

Interactive comment on “Tracing and quantifying groundwater inflow into lakes using radon-222” by T. Kluge et al.

M. Schubert (Referee)

michael.schubert@ufz.de

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"general comments"

The paper discusses the modification and combination of commercially available portable equipment for radon-in-water analysis and presents a resulting technique, which allows detection of radon concentrations down to 3 Bq/m^3 . While such low detection limit is not necessary for determination of radon concentrations in groundwater samples, the very low radon concentrations found in open water bodies demand such detection limit and accuracy. Hence, the developed methodology represents an additional useful tool for the determination of radon concentrations in lakes and rivers. An exemplary application of the developed methodology is the quantification of groundwater exfiltration into open water bodies. A respective on-site application is discussed

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Interactive Discussion

Discussion Paper

exemplarily at the end of the paper.

The used equipment and the applied method is clearly described and sufficiently discussed. However, some critical remarks have to be made. They include the following general aspects.

- Some of the assumptions made are not supported by data available to the reader and, hence, appear somewhat vague (in particular concerning the field study).
- Sampling and sample handling is one of the most difficult tasks as far as radon-in-water analysis is concerned. The errors due to radon loss by degassing, which are practically inevitable in connection with lake water sampling and sample handling, are not discussed sufficiently (the only way to avoid them would be in-situ radon extraction from the lake).
- The term "Concentration (or solubility) equilibrium" should be strictly told apart from "decay equilibrium".
- All radon concentrations are given as total radon, which might be misleading. If groundwater exfiltration into a lake is to be quantified, only "excess radon" concentrations are of interest. Hence, the radon, which is supported by ^{226}Ra dissolved in the lake water must be taken into account. Only two (which is way too few) water samples have been checked for their ^{226}Ra activity concentration (in particular, some information on ^{226}Ra in the thermocline is essential in the given case).
- The authors refer to "groundwater-lake interaction". Since the expression "lake" stands for a complete ecosystem it might be better to use the term "groundwater - lake water interaction".

"specific comments"

Title: I would rather see the main aspect discussed in the manuscript in the developed and used METHOD. The investigation of the lake is just an exemplary on-site application of that method. Since the weak points of the paper are mainly to be found

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Interactive Discussion

Discussion Paper

in the section discussing the on-site experiment, the title should rather focus on the straightforwardness of the developed sampling and measurement procedure (and the on-site section should be boiled down to the essential and data-supported facts and conclusions).

Page 1521; Line 5/6: You should quote: Lee, D.R. (1977): A device for measuring seepage flux in lake and estuaries. *Limnology & Oceanography* 22, 140-147.

Page 1521; Line 16: You better write "of environmental tracers".

Page 1521; Line 17/18: You should verify your statements by some key citations.

Page 1522; Line 7: Here you should quote: Macintyre, S., Wanninkhof, R., Chanton, J. P. (1995): Trace gas exchange across the air-water interface in freshwater and coastal marine environments. In: P. A. Matson, R. C. Harriss (Eds.): *Methods in Ecology-Biogenic Trace Gases: Measuring Emissions from Soil and Water*. Blackwell Science, Cambridge, Mass., pp. 52-97.

Page 1522; Line 7/8/9: All your citations here are older than two decades. Aren't there any more recent ones?

Page 1522; Line 10/11: Don't give the concentration ranges so matter-of-factly. Write "about". Besides, other authors report other ranges (e.g. Tuccimei, P., Salvati, R., Capelli, G., Delitala, M.C., Primavera, P. (2005): Groundwater fluxes into a submerged sinkhole area, Central Italy, using radon and water chemistry. *Applied Geochemistry* 20, 1831-1847.)

Page 1522; Line 13: Here you should quote: Schubert, M., Knoeller, K., Treutler, H.C., Weiss, H., Dehnert, J. (2006): On the Use of Naturally Occurring Radon-222 as Tracer for the Estimation of the Infiltration of Surface Waters into Aquifers. In: Povinec, P. (Ed.): *Radionuclides in the Environment, Radioactivity in the Environment*, Elsevier Book Series, Amsterdam, 326-334.

Page 1522; Line 17: Here you should quote: Trettin, R., Glaesser, W., Lerche, I.,

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Interactive Discussion

Discussion Paper

Seelig, U., Treutler, H.C. (2006): Flooding of lignite mines: isotope variations and processes in a system influenced by saline groundwater. *Isotopes Environ Health Stud.* 42,159-79.

and

Tuccimei et al. (2005), given above.

Page 1522; Line 25: You might want to read and mention here: Treutler, H.C., Just, G., Schubert, M., Weiss, H., (2007): Radon as tracer for the determination of mean residence times of groundwater in decontamination reactors. *J. Radioanal. Nucl. Chem.* 272, 583-588.

Page 1523; Line 3: Why do you distinguish between lake water and surface water? Lake water is surface water, isn't it?

Page 1523; Line 6: Modified from what?

Page 1523; Line 21: The half-life of ^{214}Pb is 26.8 minutes.

Page 1523; Line 23: If you talk about liquid/gas extraction of radionuclides, always distinguish between "concentration (or solubility) equilibrium" and "decay equilibrium". The first must have been established before the second can build up.

Mention the chamber sensitivity of the RAD7 which is, as far as I know, 1 cpm at 75 Bq/m³.

Page 1524; Line 2: You better say "radon-tight", because "air-tight" doesn't mean much in the given context.

Page 1524; Line 5: Again: "concentration equilibrium"

Page 1524; Line 11: You should add "in most cases".

Page 1524; Line 19: "Several hours" should be quantified.

Page 1525; Line 1: Again, you better say radon-tight. Besides, I would put the exper-

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Interactive Discussion

Discussion Paper

imental results, which (seem to) confirm the radon tightness of the buckets (Fig. 3), in THIS section, since the respective question emerges here. By the way, polypropylene doesn't have that much of a good reputation for being radon-tight. I would suggest some additional experiments to confirm the radon-tightness of the buckets (in particular the lids).

Page 1525; Line 6: Ahhh, here we go (see: Page 1523; Line 6). Mention that your gas exchanger is actually a modified "RAD Aqua".

Page 1525; Line 20: You must quantify that observation because all RAD7 users (as well as the Durrigge staff) will be quite surprised about the news. Actually, I am surprised myself, too, since I did some gamma spectrometry of the desiccant and couldn't find anything suspicious.

Page 1525; Line 28ff: Again: specify what equilibrium you are talking about, decay or solubility. Solubility equilibrium doesn't need "several h".

Page 1526; Line 11: You should specify room temperature. The Ostwald coefficient of radon changes dramatically between 273 K (0,5249) and 298 K (0,2263).

Page 1526; Line 16: You should add a citation where the SF6-sampling system you used is described in detail.

Page 1526; Line 17: The pump rate should be chosen in a way to avoid turbulence while filling the bucket. AND the bucket should be filled slowly from the bottom up. I guess you did it that way, but mention it.

Page 1526; Line 22: Cite at least one reference.

Page 1526; Line 25: Why four weeks? What does "negligible" mean? You can pretty much quantify that. You need exactly 23 days to get rid of 98.4% of the excess radon.

Page 1528; Line 13: We are talking about groundwater inflow and lake water outflow.

Page 1528; Line 14: Cite a reference.

Page 1529; Line 9: What do you mean by shallow groundwater? You better refer to the depth.

Page 1529; Line 11: Keep in mind that the groundwater will have to migrate through the sediment layer, which might yield a radon concentration completely different to that of the sandy gravels of the aquifer. If the thickness of the sediment layer is e.g. 25 cm and the groundwater exfiltrates with 1cm per day (which seems to be possible in the given case) the radon concentration of the exfiltrating groundwater will reflect the sediment rather than the aquifer material.

Page 1530; Line 20ff: Why? Only the EXCESS radon is of interest here. You should give the excess radon data in all tables and figures.

Page 1530; Line 27: What is the radium activity of the thermocline water? This value is very important because of the high radon activity concentrations measured there. You have to confirm that the high radon concentration in the thermocline water is not supported by dissolved 226-Ra.

Page 1531; Line 3: Why did you take all that effort and didn't just use the RAD7 for a simple on-site measurement of the radon concentration of the outside air? The RAD7 is suitable for such measurements (or even made for it).

Page 1531; Line 10: $C\text{-Ra} = \text{Lambda-Ra} * c\text{-Ra}$ looks strange to me. What do you mean with c-Ra, I mean, what unit? mg/l ?

Page 1532; Line 1: Did you carry out any batch experiments with the sediment to proof that the diffusive radon flux from the sediment is negligible or how do you know? You mention the likeliness of such flux later (Page 1532; Line 25).

Page 1532; Line 4: Why only in the hypolimnion? You found even higher radium concentrations in the epilimnion.

Page 1532; Line 4: Didn't you record depth profiles of temperature, pH and Eh?

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Discussion Paper

Page 1532; Line 22ff: Again, you cannot simply assume that. You cannot simply set $F_{sed} = 0$, in particular in the hypolimnion. You must consider F_{sed} . You even mentioned the likeliness of its influence just a couple of lines above.

Page 1533; Line 9: How can you be so sure? You don't have any information on dissolved ^{226}Ra in that layer, do you? You didn't report it, anyway.

Page 1533; Line 13: Why do you "assume" when you can calculate (Macintyre et al. 1995)? The argument that 22 Bq/l (total Rn) is valid for the whole epilimnion seems to be quite unsupported to me. You realize and correct that yourself on page 1534 line 12. Why do you mention it on page 1533 in the first place if it seems to be wrong anyhow?

Page 1537; Line 27: This reference is not mentioned in the text.

Figures and Captions There is way too much text in the table and figure captions. Everything is mentioned in the main text already.

Mark the groundwater flow direction in Fig. 4.

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