

Mobilization of Arsenic in the Groundwater of Some Char Lands in Meghna Basin, Bangladesh: A Mechanistic Study

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Received July 19, 2013; revised and accepted September 3, 2014

Abstract: In this paper, we describe the mechanistic details for arsenic mobilization in some newly formed char lands in Meghna basin, Bangladesh. We measured the concentrations of arsenic along with some other associated parameters involved in the release mechanism of arsenic in sediment-water interface. Total 38 water samples were collected and analyzed from both shallow (60 to 110 feet) and deep (≥ 200 feet) aquifers and arsenic concentration ranged from 0 to 2.5 mg/L with an average value of 0.4266 mg/L. We observed three arsenic contaminated hot spots in the shallow aquifers where one spot's arsenic concentration is as high as 2.5 mg/L which could be the second highest arsenic contaminated spot in Bangladesh reported so far. Concentration of arsenic exhibits a clear positive correlation with iron and also with ammonium, bicarbonate and phosphate significantly. From the study, it reveals that reductive dissolution of arsenic-rich iron minerals is primarily responsible for the mobilization of arsenic in the groundwater. The presence of other competitive ions such as phosphate and bicarbonate in the adsorption-desorption process of arsenic were identified for facilitating the mobilization.

Key words: Arsenic mobilization, char lands, adsorption/desorption, biogeochemical transformations, competitive ions.

Introduction

For decades presence of arsenic in groundwater has been a public health concern due to its acute toxicity in many parts of the world (Nikson et al., 1998; McArthur et al., 2001; Smedley and Kinniburgh, 2002; Smedley et al., 2003; Safiullah, 2006; Cullen, 2008) where millions of people are exposed to groundwater arsenic above the maximum permissible limit (0.001 mg/L) set by the World Health Organization (WHO) and US Environmental Protection Agency (US EPA) (WHO, 2011; US EPA, 2012). Arsenic as a public health problem was first identified in the Indian state of West Bengal in Gangetic delta. Since West Bengal and Bangladesh

are in the same deltaic plain, environmental scientists started looking for similar problem in Bangladesh and the presence of arsenic was first reported in the groundwater of Chapai Nawabganj district in 1993 by the Department of Public Health Engineering of the Government of Bangladesh (DPHE, 1993).

Arsenic is introduced to the environment mainly from natural sources intensified by anthropogenic activities. Once released, it may exist in both organic and inorganic forms with various oxidation states (-3 , 0 , $+3$, $+5$) depending on the redox potential, pH and the microbial activities of the system (Smedley and Kinniburgh, 2002). Mobilization of arsenic is a self organizing site specific geochemical process that can take place

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in a variety of ways (Smedley and Kinniburgh, 2002; Tareq et al., 2003). The mechanism of arsenic release from sediment to the aquatic environment is not clearly understood yet. Two well-known theories about the mechanism of arsenic mobilization are put forward: pyrite oxidation (Bhattacharaya et al., 1997; Nikson et al., 1998; McArthur et al., 2001; Safiullah, 2006) and oxyhydroxide reduction (Smedley and Kinniburgh, 2002; Safiullah, 2006). The toxicity of arsenicals depends on their concentration in aqueous phase, their speciation and the nature of interactions with environmental surfaces (Cullen, 2008). Thus, it is essential to know the mechanism for the biogeochemical transformations of arsenicals as well as their mobilization to the aquatic environment for the risk assessment in a given contaminated site of interest.

A number of studies have been reported on the occurrence of arsenic in the groundwater of Bangladesh mostly in local scales (Harvey et al., 2002; Swartz et al., 2004; Zheng et al., 2004; Hasan et al., 2009). The Meghna basin located in the southeast part of the country is heavily affected by arsenic contamination in its groundwater where levels of arsenic are beyond the guideline of WHO, US EPA and the maximum permissible limit set for drinking water in Bangladesh (Anawar et al., 2003; Bibi et al., 2008; Ahmed et al., 2010). Recently, we reported the significant presence of arsenic in the coconut water especially in a part of Meghna basin (Safiullah et al., 2013). The above

studies clearly described the needs for understanding the sources of pollution and a systematic monitoring for the level of contamination to predict the extent of pollution in arsenic prone areas in near future. Taking these objectives into account, the present study as a continuation of arsenic research was undertaken in some newly formed Char lands in Meghna basin of Bangladesh.

Study Area

Sonargaon upazila located in Meghna basin is one of the most arsenic affected areas in Bangladesh. Pirijpur union, one of 11 unions under Sonargaon, has 4149 households with nearly 32,382 people according to the upazila Statistical Office. Considering the geographical importance of the groundwater arsenic, the three villages—Char Bhabanathpur, Naruttampur and Jainpur (Figure 1)—were selected for investigation located on the bank of Menikhali canal which falls into the river Meghna in east, 1 km away from the study area. The canal connects the river Brahmaputra also from the western side of the Jainpur at a distance about 9 km. The south-eastern side of the villages is a vast low lying agricultural field and extends right up to the levee of the Meghna where deposition of sediments from the upper stream is evident. It is known from the local people that the deposition of sediments has been occurring over the centuries causing the villages to be emerged. The

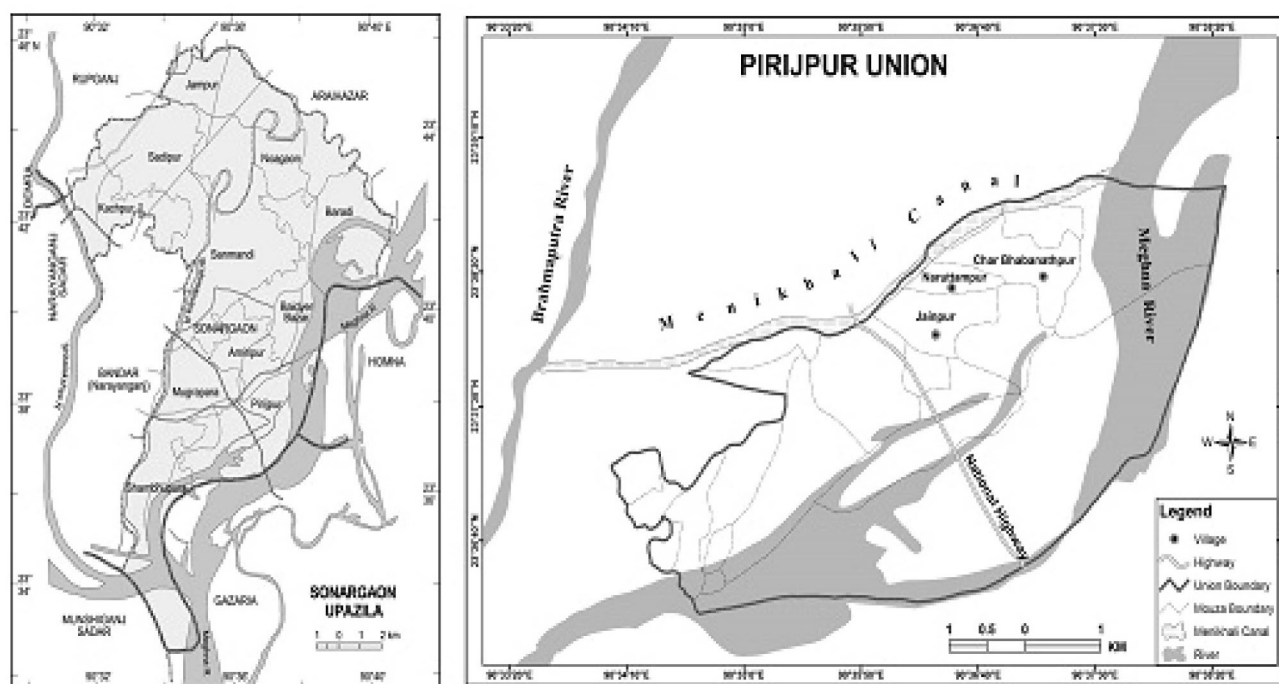


Figure 1: Location of the study area in Pirijpur union under Sonargaon upazila, Bangladesh.

presence of high level of arsenic in these areas could possibly be due to the co-deposition of arsenic bearing minerals with the sediments coming from Himalayan river Meghna. Low income group people are generally living in these villages with arsenic contaminated water to meet their daily purposes along with other poor living facilities.

Materials and Methods

Sample Collection

Sampling sites were chosen on the basis of the availability of open wells and in a manner so that the study covers the whole area uniformly. Total 38 groundwater samples were collected during the month of February-March, 2009 from both hand and deep tubewells of different depths in order to understand the whole geochemical processes. The cut-off depth for shallow and deep aquifers was taken as 110 ft because the groundwaters deeper than 110 ft show a significant decline in arsenic concentration in most of the Holocene alluvial aquifers of Bangladesh (BGS/DPHE, 2001). Before sampling, the tubewells were pumped for several minutes to stabilize the water coming out.

The water samples were collected in acid-prewashed high-density polyethylene bottles (rinsed with 1% HNO₃ followed by thorough rinsing with distilled water). The water high in iron content may transform into ferric hydroxide in contact with air and may get precipitated scavenging a fraction of arsenic present in water (Safiullah, 2006). To overcome the situation, samples for arsenic and iron analysis were slightly acidified by the addition of few drops of nitric acid. Samples for the analysis of ammonium, nitrate/nitrite, bicarbonate and phosphate were preserved without acidification. All the water samples were stored at controlled temperature and analyzed within two weeks of their collection.

Analytical Methods

For the measurement of total arsenic, spectrophotometric method (M-390, Taiwan) was used based on the arsine (AsH₃) generation by an acid-zinc reaction, which was then scrubbed through lead acetate impregnated glass wool and finally was absorbed by silver diethyldithiocarbamate (SDDC) dissolved in pyridine. The colour developed by the AsH₃-SDDC complexation was photometrically measured at 535 nm. Concentrations of arsenic were determined from standard curve which was prepared previously by using Aldrich arsenic standard. Total iron concentration in water samples was measured spectrophotometrically

using 1, 10-phenanthroline (λ_{\max} 510 nm) against a reagent blank as usual. Bicarbonate alkalinity was determined by titration method. Phosphate, ammonium and nitrate/nitrite were estimated by using field test kits (E-Merck, Germany) and pHs were determined during the sample collection with a digital pH meter (CG-818, Schott Gerate).

Results and Discussion

The occurrence of arsenic in both the shallow and deep aquifers was investigated and three hot spots were detected in the study area; two are in Char Bhabanathpur with 1.5 and 2.0 mg/L arsenic, respectively. The other is in Jainpur which is perhaps the second highest arsenic concentration (2.5 mg/L) detected so far in Bangladesh followed by 4.7 mg/L of arsenic reported by Chakrabarti et al. (2010) in a hand tubewell of the village Chiladi in Noakhali district. All these three high arsenic contaminated sites in our study were observed in the shallow aquifers. The people of Jainpur have already installed deep tubewells for lifting groundwater since the discovery of high groundwater arsenic contamination in the area. For this reason, no other operative shallow tubewells were found surrounding the hot spot.

The arsenic concentration in groundwater is relatively high in Naruttampur and Jainpur compared to Char Bhabanathpur. The eastern part of Char Bhabanathpur is relatively less contaminated by groundwater arsenic compared to its western part. This can be explained in terms of some factors we are proposing here: (i) the eastern part is relatively closer to the Meghna and hence its geological development by the river sediments occurred bit later, (ii) for the same reason, arsenic from the soil-sediments are washed out easily by natural floods, and (iii) aquifer in this area can easily be recharged by the river water as well as by flood water than the western part and this might lead to the reduction of arsenic concentration in the aforementioned site. The low groundwater arsenic concentration is detected in deep tubewells in some parts of Jainpur also and tubewells having depth of 300 ft or more had no groundwater arsenic within the capacity of our method. But in all cases, tubewells of depth 50-110 ft have very high arsenic concentration in all the three villages undertaken in our study. The concentration of arsenic in Jainpur is very high because of the deposition of sediments from the upper stream of the Meghna over 3 to 4 decades as opined by the villagers. The results obtained from the measurements show a distinct pattern of arsenic contamination: the study area

has two hotspots where each hot spot is surrounded by a concentric pattern and there is an area between them which is completely free from arsenic. These patterns have been interpreted previously as the paleo depositional profile of sediments and paleo-oxbow lakes of the study areas (McArthur et al., 1999; Safiullah, 2006). For understanding the release mechanism of arsenic in the water-sediment interface of these areas, we measured other associated parameters ($\text{Fe}^{2+}/\text{Fe}^{3+}$, PO_4^{3-} , NH_4^+ , $\text{NO}_2^-/\text{NO}_3^-$ and HCO_3^-) along with arsenic and their concentrations have been shown against the depth of the wells in Figure 2.

The results of our study demonstrates that shallow aquifers enriched in arsenic are also enriched in iron (10-25 mg/L), ammonium (2-8 mg/L) and phosphate (>17 mg/L) where the deep aquifers contain low levels of arsenic and the other parameters. The $\text{NO}_2^-/\text{NO}_3^-$ concentrations are significantly low within the groundwater pHs 6.5 to 7.0 (not shown here). We mentioned in our introduction part that arsenic concentration varies from one aquifer to another. Based on the geology and soil characteristics, arsenic concentrations in groundwater may be different in the aquifers even having the same depth. The depths of the wells were considered depending on the information given by the corresponding households. In our study, high arsenic concentrations were found within the depth

of 50-110 ft which is in complete agreement with other studies (BGS/DPHE, 2001; Anwar et al., 2003; Ahmed et al., 2010).

The contrast in arsenic concentrations between shallow and deep aquifers can be explained by resorption of arsenic onto residual FeOOH (McArthur et al., 2004) and by the nature of the aquifer sediments (Zheng et al., 2004). Pirijpur union has been in vulnerable condition due to high groundwater arsenic in its shallow aquifer. To avoid arsenic contaminated water from shallow aquifer, people of relatively high income group have already started installing deep tubewells as a source of alternative arsenic-free safe water. We found high concentrations of $\text{Fe}^{2+}/\text{Fe}^{3+}$, NH_4^+ , PO_4^{3-} and HCO_3^- with high arsenic concentration in shallow aquifers but in the deep aquifers only bicarbonate was present in significant amount. Figure 3 shows how arsenic correlates with other compatible species in the shallow aquifers.

From the results of our study, it is clear that concentrations of arsenic and iron present in the shallow aquifers are above the drinking water guidelines in Bangladesh (The Environmental Conservation Rules, 1997) and also beyond the standard set by WHO and US EPA (WHO, 2011; US EPA, 2012). We also observed that arsenic correlates strongly with iron ($R^2 = 0.9054$) which describes that presence of arsenic in

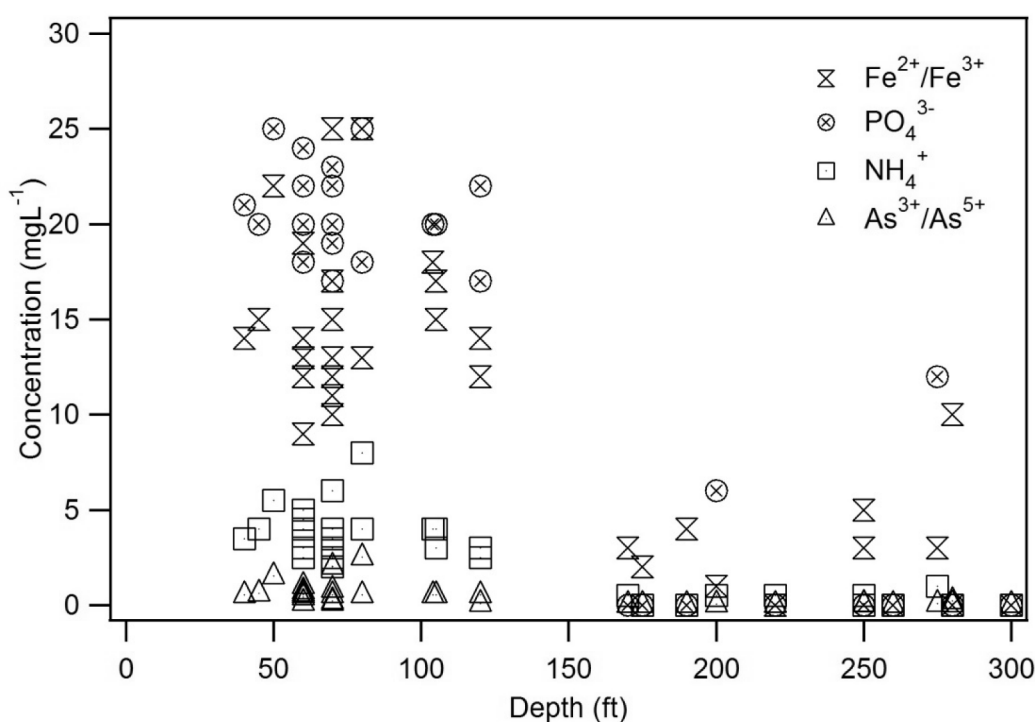


Figure 2: Concentrations of As, $\text{Fe}^{2+}/\text{Fe}^{3+}$, PO_4^{3-} , NH_4^+ as a function of depth.

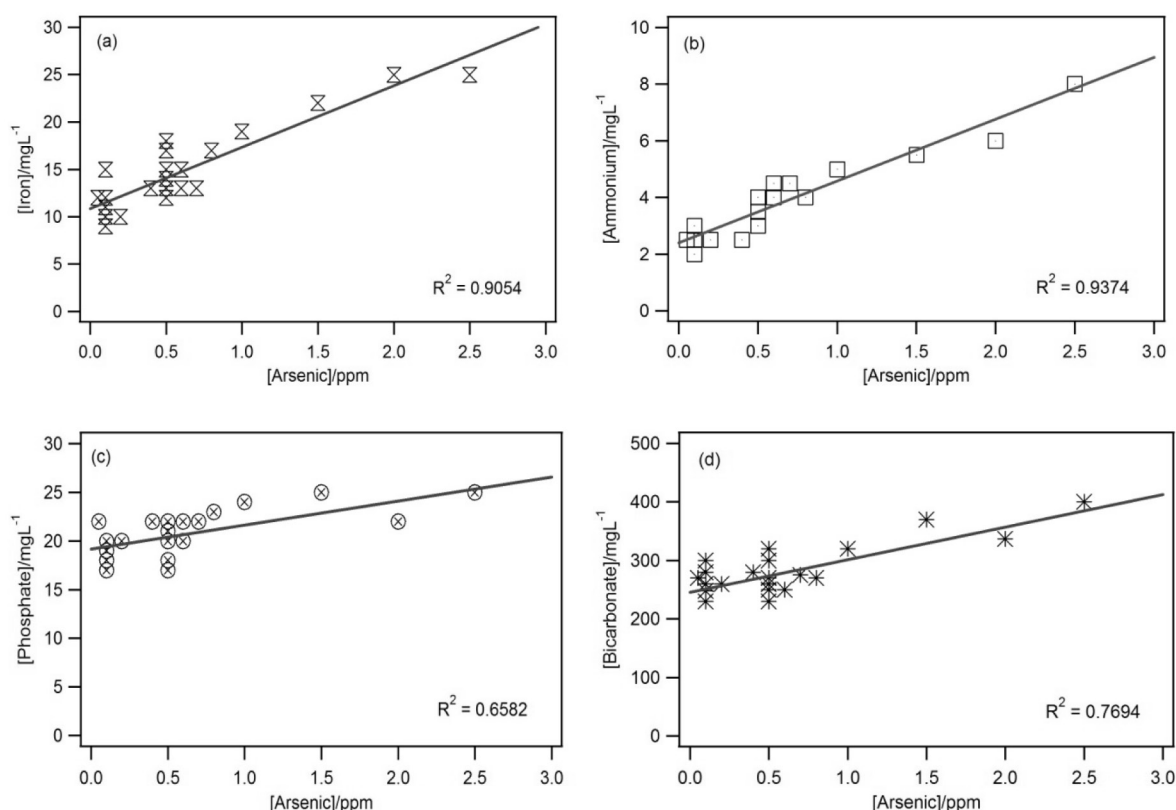


Figure 3: Correlations between (a) iron; (b) ammonium; (c) phosphate; and (d) bicarbonate with arsenic in the shallow aquifer of the study area.

the groundwater is somehow related to the dissolution of iron minerals. The high level of bicarbonate along with low of nitrate concentration might be the indication of reducing aquifer environment (Nickson et al., 2000). Concentrations of dissolved HCO_3^- reflect the degree of water-rock interactions in groundwater systems as well as integrated microbial degradation of organic matter (Zheng et al., 2004). Groundwater alkalinity or HCO_3^- originates from weathering of calcite or silicate minerals by atmospheric or respired CO_2 that leads to secondary mineral formation (Garrels and Mackenzie, 1967). Concentrations of arsenic correlates significantly ($R^2 = 0.7694$) with the concentrations of HCO_3^- in the groundwater of these areas.

Bicarbonate has been considered as a possible cause of high arsenic concentrations in the shallow aquifers in other parts of Bangladesh (Appelo et al., 2002; Anwar et al., 2004). The stability of many contaminants in groundwater is dependent on the redox aquifer conditions. Microbial degradation of the organic matter in the aquifers intensifies the reduction of NO_3^- , thereby increasing the concentrations of NH_4^+ (Mukherjee et al., 2008). The study area is located nearby paddy fields where some marshy lands are in the close vicinity of the

tubewells. Most of the tubewells are also located nearby latrine or in such a place where cow dung are kept. Ammonium ion is an important redox sensitive species and its concentration above 2 mg/L in groundwater indicates sewage input or ammonification (Nath et al., 2008). The significant correlation between arsenic with bicarbonate and ammonium and also the low level of nitrate/nitrite describe the microbial controlled reducing groundwater environment. In an overall explanation, the presence of arsenic in the shallow aquifer of the study area can be addressed by the reductive dissolution of arsenic-rich iron minerals; mainly oxide or (oxyhydr) oxides with respect to the other studies (Smedley and Kinniburgh, 2002; Safiullah, 2006).

Agricultural wastes, an important source of phosphate in groundwater, are decomposed due to water logging and in under wet condition finally leaches out to the shallow aquifers. The concentrations of phosphate in the study area are also as high as 25 mg/L. Most of the arsenicals are present in the aquatic body in anionic form as explained by their corresponding pKa values (Nuallain and Cinneidene, 1973; Dzombak and Morel, 1990; Al-Abadleh and Hoang, 2007). Phosphate is structurally similar to arsenate which may undergo

competitive adsorption with arsenicals to the same active binding sites of soil geosorbents i.e. Fe/Al/Mn-oxides present in the aquatic system (Smedley and Kinniburgh, 2002). A number of laboratory and field studies have been carried out for desorption of arsenicals (both organic and inorganic) from the reactive soil components where aqueous phosphates have been proved as potent desorbing agents (Brown et al., 2005; Jackson et al., 2006; Tofan-Lazar and Al-Abadleh, 2012; Arts et al., 2013). In our study, presence of high level of phosphate might be responsible for enhancing the release of arsenicals from the adsorbed phase. Though not structurally similar to phosphate, importance of the significant presence of anionic bicarbonate in the release mechanism of arsenic from the metal (oxyhydr)oxides surfaces cannot be ignored (Wang and Mulligan, 2006).

Conclusion

Mobilization of arsenic in sediment-water interface is a complex biogeochemical process. High groundwater arsenic along with high concentration of iron predicts that arsenic could be releasing to the aquatic system from iron-rich sediments. Presence of significant concentration of bicarbonate in both the shallow and deep aquifers has been identified for causing the reducing groundwater environment. In addition, this reducing aquifer condition is partially dominated by the microbial activities of the system as described by the high ammonium and low nitrate concentration. The significant presence of phosphate in the groundwater might play an important role for facilitating the release of arsenicals from iron-rich sediments. So taking into account the adverse effect of arsenic poisoning, groundwater of the affected areas should be monitored on a regular basis to perceive the extent of pollution and to understand the mobilization of arsenic from highly contaminated shallow aquifer to less contaminated deep aquifer, if any.

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Calendar of Events

Stormwater 2014 - 3rd National Conference on Urban Water Management

13th to 17th October 2014

Adelaide, South Australia, Australia

Website: <http://www.stormwater2014.com.au>

Contact person: Julie McGraw

Organized by: Stormwater Australia

2nd International Conference on Sustainable Environment and Agriculture (ICSEA 2014)

29th to 30th October 2014

San Diego, United States of America

Website: <http://www.icsea.org/>

Contact person: Ms. Eve Li

Organized by: CBEES

4th Journal Conference on Environmental Science and Development (JCESD 2014 4th)

29th to 30th October 2014

San Diego, United States of America

Website: <http://www.ijesd.org/jcesd/4th/>

Contact person: IJESD

Organized by: CBEES

21st Century Watershed Technology Conference and Workshop

1st to 7th November 2014

Hamilton, Waikato, New Zealand

Website: <http://watershedtech.org/>

Contact person: Lea Boodee

Organized by: ASABE and The University of Waikato

WWEM Water, Wastewater and Environmental Monitoring

5th to 6th November 2014

Telford International Centre, St. Quentins Gate, Telford, TF3 4JH, United Kingdom

Website: <http://atnd.it/6117-0>

Contact person: Marcus Pattison

Organized by: WWEM

2014 2nd International Conference on Environment Pollution and Prevention (ICEPP 2014)

12th to 13th November 2014

Auckland, New Zealand

Website: <http://www.icepp.org/>

Contact person: Ms. Eve Li

Organized by: CBEES

VIII WAC- International Conference 2014 Balancing Environment and Development

20th to 21st November 2014

New Delhi, India

Website: <http://www.worldaquacongress.org>

Contact person: Praggya Sharmaa

Global Conference on Energy, Soil, Water, Air and Environment

11th to 13th December 2014

Dubai, United Arab Emirates

Website: <http://www.globalcenter.info/eswae/>

Contact person: Dr. Nehir Varol

International Conference on Environmental Systems Research (ICESR 2014)

13th to 14th December 2014

Kuala Lumpur, Malaysia

Website: <http://www.icesr.org/>

Contact person: Ms. Eve Li

Organized by: CBEES

International Conference on Earth, Environment and Life Sciences (EELS-2014)

23rd to 24th December 2014

Dubai, United Arab Emirates

Website: <http://www.iicbe.org/2014/12/24/53>

Contact person: Conference Secretary-EELS-2014