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Discovery of Radon in Hot Spring Waters of Odisha in Eastern India

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Abstract: We report here for the very first time ever, a discovery of significant presence of radon in hot spring waters of Odisha in eastern India. Samples of water collected from two hot springs of Atri and Tarabalo of Odisha possess Rn in concentrations of 0.03×10^{-10} Ci/l of water and 19.16×10^{-10} Ci/l of water, respectively. While the Rn content of Atri hot spring is low, the Rn content of Tarabalo is significant and comparable to that of the famous hot springs of Hiroshima in Japan that average around 80×10^{-10} Ci/l of water. Uranium bearing sandstones, mudstones and shales of the underlying Athgarh Sandstones of Gondwanas appear to be the source of Rn.

Waters of Atri and Tarabalo contain small amounts of organic matter, and Tarabalo also contains minor counts of bacteria, which could be easily eliminated with proper maintenance, and these two hot springs can be developed into major health and tourist resorts.

Key words: Radon, hot spring, Tarabalo, Atri, Hiroshima.

Introduction

Geological Survey of India has recorded the existence of 340 hot springs spread over the entire landmass of India (Geological Survey of India website; Ravi Shankar et al., 1991). These thermal springs have been classified into six geothermal provinces on the basis of their geotectonic set up. The six provinces are as follows: (i) Himalayan Province; (ii) Areas of faulted blocks (Aravalli Belt, Naga-Lucha, West Coast Regions, Sone-Narmada Lineament, and Gondwana); (iii) Volcanic arc; (iv) Deep sedimentary basins of Tertiary age; (v) Radioactive Province; and (vi) Cratonic Province-Peninsular India. Atri and Tarabalo hot springs of Odisha belong to the second category of geothermal provinces, and occur in fault bound shear zone of the Gondwanas in Mahanadi Graben with post Gondwana and possibly late Tertiary/Quaternary reactivations (Sharma, 2005). None of the 340 hot springs of India have ever been systematically analysed for their radon content.

In this article we report, the first ever measurements of radon in hot spring waters of Odisha in eastern India. We collected samples of water from two hot springs, Atri and Tarabalo, of Odisha. Atri is located at a distance of 40 km west of the state capital Bhubaneswar and bears the coordinates of 20º15`N 85º30`E. Tarabalo, located in Nayagarh District of Odisha, is a cluster of hot springs spread over an area of eight acres, and the coordinates are 20º08`N 85º11`E. Tarabalo is located some 30 km further west of Atri (Figure 1).

Radon is a noble gas with atomic number 86, and has 28 isotopes. Rn^{222}, which is a daughter of Rn^{226} in the uranium-radium disintegration series, is the longest lived isotope of radon with a half life of 3.825 days

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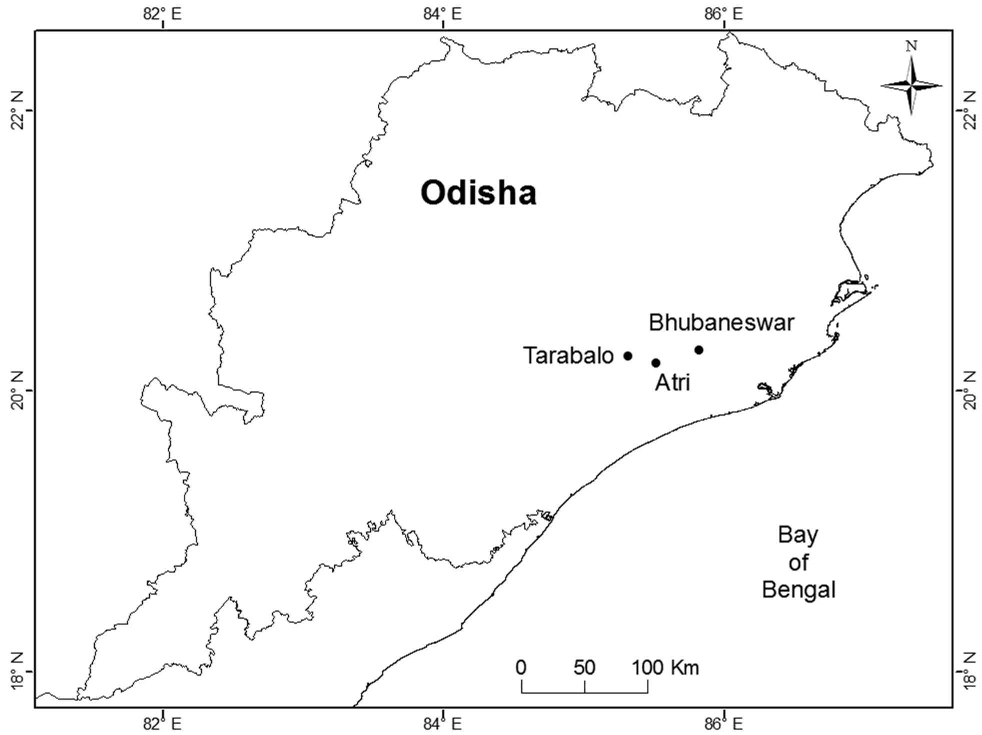

Figure 1: Location map of Atri and Tarabalo hot springs of Odisha.

(Hasegawa and Ohno, 1997). Radon measurement generally involves measuring the contents of Rn^{222} per litre of water (Sawamura, 1980).

Sampling

Samples of water from the hot springs of Atri and Tarabalo were collected on the 27th of February 2011, in 1l polyethylene terephthalate (PET) bottles. The bottles were filled to the brim and the caps were applied tightly to avoid the escape of the dissolved radon. Precise times of collection of the samples (Atri 12 noon, Tarabalo 1 pm) were recorded. The samples of water were then carried by Professor Sasaki in his personal baggage in a commercial flight to Hiroshima for analyses in his laboratories of the Department of Bio-Recycling of Hiroshima Kokusai Gakuin University of Hiroshima in Japan.

Analytical Methods

Samples of water from Atri and Tarabalo were analysed for radon on the 5th of March 2011, exactly six days after their collection, after taking the time difference of 3.5 hours between India and Japan into account.

Radon content of the samples were measured by an IM-Fontactoscope manufactured by Riken Keiki Co Ltd. IM-Fontactoscope is a newer and improved version of the pre-existing fontactoscope, and was invented in 1931 by Dr Satoyasu Iimori (1885-1982) of the Institute of Physical and Chemical Research in Tokyo (generally known as RIKEN) (Saito, 2003).

IM-Fontactoscope, called Senkoukei in Japanese, is an instrument designed for measurement of radon in natural waters. This instrument consists of a 5.920 litre (5920 cc) capacity metal ionisation chamber made of brass, with a provision to hold exactly 500 ml of water to be analysed. A central electrode is inserted through an insulator into the brass chamber, and an aluminium-leaf electroscope is attached to the central electrode. A fibre glass pointer is attached to the top of the aluminium-leaf. The rate of movement of this pointer is observable in the field of view of the microscope (Sawamura, 1980; Hasegawa and Ohno, 1997).

Five hundred milli-litre of water sample was gently poured in to the metal ionisation chamber, which was then shaken vigorously for about one minute to drive radon out of water into the gas phase. Three hours later, the rate of movement of the aluminium leaf, as evidenced from the movement of the pointer in the field of view of the microscope was measured. The measurements are generally carried out at a constant temperature in the range of 10 to 40°C and at a low humidity below 70% (Sawamura, 1980; Hasegawa and Ohno, 1997).

Readings of radon measurements, in Japanese unit Mahe, obtained from IM-Fontactoscope were plotted along a well-documented time series curve of the decay of Rn^{222} to estimate the radon content of samples in Mahe. A time series curve for Rn^{222} decay for water samples of Onomichi Track Station, collected in glass bottles as well as in polyethylene terephthalate (PET) bottles, is considered a standard curve. The results were converted to the international unit, the curie, following the conversion rate of, 5.5 Mahe = 20×10^{-10} Ci (1 Mahe = 3.636×10^{-10} Ci).

Chemical analyses of the waters of hot springs that include determination of total hardness (Ca + Mg), analyses for the various anions and cations, and tests for bacteria were also conducted following standard techniques described at length by Iwanaga et al. (1997), and Sasaki et al. (1996).

Measurements of radon and all the chemical analyses of water samples, stated above, were conducted at the laboratories of Professor Sasaki of the Department of Bio-Recycling of Hiroshima Kokusai Gakuin University of Hiroshima in Japan.

Results

The results of radon measurements and chemical analyses of waters of two hot springs of Odisha, Atri and Tarabalo, are shown in Table 1. Table 1 also presents a complete set of results of waters of two famous hot springs of Hiroshima, Takata Reisenji and Shiraki Akiyama, also analysed in the laboratories of Professor Sasaki, for purposes of comparison.

Discussion

Water temperatures of Takata Reisenji and Shiraki Akiyama hot springs of Hiroshima at 19.9°C and

Table 1: Radon content and water chemistry of hot springs of Odisha and Hiroshima

Name of hot springs	*Atri in Odisha*	*Tarabalo in Odisha*	*Takata Reisenji in Hiroshima*	*Shiraki Akiyama in Hiroshima*
Water temperature	49.7°C	42.0°C	19.9°C	13.2°C
pH	8.18	8.30	6.38	6.0
Smell	Sulphur	None	None	None
Colour	None	None	None	None
Electrical conductivity	1136.0 µS/cm	793.0 µS/cm	89.2 µS/cm	22.9 µS/cm
Total hardness	32.0 mg/l	16.0 mg/l	26.0 mg/l	2.0 mg/l
$KMnO_4$ consumption	12.0 mg/l	5.06 mg/l	<0.01 mg/l	1.90 mg/l
HCO_3^-	36.0 mg/l	64.0 mg/l	24.0 mg/l	2.0 mg/l
Cl^-	252.0 mg/l	177.3 mg/l	11.3 mg/l	7.09 mg/l
NH_4^+ - N	0.20 mg/l	0.20 mg/l	0 mg/l	0.01 mg/l
NO_3^- - N	0.01 mg/l	<0.01 mg/l	<0.01 mg/l	<0.01 mg/l
PO_4^{3-}	<0.01 mg/l	<0.01 mg/l	0.07 mg/l	0.01 mg/l
SiO^2	62.0 mg/l	59.8 mg/l	21.3 mg/l	27.3 mg/l
Na	-	-	8.03 mg/l	3.51 mg/l
K	-	-	1.45 mg/l	0.70 mg/l
Total iron	<0.01 mg/l	<0.01 mg/l	0.01 mg/l	0.01 mg/l
SO_4^{2-}	60.6 mg/l	58.5 mg/l	7.27 mg/l	0.28 mg/l
F^-	5.62 mg/l	6.05 mg/l	0.05 mg/l	0.15 mg/l
Mn	0.03 mg/l	0.03 mg/l	<0.01 mg/l	0.01 mg/l
Estimated total dissolved solids	460.48 mg/l	386.97 mg/l	99.51 mg/l	44.98 mg/l
Coliform bacteria	0 cell/ml	10 cell/ml	0 cell/ml	0 cell/ml
General bacteria	0 cell/ml	36 cell/ml	0 cell/ml	20 cell/ml
Rn estimated value	0.03×10^{-10} Ci/l	19.16×10^{-10} Ci/l	54.33×10^{-10} Ci/l	57.8×10^{-10} Ci/l

13.2ºC, respectively, are rather low. In India, springs emitting waters of such low temperatures will not qualify as hot springs. Definition of hot springs in Japan, however, is quite different, and is based on radon content of waters. Spring water that possesses a radon content of more than 5.5 Mahe (20×10^{-10} Ci) per litre of water, qualifies as a hot spring. Quite interestingly, a perfectly cold water spring like Takata Reisenji or Shiraki Akiyama, qualifies to be a hot spring based on this definition (Das, 2008).

Discussions of the results of radon measurements and chemical analyses of waters of two hot springs of Odisha, Atri and Tarabalo, and their implications are presented in this section.

Presence of Radon

Radon content of the water of Tarabalo hot spring at 19.16×10^{-10} Ci/l of water is lower than the average radon content of the hot springs of Hiroshima valued at 80×10^{-10} Ci/l of water (Das, 2008), and considerably lower than that of the most famous hot spring of Hiroshima, Yuki Onsen, which boasts a radon content of 104.72×10^{-10} Ci/l of water (www.rakudaj.net/onsen/hiroshima/yuki2.htm). Radon content of Tarabalo, however, is reasonably high, and is comparable to radon contents of the famous Takata Reisenji (54.33×10^{-10} Ci/l), and Shiraki Akiyama (57.8×10^{-10} Ci/l) hot springs of Hiroshima.

A little praise of Shiraki Akiyama hot spring is perhaps necessary here to demonstrate the importance of the radon content of Tarabalo. In its heyday during the Edo period also known as Tokugawa period (1603 to 1868 AD) of Japanese history, the famous Shiraki Akiyama hot spring received frequent visits by Governor Asano Naganori (1667-1701 AD) of the powerful Asano clan that governed Hiroshima. The governor drank and bathed in the waters of this hot spring to relax, recover and regain health. Right up to thirty years ago, children from the neighbourhood, bathed in the waters of Shiraki Akiyama hot spring, and anecdotal evidences tell that the children were cured of skin allergies and eye infection. Seen in this context, this discovery of reasonably high radon content of Tarabalo, is quite significant indeed.

Although radon content of Atri at 0.03×10^{-10} Ci/l is rather low, the very presence of radon, is significant and worth a discussion. Waters of Atri have a temperature of 49.7ºC, which is quite high and capable of driving radon gas from the waters into the atmosphere. Better results of radon measurement could most definitely be obtained by analysis of the waters at the hot spring site itself, which unfortunately is not possible due to the fragility and sensitivity of IM-Fontactoscope that prevents transportation by international air travel, and automobile transport on bumpy roads of rural Odisha. A different analytical protocol, therefore, has to be designed for a more accurate measurement of radon content of this hot spring in the future.

Presence of Bacteria and Organic Matter

Water of Tarabalo hot spring of Odisha has a high bacteria count. Count for coliform bacteria stands at 10 cell/ml, whereas general bacteria count is 36 cell/ml. This high bacteria count is essentially due to a lack of proper maintenance. In the hot spring well, we noticed rotting leaves and weeds, which are conducive for a profuse growth of general bacteria. Significant presence of coliform bacteria, evident from their high count, is particularly worrisome, as this indicates contamination of water by fecal matter, obviously due to defecation in the open by people as well as animals. Atri hot spring of Odisha, which is maintained very well, however, has a zero bacteria count. High temperature of water of Atri at 49.7°C also helps in keeping the water clean by not allowing bacteria to grow.

A count of general bacteria of waters of Shiraki Akiyama hot spring of Hiroshima at 20 cell/ml is rather high. This is essentially because this once famous and a very traditional hot spring of Hiroshima is not in use these days. This hot spring, currently nestled amidst rocks and boulders, has dried up to a great extent and lost much of its flow. Lack of use has allowed the growth of moss and decay of leaves along the conduit and along the course of the hot spring, which in turn has fostered a growth of general bacteria. The waters, however, are free from coliform bacteria. Their absence demonstrates that the waters are not contaminated by fecal matter.

Water of Takata Reisenji hot spring in Hiroshima is absolutely clean and free from any presence of any type of bacteria, evident from a zero bacteria count.

Presence of organic matter in waters of three of the four hot springs studied in this article is obvious from the consumption of $KMnO_4$ expressed in mg/l, which when divided by 2.5, even perhaps more accurately by 3, yields an approximate value of COD, expressed again in the same unit of mg/l. The approximation is necessary because of the fact that all organic matter present in a sample of water do not get oxidised by $KMnO_4$ treatment. $KMnO_4$ consumption values of the four hot springs of Atri (12.0 mg/l), Tarabalo (5.06 mg/l), Takata

Reisenji (<0.01 mg/l) and Shiraki Akiyama (1.9 mg/l) correspond to approximate COD contents of 4 mg/l, 1.68 mg/l, 0 mg/l and 0.63 mg/l, respectively. These low COD values indicate the presence of small quantities of organic matter in the waters of Atri, Tarabalo and Shiraki Akiyama hot springs.

Water Chemistry

Waters of Atri and Tarabalo have total hardness of 32.0 mg/l and 16.0 mg/l, respectively, which compare well with waters of hot springs of Takata Reisenji (26.0 mg/l) and Shiraki Akiyama (2.0 mg/l) of Hiroshima, well known for excellent soft waters (Das, 2008). Total hardness, which is a measure of the combined concentration of Ca and Mg, qualifies the water as soft or hard. Since the Japanese are very particular about quality, they adhere to a rather strict definition of soft water, which should not possess a total hardness of more than 100 mg/l. In fact currently there is a move, orchestrated by water quality experts like Professor Sasaki, to raise the standards in Japan even higher by lowering the total hardness to 50 mg/l as the upper limit for soft water.

Contents of Total Dissolved Solids of waters of both Atri and Tarabalo at 460.48 mg/l and 386.97 mg/l, respectively, although are higher than those of the two reference hot springs of Takata Reisenji (99.51 mg/l) and Shiraki Akiyama (44.98 mg/l) of Hiroshima, are quite low. Similarly, HCO_3^- contents of waters of Atri and Tarabalo at 36.0 mg/l and 64.0 mg/l, respectively, although are higher than the two hot springs of Takata Reisenji (24 mg/l) and Shiraki Akiyama (2 mg/l) of Hiroshima, are again quite low. Waters of Atri and Tarabalo because of their low total hardness, low contents of Total Dissolved Solids and low HCO_3^- concentrations are soft waters.

Values of pH of hot waters at Atri and Tarabalo are as high as 8.18 and 8.30, respectively, which is essentially due to higher concentrations of HCO_3^- contents and higher values of Total Hardness. Much higher electrical conductivities of Atri and Tarabalo at 1136.0 µS/cm and 793.0 µS/cm, respectively, than the two reference hot springs of Takata Reisenji (89.2 µS/cm) and Shiraki Akiyama (22.9 µS/cm) of Hiroshima are essentially due to much higher TDS concentrations than the two reference springs of Hiroshima.

Smell of sulphur is quite strong in the waters of Atri hot spring, and none of the other three hot springs discussed in this article, emits smells of sulphur. Atri hot spring incidentally has the lowest radon content. Smell of sulphur, however, is unrelated to radon content, as is the case in countless hot springs in Hiroshima which emit smells of sulphur and contain high concentrations of radon. Presence of NH_4^+ at 0.20 mg/l is noticed in waters of both Atri and Tarabalo hot springs. Waters from these two hot springs, however, contain very small amounts of NO_3^- recorded at 0.01 mg/l or less. Presence of NH_4^+ is perhaps due to contamination from paddy fields that surround the two hot springs. Ammonium nitrate, which is a common fertiliser, however, does not appear to be the source of this NH_4^+, as an aforesaid source would be evident from a significantly higher concentration of NO_3^-. The NH_4^+ contaminants very likely percolate from the paddy fields into the aquifers below which act as the sources of waters that eventually get released at the hot springs.

Sources of Waters

Observations of low total hardness of waters of Atri and Tarabaolo at 32 mg/l and 16 mg/l, respectively, and their low HCO_3^- contents of 36 mg/l and 64 mg/l, respectively, imply that the waters did not drain limestone terrains. Waters from the two reference hot springs of Hiroshima also show a similar, low total hardness and low HCO_3^- contents, which rule out a limestone source. The hot springs in Hiroshima in fact drain late Cretaceous granites (Sasaki et al 1992). Waters of Takata Reisenji and Shiraki Akiyama hot springs of Hiroshima moreover, contain low concentrations of Cl^- (11.3 mg/l and 7.09 mg/l, respectively), low concentrations of SO_4^{2-} (7.27 mg/l and 0.28 mg/l, respectively), and high concentrations of SiO_2 (21.3 mg/l and 27.3 mg/l, respectively), which further corroborate the circulation of waters in granitic terrains.

Waters of Atri and Tarabalo hot springs of Odisha on the other hand contain high concentrations of Cl^- (252.0 mg/l and 177.3 mg/l, respectively), high concentrations of SO_4^{2-} (60.6 mg/l and 58.5 mg/l, respectively) in addition to high concentrations of SiO_2 (62.0 mg/l and 59.8 mg/l, respectively), which indicate circulation of water in a siliceous sedimentary terrain. Such a terrain does indeed exist within the Gondwana shear zone in Mahanadi Graben bounded by faults (Nanda and Mohanty, 2006), which hosts the hot springs of Atri and Tarabalo. Numerous cracks and crevasses along the sheared zone in Gondwanas allow percolation of meteoric waters into the siliceous formations of Athgarh Sandstone which is composed of sandstones, shales, and conglomerates (Pandya, 2006). Meteoric waters very likely circulate in these sedimentary rocks before being released at the hot springs.

Sources of Radon

Sandstones, mudstones and shales contain U just like the granites do (Das and Mackenzie, 1991), and U is the source of Rn (Das, 2008). Athgarh Sandstone in addition to containing quartz arenite and lithic arenite, contains carbonaceous shale, ferruginous shale, argillaceous shales, mudstones and conglomerates (Mishra, 1998). Quartz and lithic arenites of Athgarh Sandstone contain ilmenite (about 1%, by volume, of the total), zircon (about 0.5% of the total) and smaller amounts of rutile, tourmaline and apatite (Mishra, 1988), all of which contain U. Shales and mudstones of Athgarh Sandstone also contain U. In fact ICP-MS analyses of whole rock samples of sandstones and mudstones of Athgarh Sandstone carried out in July 2009 at National Geophysical Research Institute (NGRI), Hyderabad, show U concentrations of the order of 4 ppm (personal communication, Mishra, 2011). We therefore consider the uranium bearing sandstones, mudstones and shales of Athgarh Sandstone to be the source of Rn found in waters of hot springs of Atri and Tarabalo.

Presence of Fluoride

Waters of Atri and Tarabalo contain 5.62 mg/l and 6.05 mg/l of F^-, respectively. Although these F^- concentrations are not as high as 20 ppm noted in the waters from Tattapani hot spring in Chattisgarh state (Sharma, 2003) in eastern India, are well above the maximum acceptable limit of 1.0 mg/l and maximum allowable limit of 1.5 mg/l of water in India (ISI, 1983).

Conclusion

Presence of radon in waters of Atri and Tarabalo is indeed a cause for celebration. Atri hot spring is reasonably well developed as a health and tourist resort whereas Tarabalo hot spring is totally undeveloped. High radon content of waters of Tarabalo is in fact an invitation for this hot spring to be developed into a major health care resort, like Odisha Tourism Development Corporation had proposed in 1994. The Government of Odisha may take up the development of Tarabalo hot spring complex into a major tourist and health care resort. Atri hot spring should also be further developed.

This article underlines the importance for systematic investigations to record the presence of radon in the waters of hot springs of India. The discovery of radon is likely to generate interest among our fellow earth-scientists for further research in this field.

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www.rakuda-j.net/onsen/hiroshima/yuki2.htm (Rn content of Yuki Onsen of Hiroshima).

Calendar of Events

2nd International Conference on Waste Management, Ecology and Biological Sciences (WMEBS-2015)
3rd and 4th October 2015
Phuket, Thailand
Website: http://wmebs.eacbee.org/
Contact person: Alissa Matthew
Organized by: EACBEE

Amsterdam International Water Week
2nd to 6th November 2015
Amsterdam, Netherlands
Website: http://www.internationalwaterweek.com
Contact person: Christina Boomsma
Organized by: IWA, Amsterdam RAI, IWC, NWP

Securing Sustainable Water for All: Innovation, Integration and Inclusion
17th and 18th November 2015
Karachi, Sindh, Pakistan
Website: http://www.hisaar.org/4_conferences/2015_conference.html
Contact person: Dr. Sono Khangharani
Organized by: Hisaar Foundation

Water 2015
17th and 18th November 2015
London, United Kingdom
Website: http://goo.gl/JyNdlB
Contact person: Becky Nye
Organized by: Marketforce Business Media Ltd

Drinking Water 2015: Developments in Water Quality, Treatment & Distribution
18th November 2015
London, United Kingdom
Website: http://ciwem.org/events/events-calendar/2015/nov/18/drinking-water-2015-developments-in-water-quality,-treatment--distribution.aspx
Contact person: Sophie
Organized by: CIWEM

IX World Aqua Congress
26th and 27th November 2015
New Delhi, India
Website: http://www.worldaquacongress.org
Contact person: Praggya Sharmaa
Organized by: Aqua Foundation

4th International Seminar on Enviromental Issues in Mining
2nd to 4th December 2015
Lima, Peru
Website: http://www.gecamin.com/enviromine
Contact person: Rebekah Zale
Organized by: Gecamin

3rd International Conference on Environment Pollution and Prevention (ICEPP 2015)
5th and 6th December 2015
Dubai, United Arab Emirates
Website: http://www.icepp.org/
Contact person: Ms. Eve Li
Organized by: CBEES

2nd International Conference on Environmental Systems Research (ICESR 2015)
25th and 26th December 2015
Phuket, Thailand
Website: http://www.icesr.org/
Contact person: Ms. Eve Li
Organized by: CBEES