepal (Fig. 1), although it has not been so well documented as in other parts of south Asia. As of March 2002, nearly 27% of tested wells exceeded the WHO guideline value while 5% exceeded the Interim Nepal Standard of 50 mg/l. The most affected districts were Rautahat, Nawalparasi, Parsa and Bara (Neku and Tandukar 2002). Since intensive deforestation has recently occurred in the Terai, the Terai is an ideal area for testing the hypothesis that As contamination of groundwater could result from changes in land use. In 1927, the Terai had 48% forest cover with 70% forest cover in western Terai (Fig. 2). The remainder was largely covered with open grasslands of elephant grass and reeds with a few isolated small towns near the Indian railheads (Sharma 1995). Prior to 1950, the Rana government began cutting timber in the Terai for sale to British India (Sharma 1988). Following the overthrow of the Rana government in 1950, hill people started migrating to the Terai after the malaria eradication program of the 1950's. The deforestation of the Terai intensified in the 1970's due to the clearing of agricultural land and cutting of trees for timber and fuel wood. By 1987, only 10% of the Terai was covered by forest with only 4% forest cover in western Terai (Fig. 2). As of 1994, the deforestation of all of Nepal was continuing at a rate of 2% reduction per year (Sharma 1995). Therefore, the first objective of this study was to determine whether the deforestation of the Terai could be related to As contamination of groundwater.

## ARSENIC CYCLE IN TERMS OF COMPLEXITY THEORY

The second objective of this study was to determine whether the As cycle in the Terai could be understood in terms of the new field of complexity theory, also known as the theory of complex systems. Complexity theory is the study of the properties that emerge when parts assemble into a whole (Kauffman 1993, 2000; Lewin 1993; Coveney and Highfield 1995; Holland 1995; Bak 1996; Bar-Yam 1997; Rayner 1997). An excellent example is consciousness, which is a property of a whole living organism, but which is not found in any of the cells, organs, or other parts of an organism. Complexity theory has been applied to a wide variety of fields but only recently to ecology and geology (Watson and Lovelock 1983; Klinger 1991; Jørgensen et al. 1992; Lovelock 1995; Solé and Manrubia 1995; Klinger and Short 1996; Klinger and Erickson 1997; Von Bloh et al. 1997; Levin 1998; Downing and Zvirinsky 1999; Harding 1999; Bradbury et al. 2000; Jørgensen and Müller 2000; Emerman and Parmelee 2002; Lenton and van Oijen 2002; Klinger 2003; Allen and Emerman 2003). See Clements (1916), Margalef (1963) and Odum (1969) for historical antecedents of applications of complexity to ecology.) According to complexity theory, when the parts, such as plants, animals, microorganisms and soil, assemble into an ecosystem, properties emerge that allow the ecosystem, in some ways, to mimic the behavior of a single organism. One of the properties generally associated with organisms is the ability to repel invasion by other organisms. Emerman and Parmelee (2002) and Allen and Emerman (2003) showed data consistent with the hypothesis that a prairie ecosystem can resist invasion by a shrubland ecosystem by maintaining low soil moisture and keeping N in organic form. Another property generally associated with organisms is the ability to sequester toxins. The preliminary hypothesis of this paper is that the jungle ecosystem includes a microbial population

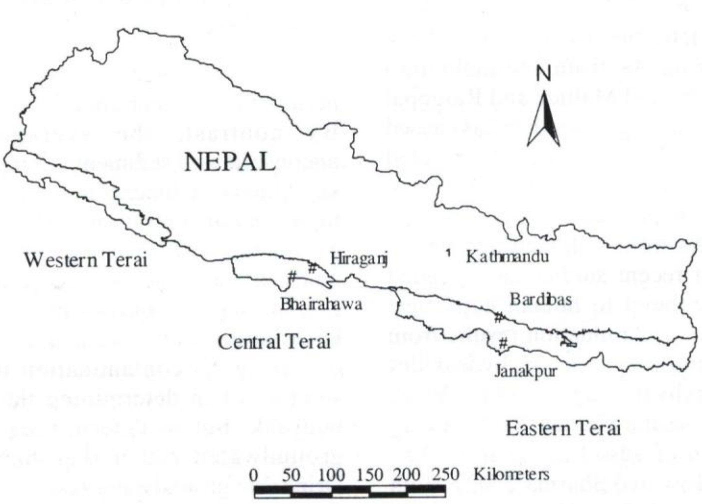

**Fig. 1: Terai region of Nepal (based upon Physiographic Map of Nepal, p. 17, Sharma (1990). Soil samples were collected at Hiraganj and Bardibas. Precipitation stations were located at Bhairahawa and Janakpur.**

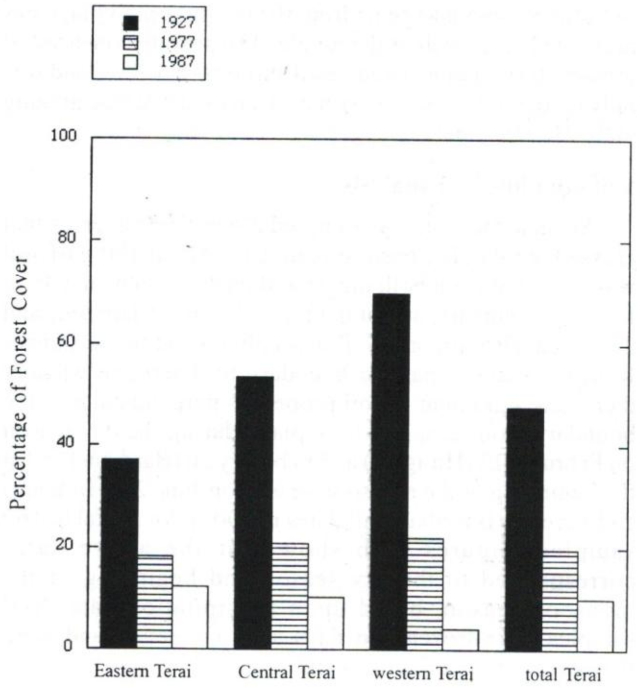

**Fig. 2: Decline in percentage of forest cover in the Terai region (data from Sharma 1995). The 1927 data are based on Survey of India topographic maps. The 1977 and 1987 data are based on satellite imageries.**

that transforms As into an immobile / non-bioavailable form such as a methylated form or an arsenosugar or arsenobetaine (Cullen and Reimer 1989; Gustafsson and Jacks 1995). When the jungle is cut down, its associated microbial population ceases to exist. The new microbial population transforms As back into a mobile / bioavailable form after which some As is leached into groundwater. The implication of the hypothesis is that, if soil were sampled in adjacent jungle and pasture, there would be higher levels of mobile soil As on the pasture side of the boundary. Pasture, rather than agricultural land, is an appropriate comparison for jungle since agricultural land in Bangladesh has been shown to be contain high levels of soil As due to irrigation with As-contaminated groundwater (Islam et al. 2000; Meharg and Rahman 2003).

Recent work in the theory of complexity suggests that such a simple hypothesis as above cannot be correct (Klinger 2003). Complex systems possess both symmetry-building behaviors and symmetry-breaking behaviors. Symmetry refers to repeatability in space and / or time. For example, the immobilization of As by microorganisms is an example of a symmetry-building behavior, it leads to consistently low mobile soil As levels both spatially and temporally. Klinger (2003) has argued that complex systems cannot attain long-term stability through symmetry-building behavior alone. Systems with only symmetry-building behavior become inflexible and unable to respond to change. The change could arise externally through a changing environment or internally through the natural evolution of a system. In the case of the jungle, there is a steady input of As into the system through uptake by plants and weathering of sulfide minerals. Eventually, the added As will overwhelm the ability of the microbial population to immobilize As unless the jungle has some means of expelling As from the system. It is proposed that the symmetry-breaking behavior that maintains the long-term stability is that the jungle microbial population converts immobile As back into mobile As when leaching potential is high, so that excess As can be leached from the system.

The complete hypothesis of this paper is stated as follows:

1. The jungle soil includes a microbial population that transforms As into immobile/non-bioavailable form during the dry season. Such a microbial population does not exist in deforested pasture.
2. The jungle soil also includes a microbial population that transforms As back into mobile/bioavailable form during the monsoon season.

The testable implications of the hypothesis are the following:

1. During the dry season, mobile soil As will be lower on the jungle side of a jungle / pasture boundary.
2. At the beginning of the monsoon season, mobile soil As on the jungle side of a jungle/pasture boundary will rise to the level of the pasture side.

The beginning of the monsoon season is the relevant time period, since, in the middle of the monsoon season, the rate of leaching may exceed the rate at which As is transformed into mobile form. The hypothesis also predicts that there should be a long-term trend toward decreasing total soil As on the pasture side. However, the measurement of total soil As requires the use of hot concentrated acids, which could not be used safely in the absence of a fume hood.

**Measurement of mobile soil As**

There is presently no consensus in the soil chemistry literature as to the procedure for measuring mobile or bioavailable soil As (Johnson and Hiltbold 1969; Woolson et al. 1971b, 1973; Woolson 1973; Johnston and Barnard 1979; Alam et al. 2001; Cai et al. 2002). Woolson et al. (1971a) showed that the growth of corn correlated poorly with total soil As, but correlated well with As extracted by a variety of dilute acids and bases. Most As in soil is coprecipitated with oxyhydroxides, silicates or sulfides and is unavailable to plants. Most studies have focused on understanding the particular sites in soil from which As was being extracted. 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 $MgCl_2$ (2 hours, two repetitions) extracted the ionically bound As and 1 M $NaH_2PO_4$ (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 coprecipitated with acid-volatile sulfides, carbonates, Mn oxides, and very amorphous Fe oxyhydroxides. Successively more aggressive extractants were used until hot 16 M $HNO_3$ and 30% $H_2O_2$ was used to extract As from orpiment and other recalcitrant As minerals. Keon et al. (2001) did not study extraction of As in organic form (Gustafson and Jacks 1995). In this study extraction with 1 M HCl was used as a measure of mobile / bioavailable soil As. One reason for this choice was that Keon et al. (2001) obtained meaningful results with only one hour of contact time. Since no automatic stirring or shaking devices were available, all solutions had to be stirred by hand. Langston (1980) and Reuther (1992) also used extraction with 1 M HCl as a measure of bioavailable soil As.

## MATERIALS AND METHODS

### Description of study sites

Two sites in the Terai were chosen where there was a sharp boundary between jungle and pasture. The first site (27°35'44"N, 83°41'20"E) was near the village of Hiraganj in Nawalparasi District, Western Development Region, while the second site (26°59'58"N, 85°51'47"E) was near the village of Bardibas, Mahottari District, Central Development Region (Fig. 1). Both sites occurred on the alluvial fan apron complex (upper piedmont erosional surface) at the foot of the Churia Hills. Both sites had very gentle slopes (<1°), soil with loamy texture and moderately good drainage (Nepal Survey Department 1982a, 1982b). Although there is no detailed soil survey of Nepal, the Department of Survey has produced land system maps that divide the Terai region into 12 land system units. According to this scheme, the soils of both sites were classified as Haplustolls / Dystochrepts / Ustochrepts (Nepal Survey Department 1982a, 1982b). The soils of both sites were clearly inceptisols, not mollisols, due to the thin epipedon (< 15 cm). The jungles of both sites were dominated by sal (*Shorea robusta*) with secondary shrub sano panheli (*Mallotus phillipensis*). The Hiraganj site also included the tree black plum (java plum, jamun) (*Syzygium cumini*) and the shrubs curry leaf tree (mitho nim) (*Murraya koenigii*) and black myrobalan (harro) (*Terminalia chebula*). The Bardibas site also included the shrub ramphal (*Dillenia indica*) and asna (saj) (*Terminalia alata*), both as a tree and shrub. All secondary species are commonly found in the sal forest of the Terai (Storrs and Storrs 1998).

At the Hiraganj site, an unnamed tributary of Dhalagirwa Khola separated the jungle and pasture. According to local residents, the pasture had been produced by deforestation 20 years previously and had never been irrigated. The pasture was intensively grazed by buffalo, cows and goats, and the grass was bitten down almost to bare dirt during both the dry and monsoon seasons. At the Bardibas site, the pasture was created by cutting down the jungle to install a high-tension line along the Mahendra Highway 15 years previously. After the jungle was cut down, a steady passage of buffalo, cows and goats from the neighboring village has prevented re-growth of the jungle. The pasture consisted of grasses, forbs and isolated small shrubs (up to 1 m) and was only moderately grazed in comparison to the intense grazing at the Hiraganj site.

### Soil sampling and analysis

At each site, soil was sampled along five traverses that crossed the jungle / pasture boundary. About 100 g of soil was collected from both jungle and pasture, 20 m and 40 m from the boundary, and at depths 5–10 cm (A horizon) and 25–30 cm (B horizon). Soil was collected at two distances from the jungle / pasture boundary to determine whether there was a gradient in soil properties perpendicular to the boundary. Soil sampling took place during the dry season on February 13 (Hiraganj) and February 20 (Bardibas) and at the beginning of the monsoon season on June 23 (Hiraganj) and June 24 (Bardibas) (all dates in 2003), for a total of 160 samples. Figures 3a-b show that the above dates corresponded to the dry season and beginning of the monsoon season, based upon precipitation data. Soil samples were frozen two days after collection and were thawed the day before analysis.

Gravitational water content was determined by weighing a sub-sample, drying it at 105°C for 24 hours, and weighing it again. For the dry season samples, particles larger than 2 mm (gravel) were removed by sieving prior to the initial weighing. The monsoon season samples were too wet to be sieved. The gravel was sieved after drying and the gravitational water content was corrected by adding the weight of gravel to the weight of the container. Soil pH was measured by adding 40 ml of distilled water to 20 g of air-dried soil. The dry season samples were sieved to remove gravel. Gravel was manually picked out from the monsoon season samples. The solution was initially stirred 30 times and then stirred 10 times every 5 minutes for 30 minutes. The solution was allowed to settle for 1 hour and pH was measured by inserting the pH sensor into the supernatant. The dry season samples were measured with the Oakton pHTestr with ATC, the monsoon season samples were measured with the Hanna HI 9025 pH meter.

Mobile soil As was measured by adding 50 ml of 1 M HCl to 5 g of field-moist soil. By contrast, Keon et al. (2001) used a ratio of extractant to freeze-dried soil of 100:1 to avoid exhausting the extractant. Johnston and Barnard (1979) used a ratio of extractant to air-dried soil of 20:1 for a wide variety of extractants. McLaren et al. (1998) used a ratio of extractant to field-moist soil of 15:1 for extraction with 1 M HCl. The ratio of extractant to field-moist soil of 10:1 was chosen here so that the solution As concentration would fall in the range of accurate measurement of the Hach Arsenic Test Kit, which was 10 –70 μg/l. This choice will be justified in the discussion section. The solution was initially stirred 30 times and then 10 times every 5 minutes for 60 minutes. The solution was then poured directly into the wide-mouth reaction vessel of the Hach Arsenic Test Kit without filtering. The principle of the Hach Arsenic Test Kit is that three reagents are

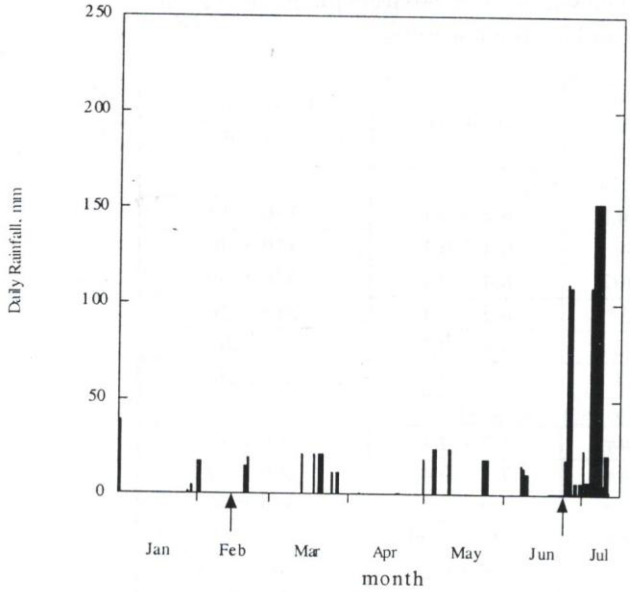

**Fig. 3a: Daily rainfall data at Bhairahawa from January 1 to July 15, 2003 (data collected by Department of Hydrology and Meteorology and reported daily in Annapurna Post (in Nepali)). Arrows indicate dates of soil collection at Hiraganj.**

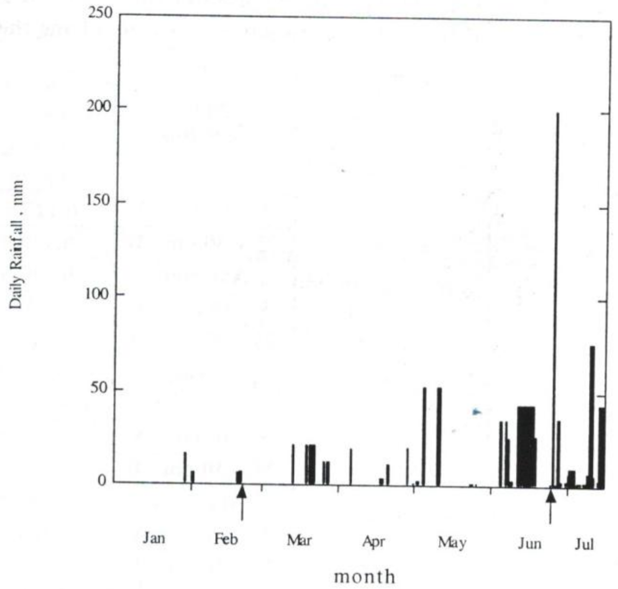

**Fig. 3b: Daily rainfall data at Janakpur from January 1 to July 15, 2003 (data collected by Department of Hydrology and Meteorology and reported daily in Annapurna Post (in Nepali)). Arrows indicate dates of soil collection at Bardibas.**

successively added to mask interference by H sulfide. Sulfamic acid and powdered Zn are then added to create strongly reducing conditions in which inorganic As is reduced to arsine gas ($AsH_3$). The arsine gas then reacts with mercuric bromide in the test strip to form mixed As / Hg halogenides that discolor the test strip. In this study, a drop of amyl alcohol was added to the solution after adding sulfamic acid and powdered Zn to prevent foaming. The addition of the first three reagents required 20 minutes. The reaction was then complete 30 minutes after adding sulfamic acid and Zn. Since the extractant continued to extract As while the solution was in the reaction vessel, the total extraction time was probably $(60 + 20 + 30 / 2) = 95$ minutes, assuming that the measured As value referred to the As concentration midway through the period of reducing conditions. Some arsenic test kits, such as the Nepali-made Environmental and Public Health Organization (ENPHO) kit use a narrow-mouth reaction vessel and it would not be possible to pour in the solution without filtering. In that case, equivalent results would probably be obtained by extracting As for 95 minutes prior to filtering, although that would need to be checked. The ENPHO method could also be modified to accommodate a wide-mouth reaction vessel.

The color chart that accompanies the Hach Arsenic Test Kit shows the colors that correspond to 0, 10, 30, 50, 70, 300 and 500 mg/l. Before carrying out the soil analyses, As test solutions were prepared with concentrations 0–70 mg/l at intervals of 5 mg/l with three replicates of each concentration. An analysis of these test solutions showed that the colors could be read to a precision of ± 5 mg/l and that the reproducibility of the Hach Arsenic Test Kit was also ± 5 mg/l. Due to the 10:1 ratio of extract solution to soil, the precision of any measurement of soil As should be ± 50 mg/kg. Therefore, the difference between two averages of soil As should be regarded as statistically significant only if the difference is at least, approximately, 100 mg/kg, in addition to other statistical considerations.

## RESULTS

Table 1a shows soil gravitational water contents, soil pH and soil mobile As for the Hiraganj site during February (dry season) and late June (beginning of monsoon season). (Statistical significance was determined by the t-test.) In the dry season, the pasture soil and jungle soil had the same gravitational water content and pH. In the A horizon, the mobile soil As was the same in both pasture and jungle. However, in the B horizon, mobile soil As was higher in the pasture soil and was higher as averaged over the two horizons. From the dry season to the beginning of the monsoon season, the gravitational water content rose in the pasture soil, but did not rise in the jungle soil. Part of the evidence that the monsoon season had only just begun or had not yet begun is that the jungle soil was not yet wet, as most of the recent precipitation was still held in the jungle canopy. The soil pH did not change in either the pasture or jungle. The mobile soil As did not change in the A horizons of the pasture and jungle, or in the B horizon of the pasture. However, the mobile soil As rose in the B horizon of the jungle, so that, at the beginning of the monsoon, mobile soil As was the same in jungle and pasture, either by comparison of horizons or as averaged over the two horizons. The average mobile soil As for all measurements at the Hiraganj site was (0.34 ± 0.01) mg/kg.

**Table 1a: Gravitational water content, soil pH, and mobile soil As in adjacent jungle and pasture at Hiraganj, Nawalparasi District, during the dry and monsoon seasons.**

| Ecosystem | Depth / Horizon | Gravitational water content (g $H_2O$/g dry soil) | Soil pH | Mobile soil As (μg As/kg dry soil) |
|---|---|---|---|---|
| February (dry season) | | | | |
| Pasture | **5 – 10 cm / A** | **0.117 ± 0.003[a]** | **6.4 ± 0.1** | **330 ± 30** |
| | **25 – 30 cm / B** | **0.118 ± 0.003** | **6.4 ± 0.1** | **410 ± 40** |
| | **Average** | **0.117 ± 0.002** | **6.4 ± 0.1** | **370 ± 30** |
| Jungle | **5 – 10 cm / A** | **0.17 ± 0.02*** | **6.2 ± 0.1** | **310 ± 20** |
| | **25 – 30 cm / B** | **0.14 ± 0.02** | **6.6 ± 0.3** | **220 ± 40**** |
| | **Average** | **0.16 ± 0.01** | **6.4 ± 0.2** | **260 ± 20*** |
| June (beginning of monsoon season) | | | | |
| Pasture | **5 – 10 cm / A** | **0.22 ± 0.02###** | **6.7 ± 0.4** | **360 ± 30** |
| | **25 – 30 cm / B** | **0.24 ± 0.02###** | **7.1 ± 0.4** | **360 ± 30** |
| | **Average** | **0.23 ± 0.01###** | **6.9 ± 0.3** | **360 ± 20** |
| Jungle | **5 – 10 cm / A** | **0.184 ± 0.008** | **6.27 ± 0.09** | **310 ± 20** |
| | **25 – 30 cm / B** | **0.16 ± 0.02**** | **6.6 ± 0.2** | **390 ± 30##** |
| | **Average** | **0.17 ± 0.01*** | **6.5 ± 0.1** | **350 ± 20#** |

[a]**Value** ± standard error

*, **, *** indicates difference between value in jungle and pasture at the same depth and season is statistically significant at the 95%, 99% and 99.9% confidence levels

#, ##, ### indicates difference between value in an ecosystem (jungle or pasture) and the same ecosystem at the previous season and the same depth is statistically significant at the 95%, 99% and 99.9% confidence levelsTable 1b. Gravitational water content, soil pH, and mobile soil As in adjacent jungle and pasture at Bardibas, Mahottari District, during the dry and monsoon seasons.

Table 1b shows soil gravitational water contents, soil pH and soil mobile As for the Bardibas site during February (dry season) and late June (beginning of monsoon season). Statistical significance was determined by the t-test. In the dry season, the gravitational water content was the same in the pasture soil and jungle soil. As averaged over the two horizons, the soil pH was lower in the jungle (6.0 ± 0.1) than in the pasture (6.4 ± 0.1). In the A horizon, mobile soil As was higher in the pasture. Mobile soil As was the same in the B horizons of pasture and jungle. Mobile soil As was higher in the pasture as averaged over the two horizons according to the t-test, but the difference was only 50 mg/kg, or less than the precision of the Hach Arsenic Test Kit, as discussed above. Between February and June, the gravitational water content rose in both pasture and jungle soil. The soil pH rose in both pasture and jungle so that, as averaged over the two horizons, the soil pH was still lower in jungle (6.3 ± 0.1) than in pasture (6.9 ± 0.1). The mobile soil As did not change in either horizon of either pasture or jungle. At the beginning of the monsoon season, the mobile soil As was the same in pasture and jungle in both horizons and as averaged over the two horizons. The average mobile soil As for all measurements at the Bardibas site was (0.23 ± 0.01) mg/kg. The difference in average mobile soil As for the two sites was statistically significant ($P = 0.0004$). In no case was the difference in soil properties within one ecosystem at a given depth, in samples collected 20 m and 40 m from the boundary with the adjacent ecosystem, found to be statistically significant.

The results of this study can be summarized as follows:

1. In the dry season, the mobile soil As was lower in the jungle than in the pasture. At the beginning of the monsoon season, the mobile soil As was the same in jungle and pasture. However, the results are more complex than predicted by the hypothesis. At the Bardibas site, the difference in mobile soil As occurred in the A horizon, while at the Hiraganj site, the difference occurred in the B horizon. Moreover, although the pasture mobile soil As was the same as the jungle mobile soil As at the beginning of the monsoon season at the Bardibas site, the rise in jungle mobile soil As from February to June was not statistically significant, contrary to what was predicted by the hypothesis.

2. Differences between mobile soil As in adjacent jungle and pasture cannot be explained simply in terms of differences in soil moisture, nor can the change in mobile soil As between seasons be understood simply in terms of changes in soil moisture.

**Table 1b: Gravitational water content, soil pH, and mobile soil As in adjacent jungle and pasture at Bardibas, Mahottari District, during the dry and monsoon seasons.**

| Ecosystem | Depth | Gravitational water content (g $H_2O$/g dry soil) | Soil pH | Mobile soil As (µg As/kg dry soil) |
|---|---|---|---|---|
| | | February (dry season) | | |
| Pasture | **5 – 10 cm / A** | **0.14 ± 0.01$^{a}$** | **6.3 ± 0.2** | **280 ± 30** |
| | **25 – 30 cm / B** | **0.09 ± 0.02** | **6.4 ± 0.3** | **200 ± 30** |
| | **Average** | **0.11 ± 0.01** | **6.4 ± 0.1** | **240 ± 20** |
| Jungle | **5 – 10 cm / A** | **0.14 ± 0.01** | **6.0 ± 0.1** | **180 ± 20****** |
| | **25 – 30 cm / B** | **0.065 ± 0.005** | **6.0 ± 0.1** | **200 ± 10** |
| | **Average** | **0.10 ± 0.01** | **6.0 ± 0.1*** | **190 ± 10*** |
| | | June (beginning of monsoon season) | | |
| Pasture | **5 – 10 cm / A** | **0.213 ± 0.006###** | **6.9 ± 0.1#** | **240 ± 20** |
| | **25 – 30 cm / B** | **0.217 ± 0.006###** | **6.9 ± 0.1** | **220 ± 20** |
| | **Average** | **0.215 ± 0.004###** | **6.9 ± 0.1#** | **230 ± 10** |
| Jungle | **5 – 10 cm / A** | **0.206 ± 0.007###** | **6.4 ± 0.1**, #** | **230 ± 20** |
| | **25 – 30 cm / B** | **0.213 ± 0.008###** | **6.3 ± 0.1***, ##** | **270 ± 70** |
| | **Average** | **0.209 ± 0.005###** | **6.3 ± 0.1***, ##** | **250 ± 40** |

$^{a}$**Value** ± standard error

*, **, *** indicates difference between value in jungle and pasture at the same depth and season is statistically significant at the 95%, 99% and 99.9% confidence levels

#, ##, ### indicates difference between value in an ecosystem (jungle or pasture) and the same ecosystem at the previous season and the same depth is statistically significant at the 95%, 99% and 99.9% confidence levels

3. Differences between mobile soil As in adjacent jungle and pasture cannot be explained simply in terms of differences in soil pH, nor can the change in mobile soil As between seasons be understood simply in terms of changes in soil pH.

## DISCUSSION

### Origin of seasonal variation in mobile soil As

What is the cause of the seasonal variation in mobile soil As if it is neither changes in soil moisture nor changes in soil pH? Changes in soil temperature and length of daylight would be consistent with the data of this study, but there are not sufficient data to reach that conclusion. Measurements of the monthly variation in mobile soil As along with monitoring of environmental parameters and microbial populations would be necessary to determine the cause of the temporal variation in mobile soil As. It is important to note that the utility of complexity theory is that it predicts the general tendencies of ecosystem behavior without the necessity of understanding the detailed chain of causes.

### Comparison with previous studies3

Although the Terai region has been shown to have high levels of groundwater As, the levels of mobile soil As for soil that has not been irrigated with As-contaminated groundwater are not large. By comparison, Johnston and Barnard (1979) measured mobile soil As for uncontaminated soils from New York by extraction with 0.5 M HCl for 1 hour and found a range of 0.23 – 0.48 mg/kg. On average, their measurements of mobile soil As corresponded to 2.5% of total soil As. Using the above percentage and the mobile soil As levels found in this study, the total soil As would be 13.6 mg/kg at the Hiraganj site and 9.2 mg/kg at the Bardibas site, which are well within the background range for total soil As. For orchard soil that had been sprayed with Pb arsenate, Johnston and Barnard (1979) found average mobile soil As to be 52 mg/kg, corresponding to 14% of total soil As. Using extraction with 2.2 M HCl for 30 minutes, Vylev et al. (1993) found average mobile soil As levels of 0.16 mg/kg for uncontaminated soils (2.4% of total As), 0.57 mg/kg for soils irrigated with contaminated groundwater (19% of total As), and 45 mg/kg for As-contaminated soils found near industrial facilities (64% of total As). Using a succession of four less aggressive extractants followed by extraction with 1 M HCl for 16 hours, McLaren et al. (1998) found average mobile soil As to be 1260 mg/kg (72% of total As) for soils contaminated with cattle dip. It has been a common observation that contaminated soils have high fractions of mobile As and that, with time, an increasing fraction of soil As transfers to immobile form (Woolson et al. 1973).

The choice of a 10:1 ratio of solution to field-moist soil for extraction of As can now be justified. Keon et al. (2001) used a 100:1 ratio of solution to freeze-dried soil to avoid exhausting the extractant. Keon et al. (2001) found As extractable by 1 M HCl and a sequence of less aggressive extractants (see Introduction) to be (1420 ± 80) mg/kg (corresponding to (73 ± 7)% of total As) in contaminated wetland soil and (23 ± 1) mg/kg (corresponding to (58 ± 4)% of total As) in contaminated fluvial sediment. If a 100:1 ratio was large enough for such contaminated soils, a 10:1 ratio was certainly sufficient for the uncontaminated soils analyzed in this study.

Paul et al. (1999) carried out laboratory studies on the effect of short-term (2–7 days) moist-dry cycles on soil pH. They related the acidification of soil under moist conditions to nitrification and Mn oxidation. There do not appear to be any field studies on seasonal variation in soil pH under monsoon conditions.

### Deforestation and arsenic contamination of groundwater

The first objective of this study was to determine whether As contamination of groundwater in the Terai region was a consequence of deforestation. It has been shown that, during the dry season, mobile soil As is higher in moderately to intensively grazed pasture than in jungle. The above conclusion does not necessarily imply that deforestation is a critical, or even an important factor, in As contamination of groundwater. Two important questions still remain to be answered. The first question is whether there is sufficient deep drainage due to occasional storms in the dry season to leach appreciable amounts of As from pasture soil into groundwater. The question could be answered by the installation of lysimeters to collect drainage water. The question could also be addressed by modeling if more data on soil physical properties were available, such as soil moisture retention curves or even soil texture. The second question is whether As introduced into underlying sediments by deep drainage remains in the aqueous form or is adsorbed onto sediments. As mentioned above, Woolson et al. (1973) showed that As introduced into soil tends to be adsorbed into less available forms with time. From this perspective, the problem of As contamination of groundwater could be self-correcting. This question could be addressed by modeling, but more knowledge of As kinetics is needed. Another approach would be to search for spatial relations among mobile soil As, groundwater As and land use in a way that takes into account the migration of groundwater in the Terai (Rao and Pathak 1996; Pathak and Rao 1998).

### Arsenic cycle in terms of complexity theory

The second objective of this study was to determine whether the problem of As contamination could be understood in terms of complexity theory. The results of this study were generally consistent with complexity theory, although, as mentioned, there were additional phenomena that were not predicted. The assumption of this study was that a jungle ecosystem includes a microbial population that sequesters As in non-bioavailable form, but a moderately to intensively grazed pasture ecosystem does not. The speculation is that, as a sequence of ecosystems proceeds through a vegetational succession from bare soil to annual grasses to perennial grasses to forest to peatland (Klinger 1991; Klinger and Short 1996), As and other toxins are sequestered into increasingly non-bioavailable forms. Since the early successional stages show relatively rapid floral growth, mechanisms of sequestration of toxins could be a waste of energy since plants can more easily grow at a rate that exceeds the rate of uptake of toxins. The above ideas could be tested by measuring changes in mobile soil As across successional gradients.

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