

In-Situ Iron Removal from Aquifer by Recharging Oxidized Groundwater

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Abstract: Malaysia is gifted with abundant groundwater resources. However, most of it has iron (Fe) content as high as 10 mg/L compelling the water supply companies to treat it using separate treatment facilities before delivering it to the consumers. Problems of filter clogging and construction of facilities to manage considerable amount of Fe sludge are the operational and financial constraints. The proposed technology for in-situ treatment of groundwater attempts to eliminate the problem at its origin. This involves pumping up the groundwater, aerating it with atmospheric oxygen by simple showering mechanism and then recharging it back into the aquifer to create an oxidation zone, where Fe, Mn and other pollutants get oxidized, sometimes by metal oxidizing bacteria. Eventually these get adsorbed in soil particles. Simple metal precipitation in soil is avoided by maintaining specific redox values in the oxidation zone. The water harvested through this oxidation zone is of superior quality and has significantly low amount of iron and other heavy metal contaminants. This technology uses no chemical and is completely sustainable. A pilot plant of 10,000 liter per day capacity has been installed in Kota Bahru, Malaysia where the Fe content in groundwater has been drastically brought down from 9.8 mg/L to 0.5 mg/L within 6 months of its operation.

Key words: Groundwater • In-situ treatment • Iron contamination and Malaysia

INTRODUCTION

The occurrence of dissolved metals in certain parts of Malaysia has restricted the use of groundwater for potable and irrigation purposes. A report published indicates that iron (Fe) concentration ranged from 88 to 95% of the dissolved metals, the remainder being lead, arsenic and manganese. The arsenic and manganese concentration ranged from 1 to 2% in most cases [1]. Among the different states in Peninsular Malaysia, Kelantan and Terengganu are worst affected where heavy metal concentration in groundwater in 2003 was 3.14 mg/L and 7.80 mg/L of, respectively. From aesthetic point of view, this water cannot be used as it would stain the surfaces at the users' end and render an unpleasant colour and odour in the water. Although most water treatment plants follow traditional iron removal steps to meet the drinking water standard, they produce large amount of Fe sludge and uses chemicals. So, a cheaper

treatment method is required to encourage more widespread use of groundwater resources in those two states.

This proposal stems from a recently concluded project on 'In-situ treatment of groundwater contaminated with arsenic' in India funded by the European Commission under the Asia Pro Eco Call (www.insituarsenic.org). Installations of a number of iron and arsenic treatment plants near Kolkata (Calcutta) based on in-situ remediation technology have been recorded, where the high level of arsenic is recorded in groundwater. The conventional technologies used for arsenic and iron removal include oxidation, chemical co-precipitation, filtration fixed bed adsorbents (arsenic removal plants), domestic and community units. However, all of these technologies encounter several operational problems in the long run. Medium to high installation cost, safe disposal of sludge, skilled manpower for regeneration, backwash and handling of

chemicals, less eco-friendly and user-friendly are only few of them [2]. Therefore, a chemical and waste free arsenic removal method can provide a long-term water security to the affected people and significantly improve the quality of life of the poor in affected areas.

In contrast to the conventional methods, in-situ treatment plants neither use any chemicals nor produce any waste stream. The installation is similar to a tube well and all parts are locally available and can be installed by local plumbers. The in-situ technology is based on abstracting the water by a submersible pump, showering it in a tank and returning a part of the water into the aquifer under gravity. The process increases the oxygen level in the subterranean water, alleviating the anoxic condition and converting the dissolved bivalent Fe to trivalent precipitate in the aquifer sand. In a complex chemical and biological process, soluble As (III) is converted to sparingly soluble As(V) and is absorbed on ferric hydroxide precipitate. The process requires periodic recharge of aerated water into the aquifer to avoid any anoxic condition in the zone of influence and periodic monitoring of the chemical and microbial quality of the water [3].

The purpose of this particular project in Malaysia is to set up a trial in-situ treatment plant for the removal of Fe in larger scale than in India. A multi-disciplinary team of geophysicists, geologists, social scientists, microbiologists and engineers conducted on-site experiments to determine the extent of oxygen transport and heavy metal mobility. They installed the plant and

designed an operation schedule for in-situ treatment in combination with microbial intervention to establish and maintain Fe concentration below the desired limits. Based on published literature, the work focused on monitoring the Fe, nitrate (NO₃), ammonium, pH, redox potential and DO in the aquifer with and without in-situ treatment. The plant operation was started in the end of August 2011 and the data of December 2011 is produced in this paper.

Project Site

Initial Groundwater Condition: In the state of Kelantan, Malaysia groundwater accounts for 70% of all the water used. Demand of groundwater increases every year by 2.5%. In 2010, Air Kelantan Sdn Bhd (AKSB), a water supply company of Kelantan estimated the demand to be 165 MLD [4]. The ICP-OES analysis of groundwater samples from an AKSB site at Kelantan (N06 07.621' E102 17.340') revealed that Kelantan groundwater has severe Fe contamination problem. Compared to USEPA's limit of 0.3 mg/L, Kelantan groundwater was found to have an average value of 9.8 mg/L for Fe at Kampung Telok wellfield site. The Na, Ca, Al, Mg and K concentrations were found to be within safe limits. Figure 1 shows the ICP-OES analysis of two samples collected from Kelantan. The groundwater composition varies over time and repeated sampling is required to obtain an average value of its composition. Some other parameters of the raw untreated groundwater in morning and afternoon are listed in Table 1.

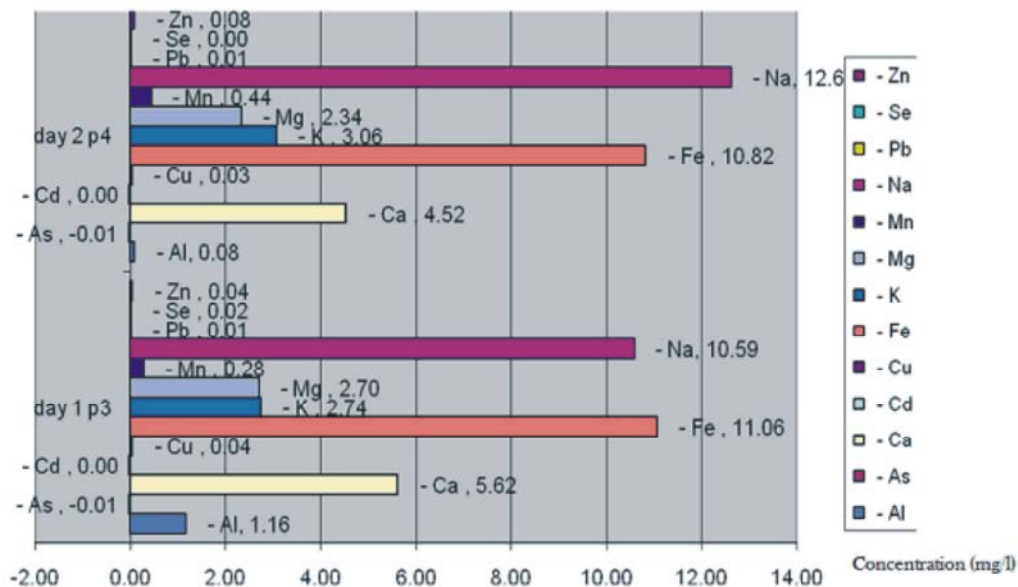


Fig. 1: ICP-OES analysis of groundwater from Kelantan, Malaysia

Table 1: Groundwater parameters in morning and afternoon before starting of treatment process

16 MAY 2011												
	Time	Turb, NTU	Tot Fe, mg/l	Fe ²⁺	Fe ³⁺	pH	Temp, °C	NO ₃	NH ₄	Ca	Mg	Mn
Morning												
Sample 1	9.50	0.65	9.76	6.30	3.40	6.05	29.4	0.8	1.23	4.8	6.35	0.303
Sample 2	10.05	0.50	9.18	5.68	3.50	6.05	28.9	1.4	1.26	4.8	6.59	0.297
Sample 3	10.20	0.91	7.65	4.32	3.33	6.06	29.1	1.4	1.25	5.6	7.32	0.307
Afternoon												
Sample 1	4.25	0.85	7.98	3.00	4.98	6.06	30.3	0.0	1.29	5.6	3.42	0.407
Sample 2	4.45	0.56	14.76	4.80	9.96	6.08	29.3	0.0	1.30	4.0	7.32	0.406
Sample 3	5.05	0.41	9.51	4.72	4.85	6.07	29.1	0.8	1.31	3.2	5.86	0.422



Fig. 2: Construction of the in-situ treatment plant.

Construction of In-situ Iron Removal Plant: The construction of the trial plant at Kampung Teluk was completed on 1st September 2011. It was built on a steel frame structure with clay brick wall for the ground room Laboratory (Fig. 2). The P and I diagram is shown in Figure 3. The 3-storey structure is 9-meter in height with floor size of 3 sq meters. The aeration tank is placed at the most upper floor with capacity of 5,500 liters (diameter of 1750 mm and height of 2060 mm), while storage tank is on the lower second floor with a capacity of 4,300 liters (diameter of 1750 mm and height of 1700 mm).

The submersible pump for abstracting raw water from tube well No. 9, with a motor capacity of 1.1kw and flow rate of 8 cubic meters per hour, is placed at a position of 6 meters below ground level in the well. The pump is controlled by variable speed drives equipment for varying

and control of the flow rate. The pumping pipeline for delivering raw water to the aeration tank is of high-density polyethylene material with diameter of 75 mm. Similarly, the delivery pipeline for transporting aerated water from the storage tank to the recharging well is made of high-density polyethylene material with diameter of 75 mm.

The operation of the system comprises a cyclic process, where the raw groundwater from tube well No. 9 is pumped to the aeration tank and is aerated through a number of shower fittings installed on top position inside the tank and the aerated water then being flowed back to the tube well No.9 (TLK 9) for recharging. The flow of the water is controlled and released back for recharging in sequential timing of twice daily routine i.e. the morning cycle and the evening cycle.

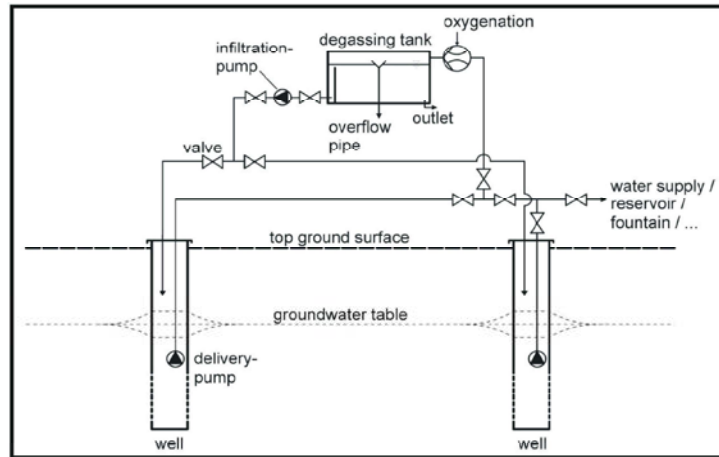


Fig. 3: P and I diagram of a two-well system.

Uncontrolled oxidation of the Fe in the aquifer can lead to Fe and Mn precipitation rather than adsorption. Also, the abrupt change in redox potential and excess oxidation may destroy the existing bacterial population, making the whole process unstable and ineffective. Thus, a mobile testing facility was made available. Other than these, the test area has a total of 16 monitoring wells and 10 recharge wells.

Well Description: The Kampung Teluk Wellfield aquifers are of shallow quaternary alluvium type and have high hydraulic conductivities. The area has intensive agriculture and the groundwater is rich in organic carbon and nitrogen. As a consequence, the reduced water condition favors a high dissolved iron level. The initial sample of raw water recorded a total Fe level of up to 14.0 mg/L and an average total Fe concentration of 9.8 mg/L. The presence of any ferric iron (Fe^{3+}) in the sample is due to fast oxidation of water during collection. It is also believed that unattended open wells (both production and observation), over a long period of time resulted in infestation of Fe reducing bacteria. It is also believed that the groundwater inflow brings in very high levels nitrogen, organics and coliform bacteria to the pumping zone. Keeping in mind that oxidation of Fe ranks after dissolved organic carbon (DOC) and nitrogen, adequate supply of dissolved oxygen is required to arrest further proliferation of Fe reducing bacteria and ensure controlled oxidation of dissolved Fe. To further complicate the problem, the area has high hydraulic conductivity, close to 20 m/day and poses a real challenge in profiling the oxidation zone created by the recharge water. The initial untreated raw water quality data are given in Table 1.

Development of Recharge System: The trial system was created to recharge a maximum of 5 cubic meters of aerated water at an average DO of 7.4 in each cycle (two cycles a day). Considering the fact that at least 0.3 mg/L of oxygen is required to keep the dissolved Fe below the drinking water limit, for design purposes, we may consider 0.5 mg/L of oxygen as the boundary of the oxidation zone. Dissolved iron (Fe^{2+}) oxidizes to the insoluble form (Fe^{3+}) above the redox value of +50 mV. However, at least +150 or higher redox value of intake water is necessary for a sustained Fe removal process over a long period of time. This implies that continuous recharge of aerated water must ensure that redox potential (ORP) increases from -40 mV (typical) to about +150 mV or more within the pumping zone. The value of raw water ORP is the best indicator for this purpose. Therefore, a redox based programmable logic (PLC) controller will stop pumping, once ORP is less than +150 for more than 5 minutes. For a large-scale installation, multiple recharge locations would ensure sustained pumping for several hours at a time. In a controlled experiment, when the same recharge water volume was distributed over a larger area (multiple water recharge), there was obvious spurt in the ammonia level, which decreased, in the following weeks. This is due to the fact that the same volume of recharge water was being spread over a larger area. Ammonia level stabilized to a value lower than 1 mg/L. It may be noted that precipitating Fe^{3+} binds phosphorous (P) with it. The in-situ system, under appropriate operating condition (such as +150 ORP) will ensure excellent P removal from groundwater.

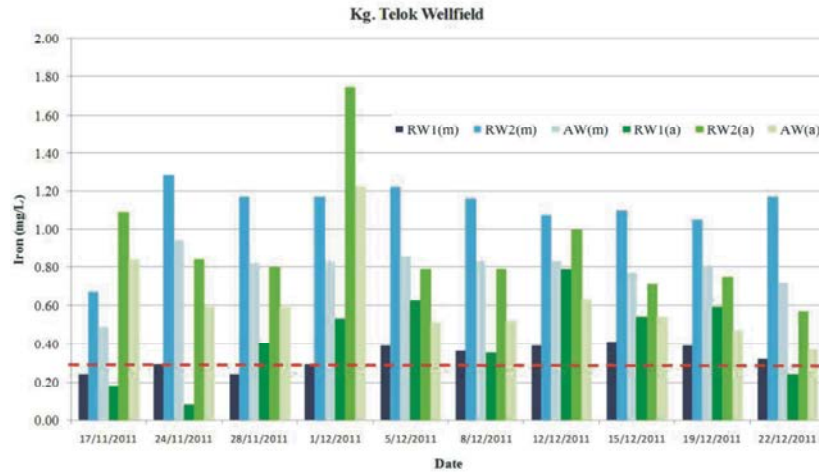


Fig. 4: Variation of Fe concentration over 1 month time after 3 months of plant operation [abbreviations: (m) morning, (a) afternoon, RW- raw water, AW- aerated water, Dash line indicate MOH drinking water quality standard].

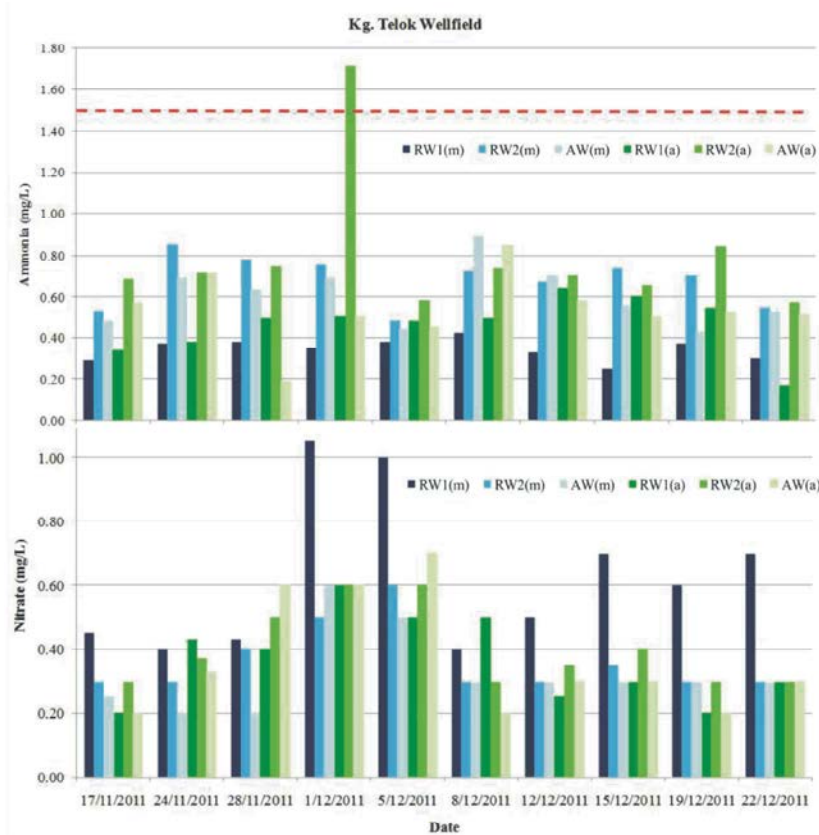


Fig. 5: Change of ammonia and nitrate concentration after 3 months of operation [abbreviations: (m) morning, (a) afternoon, RW- raw water, AW- aerated water, Dash line indicate MOH drinking water quality standard].

Water Quality after Starting the System:

There is a strong relationship between redox value (mV) and pH of water. Redox value decreased by 58 for every unit of pH rise. Therefore, it was easier to increase the redox potential of slightly acidic water than its alkaline counterpart. The pH and temperature

range of raw water in the pumping zone were, 6.5 to 7.3 and 27° to 29 °C respectively. Despite of this variability and its effect on ORP, +150 mV of ORP of the intake water at a pH just above 7 could be optimum for maximum production of water as well as target Fe level below 0.3 mg/L.

Within three months of plant operation, i.e. initialization of aerated water recharge in well 9, the groundwater was sampled from monitoring wells 1 and 2 downstream of recharge well 9 (TLK9) in the morning and afternoon. Also, the water pumped up and aerated in the tank was analyzed. All the data are presented in the Fig. 4. The Fe value is seen to come down below 1.2 mg/L. The aerated water has much less Fe content of only 0.4 mg/L. The Fe content in the morning is found to be higher than in the afternoon. Raw water withdrawn from monitoring well No. 2 (RW2) is found to contain more Fe since it is far away from the TLK9. However, monitoring well 1 is near TLK9 and registers much lower Fe concentration around 0.34 mg/L. Although the Fe concentration is still above Ministry of Health (MOH) guideline of 0.3 mg/L, it is expected to come down with longer period of operation. Also, conventional pump and treat systems will be able to remove the remaining amount of iron without any pressure in the filtration system.

The groundwater samples from monitoring wells 1 and 2 in the morning and evening have also been tested for NH_3 and NO_3 . Any increase in NH_3 would indicate increasing level of reduced condition. In-situ oxidation successfully reduced NH_3 by oxidizing it to NO_3 as evident from Fig. 5. The data indicate that the NH_3 level dropped to 0.4 mg/L from an initial value of 1.4 mg/L. The nitrate level also dropped to 0.4 mg/L from approx. 1.4 mg/L. We need to bear in mind that NH_3 is oxidized to NO_3 and by that reasoning, a 70% drop in nitrate level is quite satisfactory.

In a controlled experiment, when the same recharge water volume was distributed over a larger area (multiple water recharge), there was obvious spurt in the ammonia level and the value stabilizing at a higher level. This is due to the fact that the same volume of recharge water was being spread over a larger area. It stabilized to a value lower than 1 mg/L. It may be noted that precipitating Fe^{3+} binds phosphorous (P) with it. The in-situ system, under appropriate operating condition (such as +150 ORP) will ensure excellent P removal from groundwater. From consumer point of view, high ORP water may not necessarily have any beneficial effect or enhanced organoleptic properties. Tap water has a typical ORP of +300 and aerated water may have no unpleasant odour. In comparison, untreated (reduced) groundwater may have benefits of removing oxidizing radicals from the body and is perfectly suited for human consumption. The health benefits of sulphur rich water are well established.

In-situ oxidation is expected to reduce the coliform and fecal coliform count. A high initial total coliform count of 200 CFU/ml, dropped to 0 on 10 Oct and 11 Nov 2011 in two consecutive readings when the production well was operating under appropriate oxidizing condition. In-situ method generally reduces the coliform bacteria by removing essential nutrients from water (P for example). However, disinfection of water prior to distribution is required as a part of regulatory requirement and also to inactivate the residual coliform bacteria. During trials, the research team has observed sudden spurt in coliform count following heavy rainfall episodes in the area. Therefore, it is strongly recommended that conventional disinfection stage must be incorporated in the commercial water treatment plant using in-situ technology. Since aerated water is not secured, it shows a rise in coliform value; this is easily preventable and at the current time no measures have been taken to prevent it. TC count in raw water is more important and the redox value will have a direct bearing on the count. For inactivation of coliform by high ORP alone, the value must exceed +650 mV, when the bacterial cell becomes an electron donor to oxygen. In in-situ method, total coliform count goes down as essential nutrients such as DOC, N and P are depleted in the raw water.

Scope and Replication of Sar in Different Parts of the World: The SAR technology can be applied in removing Fe and As from groundwater in Ganges, Brahmaputra, Mekong delta and the places where As is of arsenopyrite origin. The Fe and As affected aquifers of India, Bangladesh, Cambodia, Nepal, Vietnam, Malaysia, Thailand and USA can be remediated by SAR. Currently, this technology is being replicated for Fe removal in Kolkata, India and Kelantan, Malaysia as well as for Fe and As removal in Cambodia, Vietnam and the USA with the assistance of some donor agencies and Universities. At present, University of Malaya, Malaysia, Queen's University Belfast, UK and National Metallurgical Laboratory-Jamshedpur, India is doing detailed research for further development of this technology.

CONCLUSION

This in-situ treatment technology has brought down Fe concentration in Kota Bahru test site from 9.78 mg/L to 0.4 mg/L within 3 months of operation. The NH_3 and NO_3 reduction were also achieved. Redox potential was increased from about -40 mV to approximately +150 mV, indicating oxic condition of the aquifer. Other nutrients

such as nitrogen, DOC and phosphate gets oxidized or trapped with the Fe³⁺ resulting in reduction of coliform count. However, the amount of water to be recharged in the aquifer and also the rate of recharge should be carefully calculated to make the process effective and sustainable. The greatest advantage of this process is that no metal sludge is produced and no chemicals are required. Interested readers may visit www.insituarsenic.org to gather more information on this technology.

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REFERENCES

1. Hussin, N., I. Yusoff, Y. Alias, S. Mohamad and N.Y. Rahim, 2010. Preliminary Study on the Application of Ionic Liquid as a Solvent Medium for Iron Removal in Groundwater, 1st National Conference on Natural Resources (NCNR2010): Conservation on Natural Resources and Sustainable Development, Kota Bharu, Malaysia.
2. Bhattacharyya, R., D. Chatterjee, B. Nath, J. Jana, G. Jacks and M. Vahter, 2003. High arsenic groundwater: Mobilization, metabolism and mitigation - an overview in the Bengal Delta Plain. *Mol. Cell. Biochem*, 253: 347-355.
3. Sen Gupta, B., S. Chatterjee, U. Rott, H. Kauffman, A. Bandopadhyay, W. De Groot, N.K. Nag, A.A. Carbonell-Barrachina and S. Mukherjee, 2009. A simple chemical free arsenic removal method for community water supply - A case study from West Bengal, India. *Environmental Pollution*, 157: 3351-3353.
4. Adnan, N.S.M., I. Yusoff, Y. Alias, B. Sen Gupta, M.A. Hashim and A.K.S. Bhd, 2010. Hydrogeological Investigation: Desk Study of Kampung Telok, Panji, Kota Bharu Kelantan, Dept of Geology, University Malaya, Kuala Lumpur, 50603, Malaysia.