**Reply to Ian. Cartwright (Referee #2):**

We are very grateful for the constructive comments and generous help from Prof. Ian Cartwright. We are trying to give our careful response to your comments for improving our paper quality.

Our responses to the points raised by the reviewer are listed below:

(1) a) A rethink about what material is strictly necessary in Section 5 and/or better guidance to how the information addresses the main points of the paper. This is probably the major concern.

**Reply:** We agree that better structuring of sections 4 and 5 would lead to a better manuscript. This point could be addressed by means of a careful review of section 5, removing extraneous information and discussion, and focusing on the topic of the paper- determining the natural as distinct from anthropogenic inputs to the dissolved ion load (particularly sulfate) in groundwater.

(2) b) More consideration as to how this study can inform others elsewhere in the world.

**Reply:** We agree. In a revised manuscript, we would make a greater effort to situate the study within a global context, such that others are in a position to learn from the findings. This would involve a more complete literature review of the state of knowledge regarding the techniques adopted, and a review of similar case studies worldwide.

(3) c) Some reorganisation of the introduction and results section.

**Reply:** We agree. These sections could be re-organised with a view to making the paper more concise and focused on the main topic, and more logical in structure such that the reader clearly understands which data type is useful in understanding particular aspects of the evolution of water quality.

**INTRODUCTION:**

(4) The introduction provides good background to the study and places it in an international context. Some of the referencing appears to be getting a little dated (mostly before 2012), and I'd suggest that the authors consider whether there are any important more recent papers that they can cite.

**Reply:** We agree. We have since identified newer references relevant to the topic – such as de Louw et al., 2013 (Journal of Hydrology, 501: 133-145) and Kumar et al., 2015 (Asian Journal of Earth Sciences 111: 936-947) and would insert these into a revised version of the manuscript.

(5) The first paragraph on pg. 11334 could use a few more details. Specifically, it is not clear what is meant by "more serious" and "a range of strategies" etc. Without a detailed knowledge of the area it is difficult to assess exactly the extent of the problem or what has been done to address it. If you provide a few more details, the context will be clearer.

**Reply:** We agree. These points would be addressed in any revised version of the manuscript. For example, compared with the degree of seawater intrusion in 1982, the lower fraction (less than 4.8%) of seawater reveals that the areal extent of seawater intrusion has been controlled and now has a reduced contribution to modifying the groundwater salinity (which we would clarify). This is due to ceasing the water supply from the well field to the Dalian City in 2001. The language would be improved so that ambiguities are removed.

(6) There is also some repetition of ideas in this section. The statements regarding the need to distinguish seawater intrusion from anthropogenic activity and the use of tracers appear both in the last paragraph on pg.11333 and the final paragraph in this section. I think the flow of the paper would be improved if the paragraph on seawater intrusion in the Dalian area (top pg. 11334) was merged into Section 2 (as it is really a detail about the study area) and have the introduction focus on the broader issues; the statement of aims at the end of the introduction provided sufficient information about the specifics of the study.

**Reply:** We agree. In a revised version of the paper, we would address this comment and adopt the suggestions of the reviewer in order to make the paper more concise and avoid repetition.

**Study Area:**

(7) The statement regarding the natural flow in the area (top of pg.11335) should be referenced.

**Reply:** We agree. The reference relevant to the statement is: Fan, 1984, Seawater intrusion and calculation of groundwater exploitation in the karst area of the west JinXian, Dalian. Hydrogeology & Engineering Geology (in Chinese with English abstract). 1:3-6.

(8) Pg. 11335 line 9. Define m.a.s.l on first usage.

**Reply:** We agree. This point is straightforward to address (meters above sea level).

(9) The description of the geological framework on pg. 11335 would be much easier to follow with a cross-section. I'd suggest adding a stratigraphic cross-section to Fig. 1 or if that information is on Fig. 9 to move that figure to earlier in the paper and use it to also illustrate the geology.

**Reply:** We agree. In a revised version of the paper, we would include a cross section (such as that shown in Fig 9) to the section showing the geological framework (section 2; Study area).

(10) The last paragraph (pg. 11336) just compares averages, which may or may not be informative. For example, if one area increased in salinity by say 10 fold but the rest of the areas remained similar, the average salinity has increased but the pattern is skewed by a small subset of data. Try to put more details around this as it is the main rationale for doing this study, so consider the ranges as well as the averages. If the data comes from the same suite of bores then you can apply t-tests or Z scores (or something similar) to assess this; at the very least report the ranges.

**Reply:**

We agree. Further detail on the precise changes occurring in these different areas (e.g. comparison between the recent data and previous data) would be included by making the text more specific. In some cases the number of samples required to conduct a valid t-test or Z score is too low, however we would attempt this where possible.

**Methods:**

(11) Section 3.1. Most of the methods are appropriately described, a few additional

details on the C-13 analysis (specifically the preparation device, I assume automated headspace analysis using continuous flow) and the S-34 analysis (probably combustion in an EA) would be good.

**Reply:** We agree. This point would be addressed in any revised version of the manuscript. Additional details on the stable isotope analysis could be easily added to the text – for example noting the following points:

1. - $\delta^{13}C$ values of dissolved inorganic carbon (DIC) were measured using continuous flow on a Finnigan MAT 252 mass spectrometer, with the automated headspace analysis of the preparation device, in the State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry (Guiyang), CAS. The results of $\delta^{13}C$ analysis are expressed in conventional delta ($\delta$) notation, defined as $\delta = (R_{sample} - R_{standard})/R_{standard} \times 1000$, where R is the ratio of $^{13}C/^{12}C$. The $\delta^{13}C$ values of dissolved inorganic carbon (DIC) are expressed relative to the standard Vienna Peedee Belemnite (VPDB), with an analytical precision of ±0.2‰.

2. - Samples for $^{34}S$ in dissolved sulfate were measured by a Finnigan Delta-S gas mass spectrometer after on-line pyrolysis with an EA (Elemental Analyzer) in the Laboratory for Stable Isotope Geochemistry, Institure of Geology and Geographysics, CAS. The method of Halas and Szaran (1999) was used for converting precipitated $BaSO_4$ to $SO_2$. The international standard against which $\delta^{34}S$ values are referenced is the troilite (FeS) phase of the Cañon Diablo meteorite (CDT), which has a $^{34}S/^{32}S$ abundance ratio of 0.0450 and are reported as $\delta$ (‰) difference from the standard with an analytical precision of better than or about ±0.4‰.

(12) Section 3.2. The assumption of Cl being conservative (pg. 11137, line 25) should be better justified. In addition to halite from the aquifer matrix, there is the possibility that contaminant sources introduce Cl. Did you analyse Br, in which case the Cl/Br ratios can be used. Otherwise, you need to be more definitive in ruling out addition of Cl from other sources.

**Reply:** We agree and could address this issue substantively using data found from literature in the area. Although we could not measure the Br concentration in groundwater, we were able to find Br concentrations in a study which used the same suite of monitoring bores in a previous study (Yang, 2011); Yang (2011) carried out the groundwater investigation in this area in August 2008. The molar Cl/Br ratios in their study ranged from 118.3 to 633.1 (n=11, mean value 394.3, Fig. R2), which is generally below the oceanic ratio of ~650 (Drever, 1997), hence, the groundwater show depletion compared to seawater (Figure R2, shown below).

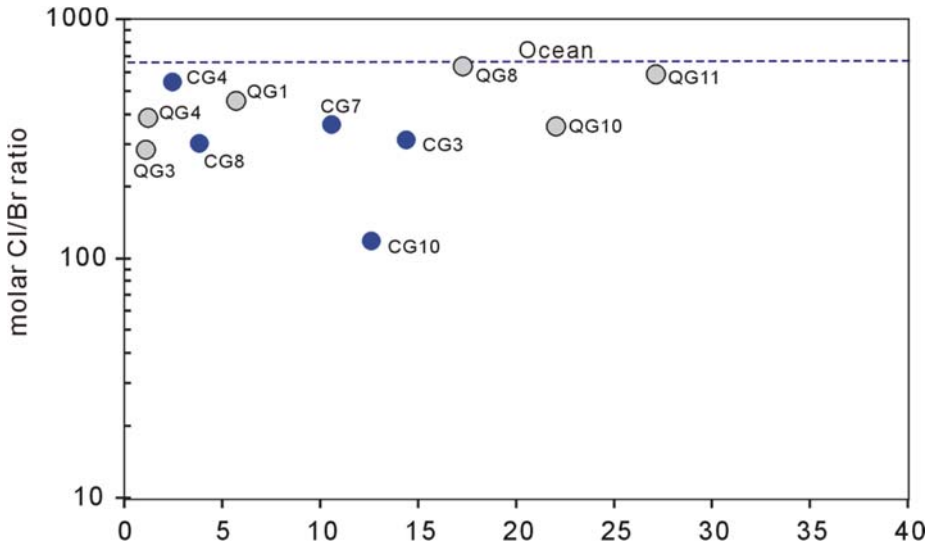

Fig. R2. Molar Cl/Br ratios vs. Cl concentrations in groundwater from the Quaternary aquifer (grey) and carbonate aquifer (dark). Data are from Yang (2011).

A range of natural and anthropogenic processes may modify Cl/Br ratios, although processes causing a decrease (e.g. enrichment in bromide) are relatively poorly known. We suspect either possible anthropogenic source (e.g., pesticides ethyl dibromine and methyl bromide), which may reduce Cl/Br ratios (Davis et al., 1998) and/or preferential adsorption of Br (e.g. on organic material) (Gerritse and George, 1988). Overall the ratios do not indicate that a major modification to mass balances of the ion load would be needed, however the issue would be noted and discussed in any revised manuscript in accordance with this observation.

**Results:**

(13) The results section is comprehensive and the data is well described. There is a tendency, however to mix observations and interpretations, for example:

Pg. 11339, line 6 – calculated seawater fractions

Pg. 11139, line 13 – inference of anthropogenic inputs

Pg. 11140, line 12 – interpretation of the C-13 data

This interpretations would be better in Section 5 where you interpret the data.

**Reply:** Thanks for these suggestions.

We agree and would address this issue in a revised version of the manuscript, making sure results and discussion are not mixed in section 5. This would be achieved by removing the discussion points and re-locating them into section 5, keeping the results free of commentary.

**Discussion:**

(14) Section 5.1. The start of this section is where the discussion of seawater mixing from Section 4 belongs.

**Reply:** We agree and would follow the reviewer's advice

(15) Page 11341 last paragraph. I struggled to understand the argument here. Surely this approach is just dealing with the details of the chemistry and whether or not major salinization is occurring should be apparent from TDS or Cl changes. I'm not sure that this adds much and it could be replaced with a statement just reiterating the changes to TDS over time (which is in Section 2).

**Reply:** We agree. We would remove the paragraph in a revised version and replace with a brief

statement regarding overall TDS changes (as distinct from other ions like sulfate) over time.

(16) Overall, Section 5 is too long and tries to describe too many things. The thrust of this paper is to distinguish between anthropogenic inputs and seawater intrusion and you should try to keep this as the main focus of the paper. You do get to this at the end of Section 5 but there is a lot of other material in this section that looks to be in there in order to provide a full geochemical interpretation. While that is OK, it does detract from the main message. I'm not convinced that the carbon story is central to answering your main questions and that section probably could be omitted or shortened substantially. If you can keep this section focused, you can make a more convincing case as to the importance of anthropogenic inputs.

It may be that you need to discuss processes such as ion exchange or mineral dissolution in order to fully understand mixing, in which case you need to guide the reader through the process better. For example, Section 5.2 discusses the interaction with carbonate minerals and while it represents a comprehensive analysis, it is not immediately clear how understanding this helps us with the question of mixing vs. anthropogenic inputs.

**Reply:** We fully agree. In order to avoid providing a full geochemical interpretation of all of the data, we would make section 5 shorter so that the discussion focusses on the main topic, as correctly identified. While some of the discussion of other aspects of the geochemistry is necessary in order to make interpretations as to the sources of dissolved ion load (natural vs anthropogenic) some sections could be cut and/or reduced in length.

(17) Try to assess critically how each piece of information informs your key hypotheses and then omit or shorten sections that might be just interesting but peripheral but explain more fully how the others relate to the key issues. The main question that you are answering seems to be that while the salinity is decreasing following a cessation of pumping, the sulfate and nitrate are behaving differently and it is important to know whether that is due to contamination – so try to keep everything focussed on that.

**Reply:** As per the response to the previous comment, we agree with this statement and would seek to make the revised version more concise and focused on this topic. As correctly noted, after the cessation of groundwater pumping in the Daweijia well field, the TDS concentration showed increases, yet the $Cl^-$ concentration did not significantly elevate. At the same time, $SO_4^{2-}$ and $NO_3^-$ concentrations increased several times compared to parameters measured in 1981. This would be made clear in the introduction and in the discussion sections so that the main points of the paper are clearer.

**Conclusions:**

(18) This section just summarises the main findings of the project. In this section explain in more detail how your project helps us to understand processes in these environments more broadly; the paper will have more impact if researchers from elsewhere in the world can see relevance to their studies and a paper in a major international journal such as HESS needs to have broad appeal.

**Reply:** We agree. Further details about the broader implications would be added to a revised version of the manuscript, with particular reference to the way our study helps to understand hydrochemical evolution in areas recovering from saltwater intrusion, and where surface inputs from agriculture

begin to manifest as the dominant control on water quality.

Figures

(19) Fig. 1. Make sure that the localities that you discuss in the text are on this Figure (or the inset) and add latitude and longitudes as you also use these in the text.

**Reply:** We agree. Localities can be updated and shown on all figures.

(20) Figures 2-4 & 6-8. I really struggled with the difference in colours (light blue vs. grey), either make these more contrasting (e.g. dark colour vs grey) or use different symbols

**Reply:** We agree, and would use a stronger contrasting colour scheme in any revised version of the figures.

If you have any further questions regarding our manuscript, please contact us as soon as possible.

Thank you very much for your kind consideration.

Sincerely Yours,

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