

Henrieta Dulaiova

We thank Henrieta Dulaiova for reviewing our manuscript. We appreciate her comments and we feel they have significantly strengthened the paper.

Below, we reply to each of her comments/suggestions.

Note: Page/line numbers in reviewer's comments refer to the original manuscript. Our references to page numbers refer also to the original manuscript. In the revised manuscript page number may differ.

1. "...radon activities in Lake A were very low (<1 Bq/m³) and one wonders if a radon cryogenic extraction method which has significantly lower MDAs would have been a better technique for this lake."

Response: We agree with Mrs. Dulaiova that the radon cryogenic extraction technique can be an excellent method if very low radon concentrations are expected. However, unfortunately this technique is not suitable to our specific case. Cryogenic techniques require LN₂ and helium. As our field conditions were very rough and we did not have access to a nearby site where a cryogenic system could be assembled, this technique is not practical in our case. Hence, we were only able to perform this investigation because a reliable and robust field-method (the RADAqua/RAD7 setup) has become available in the last few years.

2. "Although the RAD7 detector has vanishingly low background, stating that 0.5 Bq/m³ of radon was detected with confidence needs some proof. As the RADAqua is not usually applied for measurements of such low levels of radon, a list of the measurement parameters such as counting time, temperature and the corresponding MDA should be included in the manuscript."

Response: As mentioned in the manuscript, the RAD7 is a radon-in-air detector. For the determination of radon-in-water concentration the RAD7 was used in conjunction with the RADAqua. For the calculation of the radon-in-water concentration, the partition coefficient between water and air (K) was calculated (Weigel 1978). The respective measured radon-in-air concentration for Lake A was always higher than 2.5 Bq/m³.

At every spot our counting time was about 9h. By considering the system efficiency, the minimum detectable activity (MDA) was estimated to be 2.1 Bq/m³ (see MDA equations in Currie, 1968). In addition, at Lake A, we used two modified RAD7s (both have higher efficiencies than a "normal" RAD7) in parallel. The results were similar. We thus feel confident that even though the concentrations are very low, they are reliable.

We follow Mrs. Dulaiova's advice and added measurement parameters (Counting time, temperature, MDA) in the manuscript

Table 1: "...At each spot the same RAD7 setup was used (counting time: 9h, MDA radon-in-air: 2.1 Bq/m³). Water temperature during the March and July campaign was 2°C and 18°C, respectively"

Furthermore we added the following in section 2.3 Sampling and sample analysis (page 4998).

"...For determination of the radon partition coefficient between water and air, the water temperature in the system was measured continuously with a HoBo temperature

sensor and then calculated using an empirical equation presented by Weigel (1978). For the estimation of the radon concentration in the lake water, the specific counting time of the RAD-7 was set to about 9h. By considering the system efficiency, the minimum detectable activity (MDA) was estimated to be 2.1 Bq/m³ (see MDA equations in Currie, 1968)."

3. "The authors state that the surface water ²²²Rn is not supported by ²²⁶Ra dissolved in the water column because ²²⁶Ra was below detection limit. Was the surface water ²²⁶Ra measured with the same sensitivity as ²²²Rn? What was the MDA of the ²²⁶Ra measurement?"

Response: We agree that more detailed information about the ²²⁶Ra technique would be useful. The detection limit of the system for Ra-226 is < 0.4 Bq/m³. We added the following clarifications to the Methods section (page 4998):

"...For the estimation of in situ radon production through decay of ²²⁶Ra dissolved in the water column, radium was measured using gamma-spectrometry. About 50 L of lake water was passed through a manganese-impregnated acrylic fiber cartridge, which quantitatively extracts radium from water (Moore and Reid 1973). In the lab, the fiber was leached with 0.25 mol/l hydroxylamine hydrochloride and HCl to remove the manganese dioxide and the adsorbed radium. The solution was filtered through a 0.45 μm membrane filter, and 6 ml of saturated Ba(NO₃)₂ solution was added to the filtrate. H₂SO₄ (17%) was then added to the solution to precipitate Ba(Ra)SO₄, which was then collected, rinsed, dried, and stored for several days to allow daughter in-growth (Moore 1984). The precipitate was then placed on a germanium detector for the quantitative analysis of radium-226. The radionuclide was quantified by measuring the intensity of gamma-rays of ²¹⁴Pb (295.2 and 351.9 KeV) and ²¹⁴Bi (609.3 KeV). The detection limit of the system for ²²⁶Ra is < 0.4 Bq/m³."

4. "Using the parameters given in the manuscript I reproduced the groundwater discharge rate calculation for both lakes. I did not find porosity and water temperature and had to estimate the temperature by iteration until my atmospheric evasion rates matched the ones indicated in the manuscript."

Response: We really appreciated the effort put in by the reviewer to double check our calculations. To prevent any reader from having to guess temperatures and porosities in similar exercises, we added the following:

Page 4999: *"...(organic-rich silt, with a porosity of 0.8) ..."*

Page 5002: *"..., with a porosity of 0.37, ..." and: "..., with a porosity of 0.77, ..."*

Table 1: *"... Water temperature during the March and July campaign was 2°C and 18°C, respectively."*

Table 4: *"... Water temperature during the March and July campaign was 1.2°C and 18°C, respectively."*

5. "For Lake B groundwater discharge from the silty area, which represents 95% of the lake bottom, significantly influences the final groundwater discharge budget. I wonder whether those discharge rates are real. Is it possible that groundwater discharge only occurs in the sandy area and the radon measured in the other parts of the lake is only a result of mixing with high radon water? How would the authors account for this mixing?"

Response: Lake B can be divided into two parts based on the measured radon concentration - an area of high and an area of lower concentration. In each area the

radon concentration is homogeneously distributed. This implies that the lake is not well mixed related to radon, i.e. mixing of lake water takes longer than the half-life of radon.

So, if we assume that mixing rates within the lake is constant, there should be a constant decrease in the radon from the area of high concentrations (sandy area) to the area of low concentrations (silty area). However, the concentrations within the two areas do not change, implying that both areas are characterized by constant groundwater input as described in the manuscript.

6. “Would that scenario change the total groundwater discharge rate for the whole lake?”

Response: If we apply this scenario, i.e. all the groundwater input only occurs in the sandy area, the total groundwater discharge rate is different. It would be about 20% of the original estimation. We added this information on page 5004.

7. “The other question is how much confidence we should have in assessing advection rates of 0.01-0.068 cm/day. Is the radon approach really sensitive enough to detect advection rates as low as 0.01 cm/day?”

Response: We feel that the sensitivity of the approach will be largely dependent on the radon analysis method. As described above, we took precautions to make sure that the radon concentrations are above the MDA. We thus see no reason why the calculated radon fluxes and the used equations would not be sensitive enough.

8. “I agree with the authors’ conclusion that groundwater discharge is insignificant for the water budget of Lake A. How about the silty part of Lake B?”

Response: The silty part of Lake B covers 95% of the total area. Under the consideration of the calculated groundwater advective rate (0.04 cm/d) the groundwater discharge amounts to about 23,000 m³/month. This is about 80% of the total groundwater discharging into the lake. Hence, the silty part plays a significant role in the contribution of groundwater to the overall water budget of Lake B. We added this information on page 5004

9. “The authors assumed no seasonality in groundwater discharge rates. I think that is an incorrect assumption. Seasonality of groundwater discharge will depend on many factors, for example whether the lakes are recharged from a water table aquifer which would respond to rain patterns almost immediately or a confined aquifer with a slower response to climate variations. In any case, when groundwater discharge is compared to the overall water budget, one has to assume some variation. Therefore lines 20-23 on page 5004 should be worded more carefully.”

Response: We agree that there may be a large seasonal variability in groundwater discharge rates in these lakes. We emphasize, however, that logistical constraints prevent field work during the peak winter. We now highlight in the text that seasonality may have not been captured by our surveys and that our work represent a first-order approximation in these remote lakes.

We also added to the manuscript a more detailed discussion about seasonality (page 5002):

“...This is further supported by the fact that the mineral soil in that region, which represents the main aquifer, is overlain by thick saturated peat that extends deeper than seasonal frost penetration, implying that groundwater discharge is constant over

time. However, future investigations are needed for more detailed information about the seasonal variability in groundwater discharge rates.”

Furthermore we toned down the section related to the discussion of the relevance of groundwater to the water budget (page 5004).

Specific comments:

Page 4992, line 16: “...[radon] is transported with it through the aquifer.” –or until it decays. Consider rewording as depending on the transit time radon may not be transported through the aquifer but may decay or re-equilibrate with ²²⁶Ra to a different activity.

Response: We agree with the reviewer. We added: “...*radon decays or re-equilibrates with the surrounding radium-226 to a different concentration.*”

Page 4992, line 21 and throughout the manuscript: “activity concentration” pick activity or concentration, using both is redundant

Response: We agree that it is common nowadays to use only activity or concentration. We changed all “activity concentrations” to only “concentrations”.

Page 4993, line 5: “...in a water column representative of the lake water body.” The definition is incomplete as it does not consider uneven groundwater discharge through the lake bottom and inefficient mixing of the lake water.

Response: We agree. To state this in a better way, we complete the definition: “...(under the consideration of homogeneous groundwater discharge through the lake bottom and a well mixed lake water body)”

Page 4995, line 25: “[radon] is in decay equilibrium with the radium...” it should be in radioactive equilibrium instead.

Response: We changed the text to: “..., *radioactive equilibrium*”

Page 5001, lines 10-13. Homogeneous pH and conductivity distribution does not necessarily mean complete mixing and pH may be influenced by other biochemical processes.

Response: We agree that pH may be influenced by a number of processes. We have toned down this statement by using the words “...*imply a well mixed lake*”.

As conductivities in groundwaters are different than conductivities in surface waters, we believe that a homogeneous conductivity distribution in the lake supports the idea of a well mixed lake.

Table 2. This table implies that diffusion (F_{diff}) was not considered for advection rate calculations for 1-A, 2-A, and 4-A. However, based on the math it was included in the radon mass balance.

Response: We agree with Mrs. Dulaiova again - diffusion was considered for every sampling point. We changed the table caption to state this in a clear way.

Once again, we thank Ms Dulaiova for her insightful and constructive comments.

Literature:

Weigel, F.: Radon, *Chemiker-Zeitung*, 102, 287-299, 1978.

Currie, L. A.: Limits for qualitative detection and quantitative determination, *Anal. Chem.*, 40, 586-593, 1968.

Moore W.S. and Reid, D.: Extraction of radium from natural water using manganese-impregnated acrylic fiber, *J. Geophys. Res.*, 78, 8880–8886, 1973.

Moore, W.S.: Radium isotopic measurements using germanium detectors, *Nucl. Inst. Methods*, 223, 407–411, 1984.