Study of arsenic (As) mobilization in the Ganga Alluvial Plain using neutron activation analysis

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Received: 8 December 2011 / Published online: 10 January 2012 © Akadémiai Kiadó, Budapest, Hungary 2012

Abstract In northern part of Indian sub-continent, As related environmental and health issues receive attention from all over the world. Forty-eight sediment samples were collected from the Ganga Alluvial Plain (GAP) and its weathering products (from the Gomati River) for the study of As distribution and mobilization. These sediment samples were analyzed by Instrumental Neutron Activation Analysis (INAA) method using Cirus Research Reacter of Bhabha Atomic Research Centre, Mumbai. Average As concentrations in the GAP sediments $(10.44 \text{ mg kg}^{-1})$, the Gomati River bed sediments $(1.36 \text{ mg kg}^{-1})$ and the Gomati River suspended sediments $(5.30 \text{ mg kg}^{-1})$ were reported. Significant decrease of As content from the alluvial sediments to the river sediments is a clear indication of its mobilization by chemical weathering processes of mineral biotite. Present study demonstrates the importance of INAA for quantification and mobilization of As and improves our understanding related to As related environmental issues in northern India and elsewhere.

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Keywords Arsenic mobility · INAA · Sediments · Weathering - Gomati River - Ganga Alluvial Plain

Introduction

Tropical environments are continuously subjected to extreme ranges of temperatures and rainfall resulting strong geochemical fractions of elements in terms of either moderate to severe depletion or enrichment of elements [\[1](#page-5-0)]. Links between this geochemical uniqueness of environment and human health are particularly important for population who are mainly dependent on the local environment for food and water supply. The GAP, a part of the world's largest alluvial plain, experiences the sub-tropical environment and is characterized by one of the densely populated and highly agricultural region in the world. As is chemically classified as a toxic and carcinogenic element and its distribution in the environment are mainly dependent on geology, hydrology, climate, as well as various anthropogenic activities of the region [[2\]](#page-5-0). Human problems associated with As contamination in the Ganga Delta region continue to receive attention from all over the world (Fig. [1\)](#page-1-0). The recent literature survey reveals the same environmental problems in eastern part of the GAP [[3,](#page-5-0) [4](#page-5-0)].

Alluvial Rivers are the major pathways to transport weathered products (both in particulate or solid phase and dissolved phase) of the GAP to the Ganga River (Fig. [1](#page-1-0)). The INAA of unweathered and weathered sediments might provide evidence to natural processes of As mobilization from the GAP to its hydrological components. The present investigation was undertaken to know distribution and mobility of As in the GAP for better understanding of As related human health problems of this region. The aims of this reconnaissance study were to know (i) As content in

Fig. 1 The Indo–Ganga– Brahmaputra Plain, the world's largest alluvial plain located in south of the Himalayan Orogenic Belt in the Indian subcontinent. The Ganga Alluvial Plain represents its middle part and is drained by several alluvial rivers along with the Himalayan Rivers. The Gomati River drains the interfluves area of the Ganga and the Ghaghara Rivers and acts as a tributary of the Ganga River. Groundwater of the Ganga Delta plain is heavily enriched in As and has prompted the present study of As mobilization in the Ganga Alluvial Plain region

the GAP sediments and the Gomati River sediments and (ii) As mobility in weathering of the GAP.

Experimental

Study area

The GAP is an outstanding geographical region due to its low elevation, low relief, high fertility and high human population density. Geologically, it is made-up of interlayered 1–2 m thick fine sand and silty mud deposits showing extensive discontinuous calcrete horizons [\[5](#page-5-0)]. Walther's Law states that surface distribution of various geological environments is preserved in the vertical depositional section. Therefore, the exposed cliff section along the valley margins of river draining the GAP can be classified as the GAP sediments and used for the present study.

Climate is an integral part of the geochemical environment of weathering processes and also has a strong influence upon the characteristics of weathering products [\[6](#page-5-0)]. The GAP experiences a humid sub-tropical climate with four distinct seasons. The hot summer season (March–May) follows by heavy precipitation in the monsoon season (June– September). It is followed by the post-monsoon season (October–November) and the winter season (December– February). The Gomati River, a tributary of the Ganga River, drains about \sim 30, 000 km² area of the interfluves region of the Ganga and the Ghaghara Rivers in the GAP.

In the GAP, weathering processes are largely concerned with the monsoon season and alluvial lithology, and becomes intensive due to large temperature variation $(5-40 \degree C)$ in the winter and summer seasons. Figure [2a](#page-2-0) shows monthly rainfall, maximum and minimum temperatures of the study area at Lucknow. Hydrology of the Gomati River is characterized by very low discharges during the winter and summer seasons. In the monsoon season, the river's discharge increases manifold due to heavy monsoon rainfall and diminishes during the postmonsoon season (Fig. [2](#page-2-0)b). In weathering processes of humid sub-tropical climate condition, weathering products are formed on sub-surface of the GAP and are transported by fluvial processes of the Gomati River during the monsoon season. The Gomati River transports weathering products of the GAP to the Ganga River as its bedload, suspended and dissolved loads (Fig. [2c](#page-2-0)).

Sample details

Drainage area of the Gomati River has been selected for the present study. Samples of the Behta Nala Cliff section were categorized as the unweathered GAP sediments. The bed and suspended sediment samples of the Gomati River were categorized as weathered sediments of the GAP. The 9 mhigh cliff section along the Behta Nala (a small tributary of the Gomati River) was exposed near Lucknow. A set of 16 sediment samples were collected from the cliff as shown in Fig. [3](#page-3-0)a, b. A set of 15 Gomati River bed sediments were collected from freshly deposited channel bar of the Gomati River. A set of 17 Gomati River suspended sediments were collected from fresh sediment deposits from the river's floodplain. All locations of the Gomati River is shown in Fig. [3](#page-3-0)c. Sample locations were carefully chosen to avoid contamination from river or cliff sediments and from anthropogenic reworking activities. All sediment samples were collected during the summer season. The Gomati

Fig. 2 a Monthly rainfall, maximum and minimum temperature variation at Lucknow along with four seasons: the winter, the summer, the monsoon and the post-monsoon, **b** the Gomati River hydrograph displays the seasonal variability of water discharges [[9](#page-5-0)],

River flows at very low discharges and exposes its channel bed with bed load deposits in the form of mid-channel bar (Fig. [3](#page-3-0)d). A total number of 48 sediment samples weighing about 100–200 g were collected with the help of plastic scoop and were sealed in clean polythene bags with proper labels.

Sample irradiation and radioactive assay

All sediment samples weighing around 100–200 mg each were packed in a polythene packet and irradiated in CIRUS reactor at Bhabha Atomic Research Centre, Mumbai for 7 h in a neutron flux of 5×10^{13} cm⁻² s⁻¹ along with IAEA RM SOIL-7 which was used as reference material. The samples were assayed after 2 days of cooling for their gamma activity using a 40% HPGe detector coupled to an 8 K Channel Analyzer. The resolution of the detector was found to be 1.8 keV at 1,332 keV. The peak areas corresponding to gamma rays of interest were evaluated using peak-fit software PHAST (Process hazard Analysis Software Tool), developed at Bhabha Atomic Research Centre, Mumbai for data acquisition and analysis [\[7](#page-5-0)].

Results and discussion

The measured value of As concentration in sediments of the GAP and the Gomati River has been presented in Table [1](#page-4-0). In

c physical and chemical weathering processes operating on the Ganga Alluvial Plain generate weathering products transported by the Gomati River as dissolved, bedload and suspended load mainly during the monsoon season

the GAP sediments, As concentration varies from 3.04 to 19.56 mg kg^{-1} with average value of 10.44 mg kg^{-1} . As concentration varied in the Gomati River bed and suspended sediments from 0.12 to 4.36 mg kg⁻¹ and 0.66 to 14.91 mg kg^{-1} , respectively. In the GAP and the Gomati River sediments, As concentration variability may be due to their mineralogical and sedimentological inconsistency. Arsenic concentrations in the GAP sediments fall within the range of As concentration in the core of the Ganga Delta sediments [\[8](#page-5-0)]. Average As concentration of the Gomati River bed sediments $(4.15 \text{ mg kg}^{-1})$ is lower than the Gomati River suspended sediments (14.91 mg kg^{-1}). It is due to higher content of quartz and lower content of clay minerals in the bed sediments than the suspended sediments.

Figure [4](#page-4-0)a displays box and whisker diagram showing 10th, 25th, 50th, 75th and 90th percentiles of As distribution in sediments and weathering products of the GAP. Compared to the GAP sediments, As content in bed and suspended sediments of the Gomati River is markedly decreased by weathering processes of the GAP. This depletion of As content may have been caused due to high mobility of As during the GAP weathering as a result of which the Gomati River has dissolved load as shown in Fig. [4b](#page-4-0). During the monsoon season, rainwater reacts with sub-surface sediments of the GAP. As ions originate by incongruent dissolution of biotite and other ferro-magnesium minerals and are preferentially moved by solution to the Gomati River water. The Gomati River water shows distinct enrichment of As Fig. 3 a Litholog of the Behta Nala Cliff section showing sampling locations (A1-A16). b Field photograph showing the upper part of the exposed Behta Nala Cliff section exposed near Lucknow. c Sampling location map of the Gomati River (Gomati River-G1-G18, Gomati River Tributaries-T19-T23). d The Gomati River at Chandwak showing midchannel bar composed of the bed sediments and floodplain composed of the suspended load sediments

compared to the world average river water of 1.7 μ g L⁻¹. Typical As concentrations in fresh water are frequently less than 1.0 μ g L⁻¹ [\[2\]](#page-5-0). Total dissolved Fe and As concentrations in the Gomati River water range from 47.84–120.23 μ g L⁻¹ and 1.29–7.16 μ g L⁻¹, respectively. These dissolved Fe and As concentrations were well correlated ($r^2 = 0.68$), indicating their common source present in minerals of the GAP sediments [[9](#page-5-0)].

Arsenic is released from unconsolidated sediments of the GAP and mineral biotite is considered as source of As in the unconsolidated sediments of the GAP [[10,](#page-5-0) [11\]](#page-5-0). Biotite

weathers very easily and may be converted to kaolinite. In the process of kaolinization of biotite, the weathering reaction requires chemical changes by addition of $H⁺$ and release of K^+ , Al³⁺, Mg²⁺, Fe²⁺, Ti⁴⁺, Si⁴⁺ and H₂O [\[12](#page-5-0)]. $K_{0.65}(Al_{1.10}Ti_{0.15}Fe_{0.50}Mg_{0.55})(Si_{3.20}Al_{0.80})O_{10}(OH)_{2}+8H^{+}$ $=(\text{Al}_{1,71}\text{Fe}_{0,31}\text{Ti}_{0,03})\text{Si}_2\text{O}_5(\text{OH})_4+0.65\text{K}^++0.19\text{Al}^{3+}$ $+0.55Mg^{2+}+0.19Fe^{2+}+0.12Ti^{4+}+1.2Si^{4+}+3H_2O$

Element mobility in river water is defined as a ratio of dissolved concentration and upper continental crust

Table 1 Concentration of As (in mg kg^{-1}) in alluvial sediments (A1-A16), bed and suspended sediments collected from the Gomati River and its tributaries (G1-G 18, T19-T23) draining the Ganga Alluvial Plain (See Fig. [3](#page-3-0) for locations)

Gomati River Basin (alluvial sediments) $(n = 16)$ Gomati River (bedload sediments) $(n = 15)$				Gomati River (suspended load sediments) $(n = 17)$	
Sample no.	As conc. $(mg kg^{-1})$	Sample no.	As conc. $(mg kg^{-1})$	Sample no.	As conc. $(mg kg^{-1})$
A1	13.81	G ₂	0.58	G1	6.48
A2	13.33	G ₃	0.94	G ₂	5.18
A ₃	4.85	G ₅	0.91	G ₃	4.59
A4	6.65	G ₆	0.12	G4	14.91
A ₅	7.44	G7	0.47	G ₅	8.75
A ₆	6.63	G8	1.40	G ₆	6.32
A7	4.70	G10	2.06	G7	6.24
A8	8.44	G11	1.48	G8	4.86
A9	13.81	G12	1.83	G9	2.62
A10	16.24	G13	1.34	G10	4.51
A11	3.04	G14	0.50	G11	6.78
A12	3.35	G15	4.15	G12	8.13
A13	17.00	G16	0.60	T19	0.86
A14	17.26	G17	1.67	T ₂₀	0.66
A15	19.56	G18	2.28	T ₂₁	1.29
A16	10.94			T ₂₂	5.88
				T ₂₃	2.08
Minimum	3.04	Minimum	0.12	Minimum	0.66
Maximum	19.56	Maximum	4.15	Maximum	14.91
Average	10.44	Average	1.36	Average	5.30
SD	5.47	SD	1.00	SD	3.48

Fig. 4 a Box and whisker diagram showing distribution of As concentrations in the Ganga Alluvial Plain sediments, the Gomati River bed and suspended sediments. Arrow indicates a simple assessment of As mobility using depletion of As content due to the Ganga Alluvial Plain weathering processes, b distribution of dissolved As content in the Gomati River water during the winter season [[14](#page-5-0)]

concentration compared to the value for sodium. The classification of element mobility comprises the highly mobile elements (close to or greater than sodium), moderately mobile elements $({\sim}10$ times less than sodium), non-mobile elements (\sim 10–100 times less than sodium) and immobile elements $(>100$ times lower than sodium). Elements of one group can pass to another group depending on weathering condition [\[13](#page-5-0)]. In recent study, a set of the Gomati River water samples have been analyzed for their dissolved major and trace elements concentration to assess the mobility of these elements by weathering process of the GAP [[14\]](#page-5-0). Results display that relative mobility of elements follows the trend $\text{As} > \text{Mg}$ $Sr = Na > Ca > K > Cr > Zn > Rb > Ni > Cu > Si >$ $Ba > Fe > Pb > Al > Mn$ as shown in Fig. [5.](#page-5-0) Geochemical mobility of As is comparable to Mg, Sr and Na in the

Fig. 5 Normalized bar diagram of dissolved major and trace elements in the Gomati River (Cw) to Upper Continental Crust abundances (Cc). All values are compared to that of sodium. Bar heights show elemental mobility by weathering processes of the Ganga Alluvial Plain. Note the high mobility of As comparable with alkali elements such as Mg, Sr and Na [14]

GAP. It is of primarily importance due to control of As content in surface water and groundwater that define availability to participate in various components of the GAP environment.

Conclusions

Arsenic distribution in natural waters of the earth has scientific and societal issues across the world. Geogenic distribution of As in sediments the GAP is more or less same as in sediments of the Ganga Delta region. It is perhaps As mobilization from these biotite containing sediments by natural weathering processes to enter into the hydrological system. Therefore, proper understanding of As mobility from alluvial sediments is essential to provide safe and reliable drinking water along with to address the human health issues that influence nearly 500 million people living in the GAP and the Ganga Delta regions of the world.

Acknowledgments Authors are thankful to Head, Department of Geology, University of Lucknow, for providing infrastructure facility

for this work and to Mr. Pramod Kumar Joshi for making diagrams. IBS is thankful to Indian National Science Academy, New Delhi for providing the INSA Senior Scientist Position. Financial support from University Grants Commission, India—Department of Atomic Energy, India consortium (CRS/CD/MUM/CRS-M-129/07/2837) is also gratefully acknowledged.

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