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SUBJECT: Evaluation of report on the geochemistry, chronology, and sedimentation in Great Salt Lake.

Below is a list of questions that UDWQ would like Dr. Thure Cerling to respond to based on an independent review of the dating methodologies and interpretation.

1. Are the field and laboratory methods described in Levitt et al. 2012 appropriate for the purpose of dating sediment cores in Great Salt Lake back to the pre-industrial age?
2. Based on the spreadsheets provided, are the dates and 'errors' correct for all the cores used in the final analysis for purposes of establishing chronology (Core 1, Site 3 [Mid-Gilbert Bay]; Core 1, Site 4 [South Gilbert Bay]; and Core 3, site 1 [Farmington Bay])?
3. The ^{137}Cs peak dates appear to differ for the three cores: 1936 for Site 4, 1928 for Site 3, and 1962 for Farmington Bay. Is there a scientific rationale for these discrepancies?
4. In all three cores, Pb-210 activity appears to drop to a base level around 3.5 or 4.0 (dpm/g). However, this leveling out appears at different years (1932 for Site 1, 1900 for Site 3, and 1920 for Site 4). Is there a scientific explanation for these discrepancies?
5. How certain are the dates for the Farmington Bay Core (Site 1) especially for the period between 1930 and 1963?
6. UDWQ is interested in distinguishing metals concentrations and ecological indicators (macroinvertebrates and algae) for discrete periods in the past especially the mid-1970s, the early 1950s when the first causeway was constructed between Gilbert and Farmington Bay, and the mid-1850s prior to sewage discharge to Great Salt Lake. Are the dates reported in the Levitt et al. 2012 reliable for distinguishing these three periods? If not, how much differentiation can be made (e.g. if decadal dating is uncertain can the cores provide information at multi-decadal scale?).
7. Are any of the cores more or less certain in terms dating and chronology?
8. Does the reviewer have any other comments on the scientific quality of the study?

Response by Thure E. Cerling

This is a response to the questions supplied by UDWQ. I evaluated the separate reports and data of Leavitt et al. (2012), Moser et al. (2012), and Wurtsbaugh (2014).

1. *Are the field and laboratory methods described in Levitt et al. 2012 appropriate for the purpose of dating sediment cores in Great Salt Lake back to the pre-industrial age?*

The chronology of the past several hundred years can be studied by a variety of methods – each has its strong points and each has its own weaknesses. For determining sediment histories in lakes and reservoirs, the radiometric methods of ^{210}Pb and ^{137}Cs depth profiles have been favored methods for several decades, and this method is also complementary to metal concentration profiles.

The ^{210}Pb method is based on the atmospheric deposition of ^{210}Pb derived from atmospheric ^{222}Rn , and the subsequent decay of ^{210}Pb with a half-life of *ca.* 22 years. ^{210}Pb measurements need to be corrected because of "supported ^{210}Pb ", which results from the co-sedimentation of ^{226}Ra . The "supported ^{210}Pb " is the concentration of ^{210}Pb resulting from the ingrowth of the decay products of ^{226}Ra . Leavitt et al. 2012 use the ^{210}Pb methodology developed for lake and reservoir sedimentation, especially those developed by Appleby and Oldfield (1978, 1983); those papers discuss the methods of constant initial concentration ("cic") and the constant rate of supply ("crs"). The former assumes constant sedimentation rates and constant ^{210}Pb input; the latter assumes a constant ^{210}Pb input but where sedimentation rates may have changed due to human activities and with dilution by sediment.

A complementary method uses ^{137}Cs as a tracer. The deposition of ^{137}Cs to the Earth's surface is primarily due to above ground-nuclear weapons testing; the peak "fallout" of ^{137}Cs to North America was in 1963. The peak concentration of ^{137}Cs in sediments is generally taken to indicate 1963 except for rare circumstances (e.g., proximity to a nuclear facility or within the immediate fallout of a nuclear accident such as Chernobyl or Fukushima).

Elemental profiles are also used in regions with known disturbance histories related to human activities, such as mining and smelting

All of these methods have in common the assumption that the analytes (^{210}Pb , ^{137}Cs , trace metal) are particle tracers because of irreversible adsorption by clay particles. In a simple system, and certain lake or marine sediments can be considered as such, there are no additional chemical or physical transport mechanisms (see Appendix). Together, these methods can be used to test whether the sediments being analyzed have had a simple

history and are suitable for dating using these methods. I note that many lake sediments are not suitable for dating using these methods due to erosion, bioturbation, sediment focusing, or other sediment disturbance; oxidation-reduction reactions also have the potential to mobilize metals. The initial report does not evaluate scenarios of sediment disturbance, erosion, bioturbation, chemical diffusion. Here "sediment focusing" is meant to indicate changes in sediment distribution over time: in Great Salt Lake which is a shallow lake, over the past 100 years the shoreline has changes by 100s of meters laterally, channels have been dredged, causeways have been built, and water depth has changed by >5 meters it is likely that most (if not all) parts of the lake have been significantly affected by sediment focusing. It is a significant challenge to find sites suitable for the interpretations needed by DWQ for regulatory purposes.

The analytical methods and the correction methods used to determine the total ^{210}Pb and ^{137}Cs concentrations, and the "unsupported" ^{210}Pb are correct and appropriate for this problem. However, even if the methods are the appropriate approach to use, there is no guarantee that the system studied meets all the criteria for a successful use of the method.

2. Based on the spreadsheets provided, are the dates and 'errors' correct for all the cores used in the final analysis for purposes of establishing chronology (Core 1, Site 3 [Mid-Gilbert Bay]; Core 1, Site 4 [South Gilbert Bay]; and Core 3, site 1 [Farmington Bay])?

This is the crux of the issue. It is important to separate the "spreadsheets" from the interpretation.

The spreadsheet computations appear to be correct with some minor issues. The formulas for determining the uncertainties in the excess ^{210}Pb are not included: for "Core 3 Site 1" the value appears to be incorrect; however, the value given for the cores "Core 1, Site 4" and for "Core 1, Site 3" do appear to be correct. Thus, there is a slight error in the determination of the uncertainties associated with the "unsupported" ^{210}Pb for core "Core 3, Site 1" ("Excess Pb-210 Activity 1s Error"); the spreadsheet did not contain the formula used to determine these values (column R). This did not, however, affect the dates computed in the analysis because this uncertainty is not part of the final analysis. The other cores are correct (Sites 3 and 4).

This error has since been noted by the authors and was a result of a spreadsheet being copied incorrectly. I note that the authors have checked the other calculations and only the uncertainties were incorrectly copied. This does not affect the calculations in the report.

The spreadsheet assumes that the "crs-model" is correct for these cores. Great Salt Lake, and especially the near shore Farmington Bay is likely to have problems that do not

meet the assumptions of the "crs-model", especially with respect to episodes of erosion, non-deposition, or sediment focusing. I note that Core 1 in Farmington Bay was collected from a water depth of only 15 cm (Wurtsbaugh, 2014, page 9); the level of the lake in 1965 was about 1 meter lower than when the core was collected in August 2009 (Figure 1) and therefore the site was exposed at the surface for some period of time. This observation compromises the Farmington Bay Core 1 with respect to either the continuous sedimentation or the continuous rate of supply model for age determination. With such a shallow core the problem of emergence must be evaluated, as well as sediment focusing due to changing distances from the shoreline or to dredged channels; "emergence" is taken to mean that shoreline receded at the site of interest such that the site was not continuously submerged, and that it was exposed subaerially for some period of time. The "crs" model assumes a continuous rate of supply, which is very unlikely to occur under alternating submergence - emergence conditions.

If the crs model is to be used for the Farmington Bay Core 1, issues such as bioturbation, erosion, sediment deposition hiatus, chemical mobilization should be evaluated. Although these issues may have been explored, they are not reported on in detail in the final reports.

3. The ^{137}Cs peak dates appear to differ for the three cores: 1936 for Site 4, 1928 for Site 3, and 1962 for Farmington Bay. Is there a scientific rationale for these discrepancies?

There is no scientific rationale for the ^{137}Cs peak to differ in their apparent "year-of-peak concentration" for different cores unless there is mixing of sediments or some unspecified chemical transport. For this case, ^{210}Pb dating was used to determine the sedimentation rate using a model that does not include sediment mixing. Mixing of sediments is well-documented in marine and lake sediments, often to a depth of about 5 to 8 cm. Sediment mixing in the Great Salt Lake cores of this study has not been evaluated in-depth in the final reports.

We note that the Leavitt et al (2012) report noted that only three cores were deemed "acceptable" for interpretation of sedimentation rates: Site 2 (Core 1), Site 5 (core 3), and Site 6 (core 1) were analyzed for ^{210}Pb and ^{137}Cs and were rejected as being suitable for further interpretation. And indeed, Leavitt et al (2012, lines 368-371) also noted that Site 3 (Core 1) gave a ^{137}Cs peak that was clearly "too old" using the derived ^{210}Pb chronology; this was attributed to "due to isotope migration, low sampling resolution (1 sample per ~15 years), or difficulty fitting ^{210}Pb regressions due to a mid-core peak of ^{210}Pb ." They further state (lines 381-385) that "Taken together, these patterns demonstrate conclusively that sediments obtained from Sites 2, 5 and 6 were highly mixed and could not be used to establish either basic chronology or historical changes in

algal production within Great Salt Lake. Such high variability in sediment deposition and mixing is expected in large shallow lakes".

Isotope migration in the solid phase (e.g., bioturbation) should affect ^{137}Cs and ^{210}Pb together. An apparent deepening of the ^{137}Cs can occur by bioturbation for a singular event such as the "bomb-spike"; however, the ^{210}Pb age model must then also include bioturbation (Berner, 1980).

Diffusion of ^{137}Cs will not change the depth of "peak ^{137}Cs " but would broaden the peak because diffusion would occur in both the "up" and the "down" direction.

Flow of water through the sediments, because of changing water depth could result in the migration of aqueous species. At high distribution coefficients (see Appendix) this is highly unlikely, although this could occur at low distribution coefficients. Most sediments have high distribution coefficients for ^{210}Pb and ^{137}Cs , and most trace metals in clay-rich systems (and hence, their respective use in understanding geochronology and pollution histories). Distribution coefficients for ^{210}Pb , ^{137}Cs , and other metals are not reported in this study.

Thus, although the age discrepancy at Site 3 was identified by the authors, it was not further considered in the paper and the ^{210}Pb sedimentation rate was used without further comment.

4. In all three cores, Pb-210 activity appears to drop to a base level around 3.5 or 4.0 (dpm/g). However, this leveling out appears at different years (1932 for Site 1, 1900 for Site 3, and 1920 for Site 4). Is there a scientific explanation for these discrepancies?

The "leveling out" of the supported ^{210}Pb specific activity at *ca.* 3 to 4 dpm/gm likely represents the pre-anthropogenic level of the concentration of ^{226}Ra in the sedimentary supply to Great Salt Lake. *In situ* radioactive decay of ^{226}Ra to daughter products would yield a "supported" level of ^{210}Pb of about 2 to 4 dpm/gm. The year of the drop to base level is directly related to the assumption of the sediment deposition rate; this study implicitly assumption that the "crs" model is appropriate with no mixing or erosion of sediment. All cores show distinct changes in slope at about 4 to 8 cm; this could be due to changes in sedimentation rate including a hiatus in deposition or erosion, or it could be related to sediment mixing by bioturbation.

It would appear that these differences are "model-driven" rather than real.

5. How certain are the dates for the Farmington Bay Core (Site 1) especially for the period between 1930 and 1963?

I do not have high confidence in the dating of Site 1 (Farmington Bay). The report has shown that many cores of sediment from Great Salt Lake cannot be dated sufficiently well using ^{210}Pb and ^{137}Cs for the purposes needed by UDWQ. For many purposes, the dating of these cores would be quite adequate. However, these cores are not suitable for a detailed chronology over the entire period of interest.

Site 1 shows a significant discontinuity in ^{210}Pb inventory, expressed either in dpm/gm or in dpm/cm². Figure 1 shows a comparison of values for Sites 1, 3, and 4. An ideal system with constant sedimentation rate and constant supply would exhibit a linear decrease from the surface (depth = 0). Site 3 was previously identified by the authors as giving problematic results; Figure 1 shows that Site 3 is more well behaved than Site 1 which has a discontinuity at about 7 cm depth in the excess ^{210}Pb . This could be due to a depositional hiatus, or a period of slight erosion and removal of sediment. I note that the water depth at Site 1 is only 15 cm and was likely exposed for several years in the 1960s; at such a shallow depth erosion or mixing of sediments is very likely.

The approach of Applesby and Oldfield (1983) was applied to this system by Leavitt et al (2009). However, even though problems were identified with the age model for Site 3, no attempt was made to evaluate why the ^{210}Pb and ^{137}Cs gave discrepant ages for that site. Furthermore, Site 1 also has far from ideal behavior and in any analysis of ^{210}Pb alone would suggest that sedimentation has not been constant at this site; possible erosion and sediment focusing at this very shallow site must be considered in any analysis. Site 1 was likely exposed in the early 1960s due to changes in lake level.

6. UDWQ is interested in distinguishing metals concentrations and ecological indicators (macroinvertebrates and algae) for discrete periods in the past especially the mid-1970s, the early 1950s when the first causeway was constructed between Gilbert and Farmington Bay, and the mid-1850s prior to sewage discharge to Great Salt Lake. Are the dates reported in the Levitt et al. 2012 reliable for distinguishing these three periods? If not, how much differentiation can be made (e.g. if decadal dating is uncertain can the cores provide information at multi-decadal scale?).

It is likely that these cores can provide information on metal concentrations and ecological indicators in the discrete periods including pre-European settlement (*ca.* prior to 1850), the early metal extraction period (*ca.* 1860 *ca.* 1960), and the post-causeway era (*ca.* 1960 to present). Within each of those periods the stratigraphic rules of superposition give a chronological order, and within each of those periods some consistent historical inferences will be able to be made. Assignments to discrete years, or

particular decades will be challenging, as will be recognizing the changes in sedimentation rates or gaps in the records. Large hydrologic changes are likely to be associated with multiple causeway construction activities; thus, there are likely to be gaps or changes in sedimentation rates associated with those periods. Certain tools appear to give important information in the right circumstances: ^{210}Pb , ^{137}Cs , metal concentration profiles can be used in combination to get good age estimates for certain events (e.g., the weapons testing peak of 1963; the initiation of large-scale smelting in SL Valley), or rates of sedimentation for certain portions of some cores.

The present analysis on all cores does is not adequate for distinguishing these periods except in the general sense. This is due, in part, to discrepancies noted by the authors that were not further addressed in their analysis. For example, Site 3 was shown to have discordant ^{210}Pb and ^{137}Cs results; the metal concentrations in Site 1 are at extreme odds the assumed continuous sedimentation at the site. Using all results (^{210}Pb , ^{137}Cs , and metal concentrations) is may be possible to determine a sedimentation history that is compatible with all results. Reliance only on a single method for dating is likely to be correct in only a few circumstances where the sedimentation history meets all assumptions in the model.

In any case, the top few centimeters represents the "post-bomb" (i.e., post-1960) history of each site, and the basal portion represents the pre- or early settlement of the valley by Europeans. The strongly fluctuating lake levels (greater than 5 meters), the causeway construction(s), dredging, and the extremely shallowness of the Farmington Bay core (0.15 meters) will result in sediment focusing and possibly even erosion that must be considered in any analysis of these cores.

7. Are any of the cores more or less certain in terms dating and chronology?

From my analysis of this data, it appears that Site 4 may be suitable for dating using these methods. Leavitt et al (2009) concluded that Sites 2, 5 and 6 were not suitable for dating using these methods; it is likely that Sites 1 and 3 also are not suitable for dating using these methods for the purposes of UDWQ. Discontinuities in ^{210}Pb concentrations in all cores are observed at about 5 to 8 cm depth. Changes in sedimentation rate, sediment focusing, sediment mixing, or erosion may be the cause for these problems. Once again, I note that the Farmington Bay core was collected at a depth of only 15 cm, and may well be a site that was exposed and possibly eroded during the 1960s low stand. Likewise, this site has experienced water depths from *ca.* 0 to *ca.* 5 meters and will be strongly affected by sediment focusing.

8. Does the reviewer have any other comments on the scientific quality of the study?

This study included field collection of cores from Great Salt Lake and laboratory measurements that include bio-indicators and a geochemical history of sedimentation. The field collection and lab measurements appear to be of high quality. The initial interpretation of the data concluded that the collected cores from 3 of the 6 sites were not suitable for dating because of sediment mixing throughout the core. The initial interpretation also concluded that there was a problem with the dating of a fourth core (Site 3); however, in spite of that conclusion no evaluation was made as to why the ^{210}Pb and ^{137}Cs results were not concordant. This would seem to be an important follow-on to such an important observation. Furthermore, consideration of down-core concentrations shows only one core (Site 4) gives a concordant history that accommodates the trace-metal data, the ^{210}Pb data, and the ^{137}Cs data. Site 1, which was collected in a water depth of 0.15 meters, certainly does not meet the assumptions of a simple history with continuous sedimentation; Site 1 would have been exposed (i.e., above lake level) several years at the low stand of the lake in the 1960s. Such exposure compromises its value if this fact is not taken into account. Overall, it appears that Site 4 may be the most suitable to interpret using the simple sedimentation history model. Further analysis of additional cores may provide a coherent chronology that would meet the goals of UDWQ for understanding changes in Great Salt Lake including Farmington Bay related to anthropogenic discharges into the hydrologic system; at present this is not the case.

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Figure Captions.

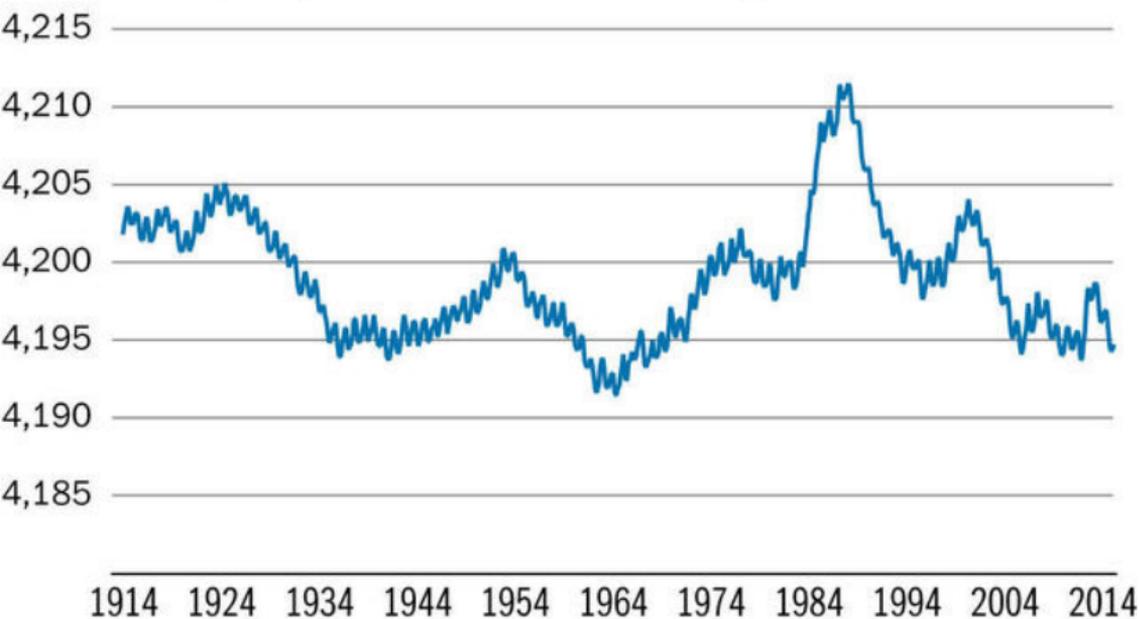
Figure 1. Level of Great Salt Lake. From:

<http://img.deseretnews.com/images/article/graphicSidebar/1393737/1393737.jpg>

Figure 2. Figure 2. Comparison of excess ^{210}Pb determinations for Site 1 (A), Site 4 (B), Site 3 (C). Dashed black line is best-fit to all data; red line is best-fit to data above and below vertical blue dashed line. Vertical blue dashed line represents possible discontinuity in sedimentation style or rate, including bioturbation.

Great Salt Lake levels

Water elevation (in feet) for the Great Salt Lake over 100 years



SOURCE: U.S. Geological Survey

DESERET NEWS GRAPHIC

Figure 1. History of the level of Great Salt Lake over the past 100 years. From: <http://img.deseretnews.com/images/article/graphicSidebar/1393737/1393737.jpg>

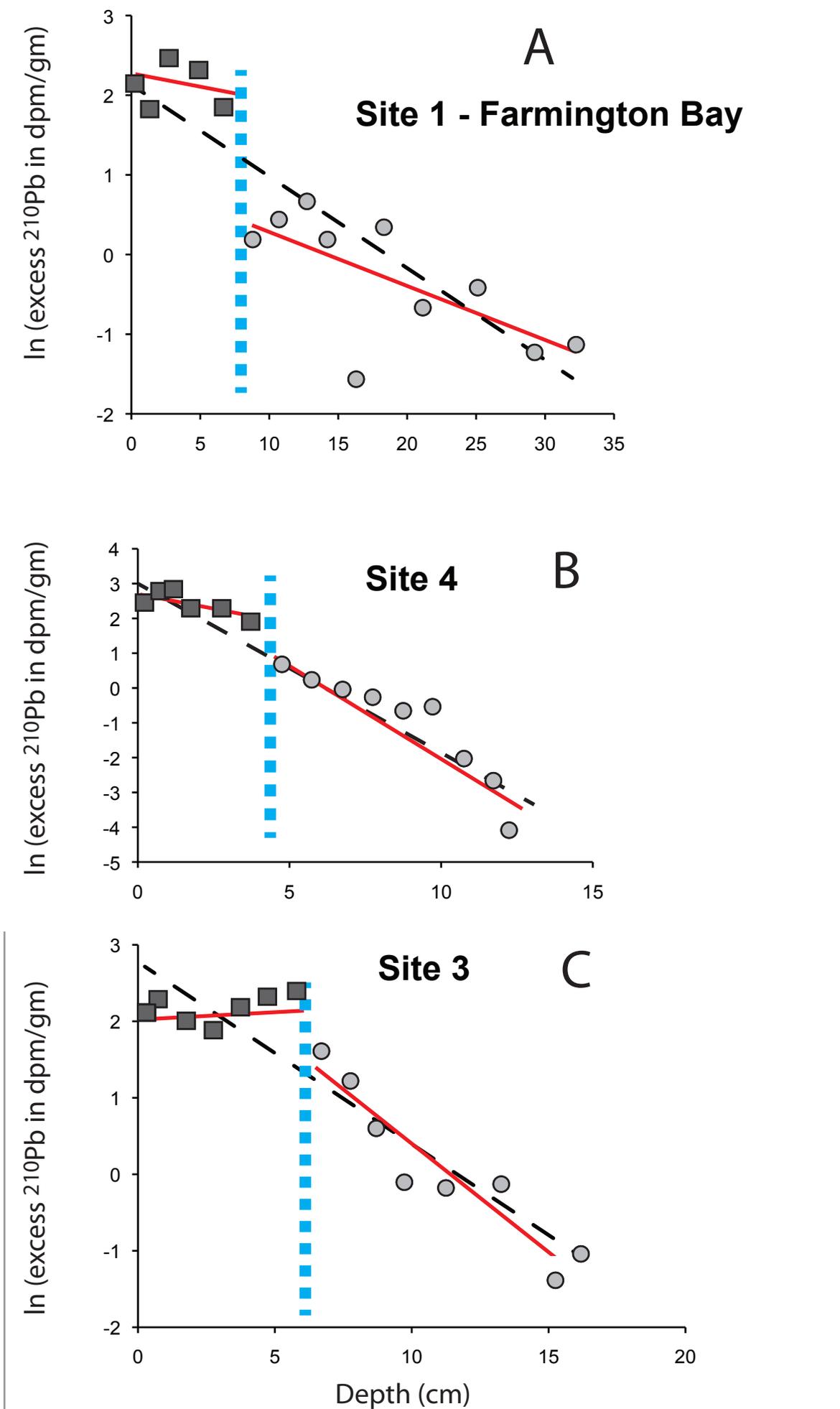


Figure 2. Comparison of excess ^{210}Pb determinations for Site 1 (A), Site 4 (B), Site 3 (C). Dashed black line is best-fit to all data; red line is best-fit to data above and below vertical blue dashed line. Vertical blue dashed line represents possible discontinuity in sedimentation style or rate, including bioturbation.

Box 1. Simple and complex systems for dating lake sediments

Many lake systems have simple sedimentation histories that are modeled using a simple one-dimensional advection-radioactive decay model. In this model, chemical reactions for relevant species are minimal because of the high distribution coefficient between the solid and mineral phase. In such system, there is no measurable transport due to diffusion in the aqueous phase because of the low dissolved concentrations, and no measurable transport by advective flow. In such a simple system, the two radiochemical methods – ^{210}Pb and ^{137}Cs – should be in broad agreement and they should also be in broad agreement with metal concentrations associated with known metal contamination.

The simple system is described by the equation:

$$\partial C_i / \partial t = -v_i \partial C_i / \partial z - \lambda_i C_i \quad [1]$$

where C_i is the concentration of the species of interest, t is time, v_i is the sedimentation rate, z is the distance below the sediment water interface, λ_i is the radioactive decay constant of the species of interest. For ^{210}Pb , the constant sedimentation rate model (csr) and the constant rate of supply model (crs) use these assumptions for the steady-state condition:

$$\partial C_i / \partial t = 0 \quad [2]$$

with the simple solution for the "csr" case:

$$\ln C_z / C_{z=0} = \lambda_i / v_i z \quad [3]$$

This equation is a simplification of a more general equation describing chemical transport of in a complex system (Berner, 1980):

$$\partial C_{i,s} / \partial t = D_{i,s} \partial^2 C_{i,s} / \partial z^2 - v_{i,s} \partial C_{i,s} / \partial z - \lambda_{i,s} C_{i,s} + \Sigma R_{i,s} \quad [4]$$

where $D_{i,s}$ is the diffusion coefficient for solids and $\Sigma R_{i,s}$ represents the sum of all chemical reactions involving the species of interest (i). Because diffusion coefficient for solids is on the order of 10^{-12} or smaller, and the characteristic length L for diffusion is

$$L^2 = 4Dt \quad [5]$$

diffusion for solids can be ignored for length scales greater than one centimeter on times scales of 100s of years. Biodiffusion (sediment mixing by organisms) can be considered as part of this simple system, but must be explicitly accounted for if included. Biodiffusion often has relevant depths of *ca.* 4 to 8 cm (e.g., see Turekian and Bacon, 2014).

Chemical reactions are coupled to the aqueous phases by chemical reactions, including the distribution coefficient K_d for trace elements where:

$$K_{d,i} = C_{i,s} / C_{i,aq} \quad [6]$$

$C_{i,s}$ and $C_{i,aq}$ are the concentrations of species i in the solid and aqueous phases, respectively. For clay-rich systems where the distribution coefficient for ^{210}Pb , ^{137}Cs , and other metals is high (i.e., > 100), and therefore the concentrations of these species in the aqueous phase are low, aqueous transport by diffusion or advection can be ignored. For systems where $K_{d,i}$ values are small (i.e., < 10), or oxidation/reduction reactions need to be considered, transport and back reaction with the solid phase needs to be considered and the related coupled equation be evaluated:

$$\partial C_{i,aq} / \partial t = D_{i,aq} \partial^2 C_{i,aq} / \partial z^2 - v_{i,aq} \partial C_{i,aq} / \partial z - \lambda_i C_{i,aq} - \Sigma R_{i,aq} \quad [7]$$

When these reactions are significant, this is considered to be a complex system and significant modeling is needed to help understand the system. Only at negligible aqueous concentrations (i.e., large $K_{d,i}$) can diffusion be ignored on these time scales because aqueous diffusion coefficients are on the order of 10^{-6} (i.e., the characteristic length for diffusion on 100-year time scales is > 10 cm).

In addition, a simple system has continuous sedimentation (i.e., no hiatus in sedimentation) or a constant rate of supply (i.e., no sediment focusing changes through the interval of interest), no erosion events. Occasional exposure, such as due a drop in lake level, is an obvious problem whereby the assumption of a "simple system" has been violated.

Understandably, the authors have used the "simple system" for calculating ages and sedimentation rates. Mismatch between methods of determining the age models is a good way to determine whether the "simple system" model is valid for each site. Transient solutions to equation [4] are not unique.

References

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