Hydrol. Earth Syst. Sci. Discuss., 10, C7519–C7523, 2014
www.hydrol-earth-syst-sci-discuss.net/10/C7519/2014/

Hydrology and
Earth System
Sciences
Discussions

Interactive comment on "Identifying sources and controlling factors of arsenic release in saline groundwater aquifers" *by* C.-W. Liu et al.

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Received and published: 29 January 2014

Re: Ms. Ref. No.: hess-2013-337

Responses to the comments from reviewers.

Please note. Reviewer's comments are italicized. Author's responses follow immediately below the specific comments.

We all authors are very much thankful to the reviewers for making the comments on our communicated manuscript "Identifying sources and controlling factors of arsenic release in saline groundwater aquifers" for the possible publication in the HESSD. After going through the comments made, we have tried our best to give the answer as

follows.

REFEREE 1

General comments Organize the context in a more comprehensive way especially the result section and the writing/ grammar can be improved. The result section is reorganized to three subsections in a more comprehensive way. Moreover, the writing/ grammar of the manuscript has been improved by a native English speak editor.

1. Page 10575: Line 15: Figure 3 does not look very useful. Instead of relating arsenic concentration with depth and EC, I think it would be better to link with lithology and different ion composition (Like Figure 4) Figure 3 illustrates that the saline water mostly distributed in the uppermost aquifer. But high arsenic concentrations were found in both uppermost and lower groundwater aquifers (Fig. 3b). It suggests that the arsenic release to groundwater may be involved in complex ionic reaction in the groundwater aquifers. Fig. 4 only shows the lithology of the uppermost aquifer. We thus retain the Fig. 3.

2. In Figure 4: the lithological data can be presented more prominently, as it is very useful piece of information. Lithological data presentation in Fig. 4 is revised.

3. Page 10576:Line 25: ": : :.and interaction seawater and non-saline groundwater in this region (Figure 5)": : :from the discussion and from the figure 5 the above statement cannot be derived. The sentence ", indicating the complex. . .(Fig. 5)." is deleted.

4. Page 10577: Line 25-30: High As(V) in YL6-1 and As(III) in YL6-1 was observed. Here the difference in EC was mentioned, however I feel mentioning difference in pH, redox condition would be more relevant. The average pH of groundwater in YL6 and YL7 were 8.1 and 8.0 respectively. But we did not measured the redox potential (Eh) of both well waters. According to the data of arsenic species concentration, we supposed that the YL6-1 is under reducing condition and YL7-1 is under oxidizing condition.

5. Section 3.5: If Figure 9 represents only the calculated/simulated values, then line

plot is more suitable and it can be combined with Figure 8. Fig. 9 represents the arsenic desorption of YL6-1 and YL-7 at various anionic concentration. If we overlay these data on Fig. 8, it will be difficult to distinguish the behavior of arsenic desorption at different anionic concentration. We thus retain the experimental and simulated data in two separated Figs.

REFEREE 2

1. The reviewer has made many hand-written comments in the attached file. We appreciate the hand-written comments by the reviewer and made all the necessary corrections in red color.

2. The reviewer suggested that the desorption experiments in the presence of different co-existing ions (in the batch mode) are erroneous. We are very sorry for that and strongly oppose against the made comments. The all experiments were conducted to see the leaching effect of As in the presence of different co-existing ions that are common in the water environment, in the effluents and are important and useful. The EDX study conveys that the core sample has the different minerals (Table 2) as the composition especially Fe, Al. It is well known that As has greater affinity toward Fe and Al. Hence, in core samples As is tightly bonded with these minerals. Sulfate ion can from an outer-sphere complex on the surfaces of the adsorbent with the said components [1, 2]. The presence of PO43- strongly affected the As leaching as it could bind with the components of adsorbent (Al, Fe) [3]. Bicarbonate also formed inner-sphere complex with the adsorbent components (Al, Fe) [4]. Hence, due to the combine effects of those said ions with As-contained core samples in the medium inhibit the leaching of As from the core samples into the effluent and is observed not too much and is evidenced from the Table 4. So, the concept of the desorption study in the presence of those ions is appropriate in the present context.

3. The reviewer has rightly pointed out the conversion of HCO3- according to the following equation:

We appreciate the made comments and it is true. In the present study our main motto was to know the leaching of As from the core sample in the presence of bicarbonate ions and we did. The obtained result has been provided in the Table 4. Hence, the crucial experiment, the rate of conversion of bicarbonate ion into the product we did not calculated assuming the low or minute conversion rate. Once again, if the decomposition rate of the inner-sphere complex (formed by the bicarbonate ion) is more, then the As leaching would be more. In the present study, from the Table 4, a regulated As leaching was observed. Hence, we can say that the bicarbonate conversion rate was very low. However, regretfully we say that we did not measure the conversion rate.

4. The experiments should have been performed under natural redox conditions. We very much appreciate the comment regarding the redox environment in anaerobic conditions. Yes, it is true that the redox conditions of the environment will be predicted in a semi-anaerobic condition. However, the major causes of shallow groundwater salinization were the vertical infiltration of salt water from fish ponds farming and the seawater over flooding in the subsidence coastal area. Seawater intrusion plays a minor role on the local groundwater salinization due to aquifers are enclosed by the aquidard which does not directly contact the seawater. The oxygenic seawater mixed with shallow groundwater may create an oxidative environment. We thus had conducted the all experiments in the aerobic conditions.

5. There are no reduced forms of Fe oxyhydroxides. To the dest oxides exist in nature that contain both Fe(II) and Fe(III), but Fe(II)-oxides are rather irrelevant in environmental geochemistry. I wonder whether siderite, Fe-sulphides or glauconite and other Fe-bearing Al-silicates become measured with the technique in addition to pure Fe-oxyhyroxides. All these minerals may be present in a coastal groundwater environment. We are very much thankful to the reviewer for making comments and giving the suggestions. The major types of Fe minerals characterized by HR-XPS were FeO, Fe2O3 and FeOOH. Siderite, Fe-sulphides or glauconite and other Fe-bearing Al-silicates may be present in a coastal groundwater environment. The HR-XPS analysis is not able to

recover the signals of the existence of these minerals in the study.

References [1] J. F. Hingston, A. M. Posner, J. P. Quirk, Anion adsorption by goethite and gibbsite:1. The role of the proton in determining adsorption envelopes, J. Soil Sci., 23 (1972) 177-192. [2] C. Su, R .W . Puls, Arsenate and arsenite removal by zerovalent iron: Effects of phosphate, silicate, carbonate, borate, sulfate, chromate, molybdate, and nitrate, relative to chloride, Environ. Sci. Technol. 35 (2001) 4562-4568. [3] S. K. Maji, A. Pal, T. Pal, Arsenic removal from aqueous solution by adsorption on laterite soil, and references there in, J. Environ. Sci. Health-A 42 (2007) 989-996. [4] C. C. Fullar, J. A. Davis, G. A. Waychunas, kinetics of arsenate adsorption and co-precipitation, Geochimica et Cosmochimica Acta 57 (1993) 2271-2282.

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., 10, 10565, 2013.