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Hydrology and Earth System Sciences

# Tracing and quantifying groundwater inflow into lakes using a simple method for radon-222 analysis

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**Abstract.** Due to its high activities in groundwater, the radionuclide $^{222}$Rn is a sensitive natural tracer to detect and quantify groundwater inflow into lakes, provided the comparatively low activities in the lakes can be measured accurately. Here we present a simple method for radon measurements in the low-level range down to 3 Bq m$^{-3}$, appropriate for groundwater-influenced lakes, together with a concept to derive inflow rates from the radon budget in lakes. The analytical method is based on a commercially available radon detector and combines the advantages of established procedures with regard to efficient sampling and sensitive analysis. Large volume (12 l) water samples are taken in the field and analyzed in the laboratory by equilibration with a closed air loop and alpha spectrometry of radon in the gas phase. After successful laboratory tests, the method has been applied to a small dredging lake without surface in- or outflow in order to estimate the groundwater contribution to the hydrological budget. The inflow rate calculated from a $^{222}$Rn balance for the lake is around 530 m$^3$ per day, which is comparable to the results of previous studies. In addition to the inflow rate, the vertical and horizontal radon distribution in the lake provides information on the spatial distribution of groundwater inflow to the lake. The simple measurement and sampling technique encourages further use of radon to examine groundwater-lake water interaction.

## 1 Introduction

In the Rhine Valley a large number of dredging lakes have been created during the last century as a result of extraction of gravel and sand. These lakes are now often used as recreation areas. To estimate the impact of the newly created lakes on the hydrological system or the ecosystem by changes in the lakes themselves (direct introduction of pollutants or nutrients) or in the surroundings (fertilization, creation of new lakes and wells for water supply) more information about the groundwater-lake water interaction is needed. This information is also of interest for the use of groundwater wells for drinking water purposes. The determination of the groundwater-lake water interaction is also important for the estimation of the nutrient budget and the prediction of the bio-geochemical evolution of the lake water, especially the eutrophication. The connection to the groundwater system is the most important component in the water budget of these lakes.

A network of seepage meters can be used to estimate directly the inflow of groundwater (Lee, 1977). However, the uncertainty of this approach is high. If the aquifer is heterogeneous, the spatial variation of the inflow can be large (Krabbenhoft et al., 1990), requiring a considerable number of seepage meters and a large effort to get acceptable results.

Hydraulic heads of the groundwater in the lake's surroundings and typical evaporation and precipitation values can be used to determine the groundwater inflow indirectly. Using this method leads to large uncertainties due to unknown flow paths and small scale changes in the hydraulic conductivity (Wollschläger et al., 2007). Furthermore, low hydraulic head gradients as in our study area may make it difficult to obtain sufficient precision.

The groundwater inflow can also be estimated indirectly by balancing of environmental tracers, such as stable isotopes, tritium, salts or radon. The stable isotopes deuterium and $^{18}$O are a useful tool to examine the groundwater-lake interaction (Krabbenhoft et al., 1990). However, the problem of this method is the relatively large uncertainty resulting from the measurement error and seasonal variability of the isotopic composition of the evaporation flux, the lake water and the groundwater inflow (Yehdegho et al., 1997). The isotopic composition of the evaporative flux can not be measured directly and theoretical approaches are charged with high uncertainty. Although the isotopic composition of the

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lake water can be measured easily, small uncertainties have a large influence on the mass balance because of the comparatively small signal differences between lake- and groundwater.

We used in a similar way the radon isotope $^{222}$Rn as a budget tracer. $^{222}$Rn is a radioactive noble gas isotope, which is produced by the decay of $^{226}$Ra within the natural decay chain of $^{238}$U. It decays by $\alpha$-emission to $^{218}$Po with a half life of 3.82 d. Despite the existence of other, more short-lived radon isotopes, we use the term radon synonymous to $^{222}$Rn, which is the relevant isotope in our context. Radon emitted from the rocks or the sediment grains is dissolved and transported by the groundwater. As a result of outgassing to the atmosphere and the short lifetime of radon, concentration gradients are created between surface waters and groundwater.

The strong gradients of radon between the reservoirs are used for applications in aquatic systems. For example, $^{222}$Rn has been used as a tracer for the examination of the air-sea gas exchange (Roether and Kromer, 1978; Kawabata et al., 2003) or the estimation of vertical and horizontal mixing near the bottom boundary of lakes (Imboden and Emerson, 1984; Colman and Armstrong, 1987). Radon is particularly well suited to study groundwater-surface water interaction, because activity concentrations in groundwater (on the order of 1 to 100 kBq m$^{-3}$, depending on the lithology) are much higher than in surface water (about 1 to 100 Bq m$^{-3}$). This contrast has been used to study groundwater recharge and flow in the vicinity of rivers (Hoehn and von Gunten, 1989; Schubert et al., 2006). Furthermore, radon has been applied successfully and quite extensively in the investigation of submarine groundwater discharge (e.g. Cable et al., 1996; Corbett et al., 1999, 2000; Crusius et al., 2005). Similarly, some studies used radon to asses groundwater exfiltration into lakes (Corbett et al., 1997; Tuccimei et al., 2005; Trettin et al., 2006).

More frequent application of radon in groundwater-lake water interaction studies may be impeded by the specialized and extensive measurement procedure. In a conventional method for radon detection at low activity levels described by Mathieu et al. (1988), used for instance by Corbett et al. (1997, 1999, 2000), radon is extracted from large water samples by stripping with radon-free He, enriched on a charcoal trap cooled with dry ice, and subsequently detected in a scintillation cell. More recently, methods for continuous radon measurement have been proposed and used in studies of submarine groundwater discharge (Burnett et al., 2001; Lambert and Burnett, 2003; Dulaiova et al., 2005). This approach offers the advantage of time series but requires time consuming measurements in the field with the respective logistical effort. While adequate for studies in coastal regions with radon activity levels in the range of 100 Bq m$^{-3}$ and variability on the tidal time scale, this approach may be less suitable for application in lakes, where even lower but steady radon activities are to be expected.

In this paper, an alternative, comparatively simple and low-cost method is presented, which can be used in the low-level range of surface waters without enrichment of radon and with a low effort in the field. As the continuous techniques, our method is based on a commercially available radon alpha spectrometer, which detects the radon activity in the gas phase. Using large sample containers and an appropriate transfer system for radon from the sample water to the gas phase, the water activity can be determined at low levels with a sufficient accuracy with regard to the application in lakes.

In addition to the analytical method, a concept for the derivation of the groundwater inflow rate from the radon balance within the lake is discussed. The applicability of this approach is demonstrated by a case study at a dredging lake in the Rhine Valley.

## 2 Materials and methods

Our method is based on the determination of the radon activity with the commercial alpha spectrometer RAD7 (Durridge Company Inc., Bedford, MA, USA), which measures radon in the gas phase. The radon activity is deduced from the activity of its decay products (primarily $^{218}$Po). The alpha particles are detected in a discrete energy spectrum by a semiconductor crystal. The energy sensitive counting allows to separate all alpha-emanating polonium isotopes. The half-life of $^{218}$Po (daughter of $^{222}$Rn) is 3.1 min. The radon concentration can be derived by counting of the decay of $^{218}$Po, which is almost in equilibrium with the radon decay in about 15 min. To increase the precision, $^{214}$Po can be taken into account as well. Due to the longer half-life (26.8 min, 19.9 min) of the intermediate nuclides $^{214}$Pb and $^{214}$Bi this can only be done after about 3 h when the secular equilibrium is reached. The spectrum is recorded from 0 to 10 MeV with a resolution of 0.05 MeV (Durridge Co., 2000). Our device has a chamber sensitivity of 1 cpm at 151.7 Bq m$^{-3}$. Typical background count rates are 1.2 to 1.5 counts per hour within the energy window of $^{218}$Po.

Durridge Company offers two systems (Rad $H_2O$, Rad Aqua) for the extraction of radon from water samples and measurement with the RAD7 detector. Rad $H_2O$ is designed for the analysis of small water samples (250 ml) with high activities (>400 Bq m$^{-3}$). The air is circulated in a closed cycle through a radon-tight water bottle, containing the sample to extract the radon. The other system (Rad Aqua) is designed for continuous measurements. Sample water is pumped through a gas exchange cylinder, which is connected to the air cycle. Radon outgasses from the water until solubility equilibrium is reached. In both cases the air cycle is connected to the radon alpha detector via a drying unit to remove water vapor. Detailed descriptions of the RAD7 detector as well as the two water extraction procedures are available (Durridge Co., 2000, 2001a, 2001b).

**Table 1.** Different analytical methods and their detection limit. Counting times are typical values for the respective methods and are in part limited by the increasing background from the drying medium, except if a water trap is used. The detection limit is defined as the activity concentration that – using the stated sample size and during the given counting time – produces a background-corrected signal which deviates two standard deviations from zero (i.e., the $1\sigma$ uncertainties of the listed values are 50%).

| Method | Sample volume (L) | Counting time (min) | Detection limit (Bq m$^{-3}$) |
|---|---|---|---|
| Rad $H_2O$ | 0.25 | 20 | 400 |
| Rad $H_2O$ with $N_2$-purging | 0.25 | 60 | 160 |
| Rad Aqua | continuous | 30 | 10 |
| Rad Aqua Plus with $N_2$-purging | 12 | 60 | 8 |
| Rad Aqua Plus with $N_2$-purging and water trap | 12 | 240 | 3 |

The Rad $H_2O$ procedure is suitable for groundwater analyses, but its detection limit is in most cases too high to analyze surface water (typical detection limits achieved for the two standard techniques compared to improvements discussed below are listed in Table 1). In contrast, the Rad Aqua technique is in principle suitable for the detection of groundwater exfiltration into surface waters, but due to the long equilibration times, the measurements in the field are very time consuming.

The Rad Aqua method has been used for continuous radon detection in coastal environments in order to investigate submarine groundwater discharge (Burnett et al., 2001; Lambert and Burnett, 2003). In this application, at least 20 min are required to reach equilibrium and up to several hours for the subsequent counting to obtain an acceptable uncertainty, thus a long time has to be spent at each sampling point. In order to be able to measure depth profiles in a reasonable time, several measurement systems have to be run in parallel (Dulaiova et al., 2005). Therefore we searched for an alternative way that provides simple and quick sampling in the field and measurement with adequate precision in the laboratory.

We developed a new setup for the sample preparation, which we call Rad Aqua Plus, that combines the advantages of the two standard techniques for our purpose. The method involves discrete samples as in Rad $H_2O$, which can be measured sequentially in the laboratory with only one detector. The required precision can be reached within reasonable counting times. However, larger (12 l) samples are used, which are taken in low-cost closed polypropylene buckets (diameter 26 cm, height 22.7 cm). The gas extraction from the samples is accomplished by an equilibration procedure as in Rad Aqua, but based on two closed loops (Fig. 1), one for the gas- and the other for the water circulation. The gas loop is connected to the alpha-detector RAD7 and the water circulation to the sample bucket. The water from the closed, air-free bucket is sprayed in a gas-exchange cylinder, in which the gas of the closed gas loop is pumped in counter current. Both loops are driven by pumps. The internal pump of RAD7 with a flow rate of about 1 L min$^{-1}$ is used for the gas loop. The water cycle is driven by a submersible pump (Barwig, 12V) with a flow rate of about 1.5 L min$^{-1}$.

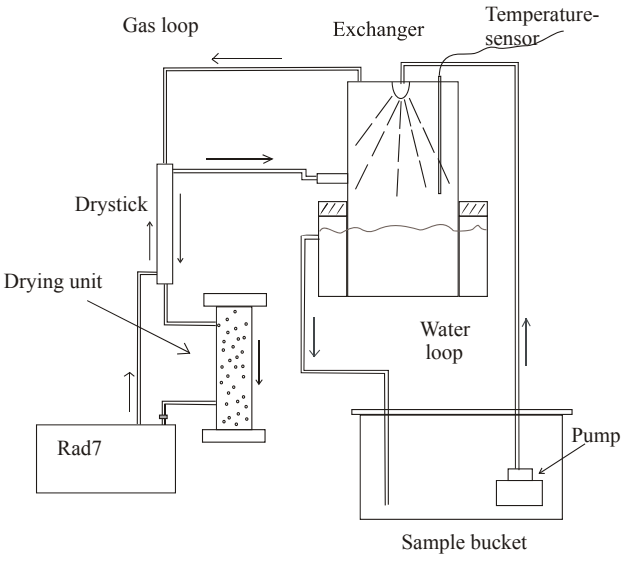



**Fig. 1.** Schematic diagram of the setup consisting of closed water and gas loops, interconnected at the exchanger, where radon is transferred from the water to the gas cycle until equilibrium is reached. The equilibrium partitioning depends on temperature and the volume ratio of the two loops. The radon activity is measured in the gas loop with an alpha spectrometer (RAD7).

In order to enable the analysis of low radon activities, special care was taken to reduce the background counting rate. The background caused by the ambient air in the gas loop is unimportant in case of high radon concentrations or the continuous measurement system. In the continuous method, the initial radon activity from the air is gradually removed by equilibration with the through flowing water. In a system with a closed water- and air cycle it is not negligible if the water activity is below 50 Bq m$^{-3}$. Typical air activities in the Rhine Valley (Heidelberg) are 2 to 30 Bq m$^{-3}$ (Levin et al., 2002). Thus, in order to get rid of the air background and to prepare a well defined initial state, the whole system is purged with Rn-free nitrogen. We found that the background can be substantially reduced by flushing the air loop with nitrogen for 10 min prior to each analysis.

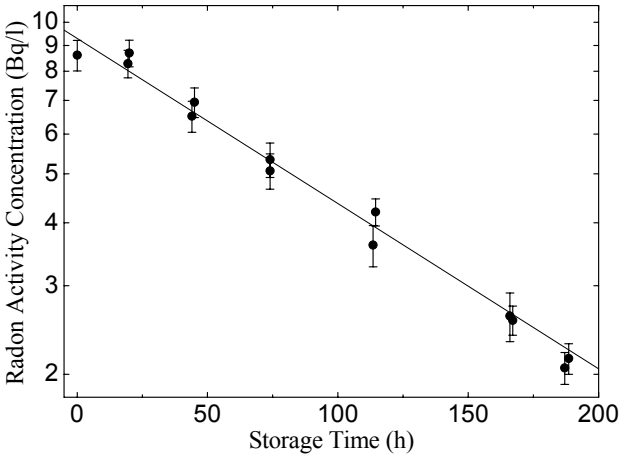



**Fig. 2.** Test of the tightness of the sampling buckets. Several buckets were filled with the same water and then stored. The results are compared to the decay curve.

Furthermore, we observed that the desiccant used to remove humidity from the air loop caused a significant background. In a closed loop containing the $CaSO_4$-drying unit and the RAD7, the radon signal increased linearly with time up to some tens of counts per hour, even after intensive flushing of the whole system with nitrogen (Kluge, 2005). Using a cold trap instead of the standard drying unit and purging with nitrogen reduces the background to a value of 1.2 to 1.5 counts per hour in the energy window of $^{218}$Po. Detection limits achieved with these modifications are listed in Table 1. For a typical 4 h measurement with the Rad Aqua Plus method a detection limit of 3 Bq m$^{-3}$ is obtained.

At the beginning of the measurement procedure, the gas cycle contains no radon. Then outgassing starts. This process continues until solubility equilibrium between the two phases is reached. Depending on the volume of the gas loop and the water flow rate through the exchange chamber, the equilibration takes 30 min up to several hours (Burnett et al., 2001). In our setup, equilibrium is reached after 40 min at a flow rate of 1.5 l min$^{-1}$.

The radon extracted from the water samples is measured with the RAD7 radon monitor as a gas phase activity concentration $C_{\mathrm{gas}}$, which has to be converted to the activity concentration in the water $C_{\mathrm{H_2O}}$, as follows:

$$C_{\mathrm{H_2O}} = C_{\mathrm{gas}} \left( \frac{V_{\mathrm{gas}}}{V_{\mathrm{H_2O}}} + \alpha(T) \right) \quad (1)$$

where $V_{\mathrm{gas}}$ and $V_{\mathrm{H_2O}}$ are the volumes of the closed air and water loops, respectively, and $\alpha(T)$ is the temperature dependent partition coefficient (or Ostwald solubility) of radon. An equation for $\alpha(T)$ is given by Weigel (1978):

$$\alpha(T) = 0.105 + 0.405 \times e^{-0.0502 \times T} \quad (T \text{ in } ^\circ\mathrm{C}). \quad (2)$$

At room temperature, $\alpha$ is about 0.25, indicating that the volumetric concentration in the gas phase is 4 times higher than in the water phase.

The buckets were filled on the lake by lowering a submersible pump to the sampling depth and pumping the water up through a metal tube system. The tubes that are originally designed for $SF_6$-sampling provide an exact depth determination and prevent contamination (von Rohden and Ilmberger, 2001). The pump (Comet-Combi 12-4 T) is driven by 12 V batteries and provides a flow rate of 5 to 10 l per minute. The water from the sampling depth is first used to fill a large barrel, then a bucket is slowly filled brimful. The full bucket and its lid are immersed in the water of the barrel and further flushed for a few minutes before the sample container is closed without leaving any air bubbles.

To test the Rn-tightness of the 12 l buckets used for sampling, a series of water samples from the same source was stored for several days. One sample was measured immediately to determine the initial activity concentration of the water. Afterwards every day a sample was measured and compared to the expected value according to the decay curve. The test shows that the measured radon activity corresponds to the expected value for at least 7 d (Fig. 2).

In applications of radon in lakes it is necessary to differentiate between the radon entering through the sediment boundary and the radon constantly being produced in the water column by dissolved $^{226}$Ra. Often calculations only refer to the radon that is not supported by dissolved radium, the so-called excess radon (Schwartz, 2001). The contribution of the decay of dissolved radium to the radon concentration can be determined by measuring the samples a second time after a certain storage period. After 4 weeks the excess-radon in the lake sample is reduced to a negligible value (0.6% of the initial value) and the measured radon activity is equal to the decay rate of radium (Roether and Kromer, 1978). This equilibration technique allows to determine the radon produced by the decay of dissolved radium at the same precision as the water samples.

Using large samples and radon transfer by equilibration in closed loops it is possible to measure water samples in the low level range. Typical uncertainties are about 10%, depending on the duration of the measurement and the activity concentration. For example, using a counting time of 4 h on a 12 l sample with an activity concentration of 25 Bq m$^{-3}$, 93 counts with a background of 6±2 counts are detected. This leads to an uncertainty of 11%. The advantages of the method are the fast sampling in the field (only a few minutes are necessary to fill the bucket) and the straightforward measurement process in the laboratory.

Compared to the radon analysis method described by Mathieu et al. (1988), radon is not enriched on charcoal and no extensive extraction with vacuum pumps and helium purging is performed. The whole process described by Mathieu et al. (1988) takes about 2 h for extraction and gas transfer from a 19 l water sample, whereas in our procedure the

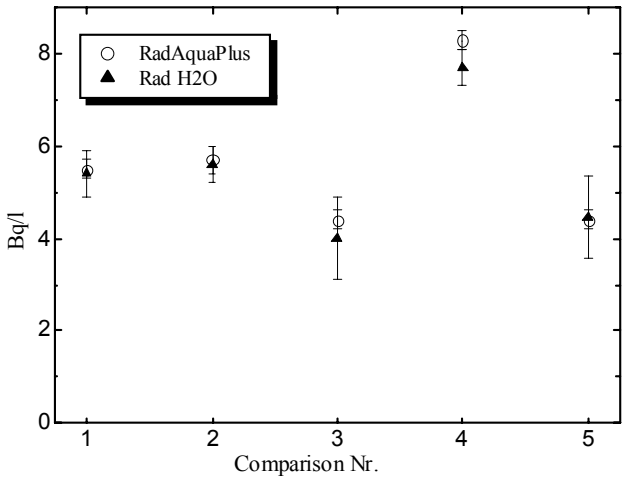



**Fig. 3.** Comparison of the new method, called Rad Aqua Plus, with Rad $H_2O$ measurements at different activites.

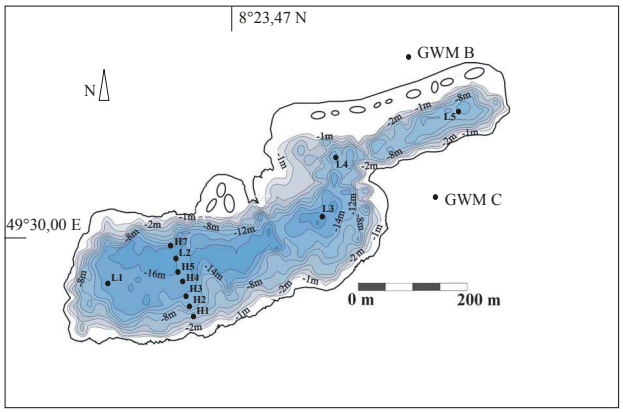



**Fig. 4.** Lake Willersinnweiher, modified from Sandler (2000). The sampling points in the lake are numbered from L1 to L5 in case of the SE to NW longitudinal transect and from H1 to H7 for the transverse transect from South to North. The groundwater wells are GWM B and GWM C. Groundwater enters mainly on the southern lake shore, especially in the south-western part, and leaves the lake on the northeastern shore (Wollschläger et al., 2007).

counting can start after a simple equilibration process of about 40 min. The minimum detectable activity reported by Mathieu et al. (1988) is about 0.7 mBq. The here presented method does not reach this detection limit (about 3 Bq m$^{-3}$, see Table 1). However, for the study of the groundwater coupling in lakes that are strongly influenced by groundwater, as in the example discussed below, the sensitivity is sufficient and is a good tradeoff between applicability and precision.

Figure 3 shows the results of a comparison with Rad $H_2O$ (extraction of 250 ml water samples) as reference method, performed on samples with relatively high activity concentrations. It demonstrates that the two methods agree within uncertainties, but the measurements with Rad Aqua Plus are faster and yield smaller errors for the same counting period due to a higher counting rate. A sample activity concentration of 6.5 Bq l$^{-1}$ leads to 10 cpm in case of Rad $H_2O$ and about 90 cpm in case of Rad Aqua Plus.

## 3 Results

In order to demonstrate the practical applicability of the method to the investigation of groundwater-lake water interactions, a study at a dredging lake, where the groundwater inflow had previously been studied, was conducted. Lake Willersinnweiher is situated in the upper Rhine Valley in Southern Germany. It is one of several dredging lakes in this area. It has a narrow elongated shape (≈200×850 m) with a mean depth of ≈8 m and a maximum depth of 20 m (Fig. 4). Its surface area is 0.145 km$^2$ and its volume is about $1.3\times10^6$ m$^3$. Because there are no surface in- or outflows, the water budget is controlled only by groundwater inflow and subsurface lake water outflow, precipitation and evaporation. In this part of the Rhine Valley precipitation and evaporation are in the same range averaged over one year. We therefore neglect their influence on the water balance.

Lake Willersinnweiher is embedded in about 30 m of young quarternary sandy-gravel sediments (HGK, 1999). Only a thin layer of lake sediment (up to 20 cm in the deepest part) has accumulated since the lake's present morphology was formed in 1975 (Laukenmann, 2002). Groundwater flow is generally from south-west to north-east with typical flow velocities between 0.01 m d$^{-1}$ and at most 0.4 m d$^{-1}$ in the inflow area. In the vicinity of the lake, samples were taken at two groundwater observation wells (GWM B, GWM C, Fig. 4) with a maximum depth of 25 m.

Water from the lake and the observation wells was sampled differently, according to the expected activity concentration. Groundwater in the study area has radon activities of 5 to 10 kBq m$^{-3}$. Therefore, 250 ml glass bottles analyzed with the standard Rad $H_2O$ method are adequate. Because the activity of the lake water is lower (<50 Bq m$^{-3}$), larger sample volumes are necessary. In this case we used 12 l buckets and the new Rad Aqua Plus measurement procedure.

The groundwater wells were sampled twice (31/01 and 4/03/2005), using three pumps operating simultaneously at different depths. The mean radon activity in the upper 5 m of the aquifer is 5700±100 Bq m$^{-3}$. The concentration 7, respectively 9 m below the water table differs between the two wells (mean at GWM B 5700 Bq m$^{-3}$, at GWM C 6800 Bq m$^{-3}$), perhaps due to heterogeneous deposits. In aquifer depths deeper than the lake bottom, the radon activity in both wells is similar with a mean of 6700±100 Bq m$^{-3}$. In the vicinity of the wells the lake depth is in general less than 8 m. Therefore only the shallow groundwater can enter the

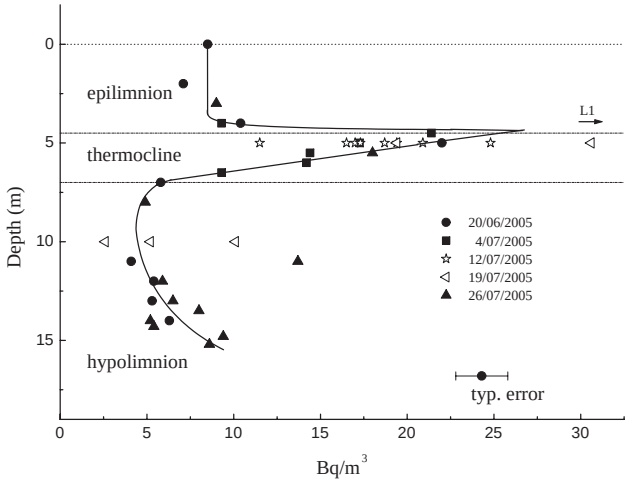



**Fig. 5.** Summary of vertical radon profiles in the middle of Lake Willersinnweiher at station L2 (full symbols) and horizontal transects including all stations in the main basin (open symbols), taken at different dates during the summer stratification period. The result at station L1 (5 m depth, 19/07/2005) is off scale (52.4 Bq m$^{-3}$). The black line is a subjective fit based on the L2 data. The strong radon peak in the upper thermocline and the higher mean radon activity in the epilimnion compared to the hypolimnion indicate a groundwater inflow mainly in the upper part of the lake.

lake and a value of 5700 Bq m$^{-3}$ is used for the inflowing groundwater in the budget calculations discussed below.

Samples for the radon analysis of lake water were taken with a submersed pump at different locations and from different depths. Vertical profiles at the central station L2 in the main lake basin (Fig. 4) were sampled at three days (20/06, 4/07, and 26/07/2005). Horizontal sections along and perpendicular to the lake axis were taken at two other days (12/07 and 19/07/2005). The samplings took place during a period of stable summer stratification. The results of all samples are listed in Table 2 (vertical profiles) and Table 3 (horizontal sections).

The results of the vertical profiles from site L2 (Fig. 5) do not show strong temporal changes within the sampling period and are therefore interpreted together. The vertical resolution of the combined profile is about 0.5 m in the thermocline and towards the lake bottom and between 1 and 3 m in the rest of the water column. The profiles show activity concentrations ranging from 5 to 30 Bq m$^{-3}$. In the epilimnion, including samples from the water surface, concentrations are constant within the uncertainty, at a mean value of 8.9±1.2 Bq m$^{-3}$. In the thermocline the activity is increasing to a maximum of ≈30 Bq m$^{-3}$. In the hypolimnion, the radon concentration is rather low with a mean of 6.1±1.9 Bq m$^{-3}$.

The first horizontal transect of the radon activity was performed from south to north at the widest part of the lake, perpendicular to the lake's main axis. Samples were taken from 5 m depth with a horizontal spacing of the sampling sites of about 20 to 30 m (H1 to H7, see Fig. 4). The second horizontal transect was performed from southwest to northeast along the main axis with samples from 5 and 10 m depth with a horizontal spacing of the sites of about 200 m (L1 to L5). Both sections also include samples from the central station L2, where they intersect.

Results from the sites of the horizontal transects located in the main part of the lake are shown in Fig. 5 along with the profiles from site L2. Sites L4 and L5 in the small northeastern basin are excluded in Fig. 5 because this part of the lake is separated by a sill at about 8 m depth. The radon activities in the small basin are comparatively high, probably due to the proximity of the sampling points to the sediment as well as to possible groundwater inflow along the southern shore of this basin. In the main basin, the results from most of the stations are comparable to those from the central profiles at station L2, although the variability especially at a depth of 5 m is high. The large variation at this depth is probably due to the strong vertical gradients near the thermocline peak of the radon activity. Nevertheless, the highest activity of more than 50 Bq m$^{-3}$ was found at station L1 towards the southwestern end of the lake, indicating that the main groundwater inflow area is located there. The overall groundwater flow direction deduced from a numerical model (Wollschläger, 2003) is in agreement with this result.

The radon activities in Lake Willersinnweiher displayed in Tables 2 and 3 and Fig. 5 are uncorrected total measured activities including the contribution from dissolved radium and excess radon. We refer to total radon activities and take the production from radium explicitly into account in the budget calculations in order to correctly represent the gas exchange with the atmosphere. The activity of dissolved radium was determined by measurements on two lake samples stored for several weeks. The radon activity concentration in these samples was in equilibrium with that of the dissolved radium, and yielded radium activites of about 4 Bq m$^{-3}$ in the epilimnion and 2.6 Bq m$^{-3}$ in the hypolimnion. The representativeness of these few measurements was checked by a high resolution radium profile taken in 2007. This profile showed that the still present radon peak in the thermocline is not supported by dissolved radium, as the radium values were in general smaller than 2 Bq m$^{-3}$.

For the estimation of the gas exchange flux, the atmospheric radon activity was measured. A 10 l air sample was taken by enriching the radon on a dry ice cooled charcoal trap. By heating the charcoal in the laboratory, the radon was purged into a closed loop with the RAD7, where the activity was measured to be 7.4±0.8 Bq m$^{-3}$, which is in the range of typical air activities in the Rhine Valley (Levin et al., 2002).

**Table 2.** Results of vertical profile measurements in Lake Willersinnweiher using the equilibration method Rad Aqua Plus. The samples were taken at the indicated dates and depths at station L2 (middle of the main basin, see Fig. 4). Typical uncertainties are 10 to 15% of the value.

| 20/06/2005 | | 04/07/2005 | | 26/07/2005 | |
|---|---|---|---|---|---|
| depth (m) | $C_{Rn}$ (Bq m$^{-3}$) | depth (m) | $C_{Rn}$ (Bq m$^{-3}$) | depth (m) | $C_{Rn}$ (Bq m$^{-3}$) |
| 0 | 8.5 | 4 | 9.3 | 3 | 9.0 |
| 2 | 7.1 | 4.5 | 21.4 | 5.5 | 18.0 |
| 4 | 10.4 | 5 | 17.3 | 8 | 4.9 |
| 5 | 22.0 | 5.5 | 14.4 | 11 | 13.7 |
| 7 | 5.8 | 6 | 14.2 | 12 | 5.9 |
| 11 | 4.1 | 6.5 | 9.3 | 13 | 6.5 |
| 12 | 5.4 | | | 13.5 | 8.0 |
| 13 | 5.3 | | | 14 | 6.2 |
| 14 | 6.5 | | | 14.3 | 5.4 |
| 16.8 | 24.3 | | | 14.8 | 9.4 |
| | | | | 15.2 | 8.6 |

**Table 3.** Results of horizontal sections of radon measurements through Lake Willersinnweiher using the equilibration method Rad Aqua Plus. The samples were taken on 12/07/2005 in a transversal section (stations H1 to H7, see Fig. 4) at a depth of 5 m (4.6 m for H1) and on 19/07/2005 in a longitudinal section (stations L1 to L5) at depths of 5 and 10 m. Typical uncertainties are 10 to 15% of the value.

| 12/07/2005 | | 19/07/2005 | | |
|---|---|---|---|---|
| 5 m depth | | 5 m depth | | 10 m depth |
| station | $C_{Rn}$ (Bq m$^{-3}$) | station | $C_{Rn}$ (Bq m$^{-3}$) | $C_{Rn}$ (Bq m$^{-3}$) |
| H1[a] | 24.0 | L1 | 52.4 | 2.6 |
| H2 | 14.7 | L2 | 30.6 | 10.1 |
| H3 | 16.2 | L3 | 19.4 | 5.2 |
| H4 | 10.4 | L4 | 13.8 | 24.8 |
| H5 | 14.4 | L5 | 48.0 | 21.0 |
| L2 | 19.4 | | | |
| H7 | 16.9 | | | |

[a] the sample at station H1 was taken at 4.6 m depth

## 4 Discussion

The basic idea for the calculation of the groundwater inflow is to use the mass balance for the radon isotope (see Fig. 6). If the entire lake is homogeneously mixed and radon activity is at steady state, only the following quantities have to be determined: The mean radon activity concentration (Bq m$^{-3}$) of the lake water $C_{la}$, a representative activity concentration of the inflowing groundwater $C_{gw}$, the radium activity concentration (Bq m$^{-3}$) of the lake water $C_{Ra}$, a representative value for the sediment flux $F_{\text{sed}}$, and the gas exchange flux of radon from the lake to the atmosphere $F_{\text{surf}}$.

The variation in the lake level during the sampling campaign was in the cm range, so the lake volume as well as the exchange with the groundwater can be taken as constant. As the Rn activity concentrations in distinct water depths do not change in time (see Fig. 5) it seems reasonable to assume steady state (i.e., $dC_{la}/dt$=0). Then the radon balance can be written (compare Corbett et al., 1997):

$$Q_{gw}(C_{gw} - C_{la}) + F_{\text{sed}}A_{\text{sed}} - F_{\text{surf}}A_{\text{surf}} - \lambda_{\text{Rn}}V(C_{la} - C_{Ra}) = 0 \quad (3)$$

$Q_{gw}$ is the groundwater in- and outflow in m$^3$ d$^{-1}$, $A_{\text{surf}}$ the lake surface, $A_{\text{sed}}$ the sediment surface area and $V$ the total volume of the lake. In case of lake Willersinnweiher, groundwater in- and outflow is assumed to be the same, because there is no surface in- and outflow and furthermore precipitation and evaporation are comparable.

Lake Willersinnweiher is stratified during the summer period. Therefore we can divide the lake into different parts (see Fig. 6). The well mixed upper layer (epilimnion), which extends to a depth of 4.5 m, the region of the strong tempera-

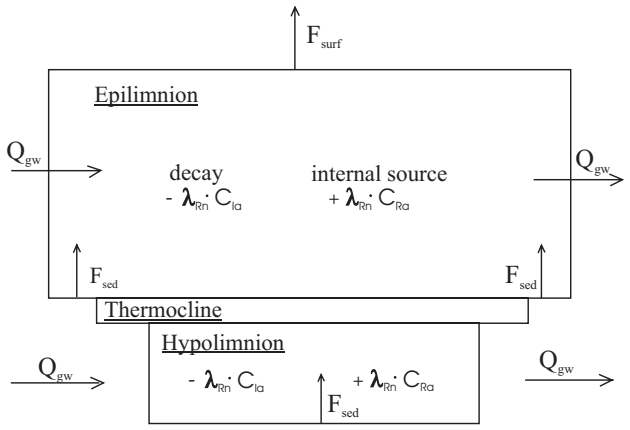



**Fig. 6.** Box model of the stratified lake. Radon sources are groundwater inflow $Q_{gw}$, sediment flux $F_{\mathrm{sed}}$ and the decay of dissolved radium in the lake water ($\lambda_{\mathrm{Rn}} \times C_{Ra}$). The radon concentration is decreased by decay ($\lambda_{\mathrm{Rn}} \times C_{la}$), outgassing to the atmosphere $F_{\mathrm{surf}}$ and the outflow. The radon exchange between the two boxes is neglected, because of the stratification and the reduced transport through the thermocline.

ture change (thermocline) between 4.5 m and 7 m below the surface and the deepest layer (hypolimnion), which reaches from a depth of 7 m to the bottom. As the radon transport through the thermocline is strongly reduced, we neglect the radon fluxes from the epilimnion through the thermocline to the hypolimnion and vice versa. Therefore each layer can be regarded as a box, which does not exchange radon with the other parts. Furthermore, in the hypolimnion and the thermocline the gas exchange with the atmosphere does not have to be taken into account. Thus the radon balance (Eq. 3) for these boxes can be simplified, the gas exchange has to be included only in the epilimnion. For $C_{la}$, $A_{\mathrm{sed}}$ and $V$ the corresponding values for the respective boxes have to be used (Table 4). Calculating the mass balance for the hypolimnion and the epilimnion separately, vertical differences in the groundwater inflow can be detected.

In the hypolimnion below 7 m depth no major groundwater inflow appears to be present, as the radon concentration is rather low (Fig. 5). The increase of activity towards the lake bottom could be due to a small, localized groundwater inflow, but more likely it is produced by the sediment flux which is most important in the bottom layer with a large ratio of sediment area to water volume. In general, radium decay, groundwater inflow and sediment flux are the sources for the hypolimnic radon. In order to estimate the maximum possible groundwater inflow, we consider the limiting case of negligible sediment flux. Balancing the groundwater inflow and the decay terms yields a maximum groundwater inflow of $Q_{gw}$=(52±30) $\mathrm{m^3\,d^{-1}}$, which is small compared to the total volume of the hypolimnion (470 000 $\mathrm{m^3}$). Even with this maximum groundwater inflow, the hypolimnion water mass would be renewed only once in about 25 a, in the absence of the winterly overturn.

The constant radon concentrations in the epilimnion show that the upper 4.5 m of the lake's water column are well mixed. In contrast, vertical mixing is strongly suppressed in the thermocline between about 4.5 and 7 m depth (von Rohden et al., 2007). A strong radon peak, representing a clear groundwater signal, is present in the upper thermocline (Fig. 5). As there is a minimum of the sediment surface to water volume ratio at this depth, we do not ascribe this to the morphology of the lake in conjunction with the sediment flux.

For a first estimation, we interpret the peak as a result of a uniform radon input due to groundwater inflow into the upper part of the lake. This signal is preserved in the thermocline but strongly reduced in the epilimnion due to gas exchange. In other words, we assume that if there was no gas exchange, the radon concentration would be 22 $\mathrm{Bq\,m^{-3}}$ (mean value at L2 in 5 m depth) throughout the epilimnion and the thermocline. Using this value and neglecting the sediment flux, the balance calculation leads to a maximum value for the groundwater inflow of (475±140) $\mathrm{m^3\,d^{-1}}$ for the upper part of the lake (<7 m depth, volume 830 000 $\mathrm{m^3}$).

Combining the above first estimates for the hypolimnion and the epilimnion, the total groundwater inflow to the lake is estimated to (530±140) $\mathrm{m^3\,d^{-1}}$, which corresponds to (1350±350) $\mathrm{mm\,a^{-1}}$ and a replacement time between 5.2 and 9.1 a. This maximum estimate is close to the lower end of the results of former studies: A calculation of the inflow with a groundwater flow model based on piezometer data and stable isotopes (Wollschläger, 2003) yielded a range of 446 to 1726 $\mathrm{m^3\,d^{-1}}$ (replacement time: 2 to 8 a). Balancing of the calcite sedimentation (Schmid, 2002) resulted in 630 to 1060 $\mathrm{m^3\,d^{-1}}$ (3 to 5 a).

The actual radon activity in the epilimnion above 4.5 m depth is much lower than the maximum in the thermocline. Assuming that the difference is entirely due to outgassing to the atmosphere, the gas exchange flux can be calculated, and using the difference between the radon activity at the surface (8.9±1.2 $\mathrm{Bq\,m^{-3}}$) and the equilibrium concentration with the air (≈1.8 $\mathrm{Bq\,m^{-3}}$), the gas transfer velocity can be computed. This calculation performed for the water body above 4.5 m depth (volume 580 000 $\mathrm{m^3}$) results in a transfer velocity $k$ for Rn of 1.3 $\mathrm{m\,d^{-1}}$, which is a rather high value compared to a value of $k$=0.6 $\mathrm{m\,d^{-1}}$ calculated from wind data (mean wind speed is around 2 $\mathrm{m\,s^{-1}}$) using a relationship given by Upstill-Goddard et al. (1990) or $k$=0.3 $\mathrm{m\,d^{-1}}$ according to a relationship of Wanninkhof (1992).

This result indicates that the actual radon input into the epilimnion is likely smaller than estimated above, probably because the assumption of a uniform inflow into epilimnion and thermocline is oversimplified. A reason for a comparatively high apparent input into the thermocline could be that groundwater entering the lake above or below the thermo-

**Table 4.** Values used for the balance calculations. $A_{\text{sed}}$ is the sediment surface in each box. The according mean radon activity is $C_{la}$. Throughout the thermocline a mean radon activity concentration of 22 (Bq m$^{-3}$) was assumed. The groundwater inflow rate $Q_{gw}$ is calculated neglecting the sediment flux. Therefore the results represent an upper limit.

| Box | Volume (m$^3$) | $A_{\text{sed}}$ (m$^2$) | $C_{la}$ (Bq m$^{-3}$) | $Q_{gw}$ (m$^3$/d) |
|---|---|---|---|---|
| Epilimnion | 580 000 | 31 000 | 8.9 | 330 |
| Thermocline | 250 000 | 27 500 | 22 | 145 |
| Hypolimnion | 470 000 | 86 000 | 6.1 | 52 |
| Total | 1 300 000 | 144 500 | – | 527 |

cline could sink or rise to the layer where the probability of achieving neutral buoyancy is largest.

The above calculations rely on several assumptions. An essential requirement is a complete horizontal mixing of the lake on a time scale smaller than the half-life of $^{222}$Rn, admitting the assumption of horizontal homogeneity so that the central profile can be taken as representative for the entire lake. Peeters et al. (1996) reported horizontal eddy diffusivities in lakes in the range of 0.02 to 0.3 m$^2$ s$^{-1}$. Using these values and a typical horizontal dimension of 100 m (half width) for Lake Willersinnweiher, the mixing timescale lies between 5 and 70 h. This is comparable to the radon half-life of 92 h, hence the requirement of horizontal homogeneity is at best approximately fulfilled. In fact, the horizontal sections revealed inhomogeneity especially in the epilimnion (see Fig. 5 and Table 3). The activity is decreasing from the suspected groundwater entry zone (south-west) to the north-eastern end of the main basin of the lake.

The requirement of a good horizontal mixing will limit the application of the simple box model concept based on only one central radon profile to rather small lakes. In case of larger lakes, the horizontal mixing time will be in the range of days or weeks, which in view of the short half-life of radon must lead to horizontal concentration gradients. In this case it is not possible to calculate the groundwater inflow from one vertical profile, but a three dimensional mapping of the radon distribution would be required. This has, on the other hand, the advantage that the groundwater entry areas can be located.

A large uncertainty originates from the scatter of the activity concentration in the thermocline. The values measured at 5 m depth at station L2 vary from 19 to 30 Bq m$^{-3}$, possibly due to horizontal inhomogeneity and a variable degree of advection of groundwater influenced water. Further uncertainties arise from the assumptions about the distribution of the groundwater inflow in the upper part of the lake and the effect of gas exchange. The sharp peak in the thermocline is possibly not only the result of the gas exchange at the surface, but also due to the buoyancy driven interstratification of the inflowing groundwater in the thermocline.

A way to reduce the uncertainty of the effect of gas exchange would be to monitor the temporal evolution of the radon concentration in the epilimnion and the radon inventory in the entire lake over an extended period of time, as demonstrated by Corbett et al. (1997). Seasonal changes in the vertical stratification of the lake should lead to a dynamic evolution of the radon concentrations that can be interpreted in terms of gas exchange, vertical mixing, and groundwater input. The analytical method presented here is suitable to conduct such an extended study, which we plan to realize in the future.

## 5 Conclusions

The method using large buckets for sampling, radon transfer by equilibration between water- and gas cycles, and the measurement with an energy sensitive alpha spectrometer is well suited for the detection of the radon activities in lakes which are influenced by groundwater. The method involves rapid sampling, followed by precise radon measurements in the laboratory. The presented field example from Lake Willersinnweiher shows that the approach allows the detection and quantitative estimation of groundwater inflow to lakes.

The groundwater inflow estimated by a simple radon balance is comparable to the results of former studies. A significant advantage of the radon method is that it allows to distinguish groundwater inflow to the hypolimnion and the epilimnion during stratification, and to locate groundwater entry areas in the horizontal dimension. However, more data over a longer time period with changing vertical stratification and better horizontal resolution would be needed to reduce the uncertainties of the inflow estimation and to obtain a more detailed picture of the spatial distribution of the groundwater inflow.

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## References


Burnett, W. C., Kim, G., and Lane-Smith, D.: A continuous monitor for assessment of $^{222}$Rn in the coastal ocean, J. Radioanal. Nucl. Chem., 249, 167–172, 2001.

Cable, J. E., Bugna, G. C., Burnett, W. C., and Chanton, J. P.: Application of $^{222}$Rn and $CH_4$ for assessment of groundwater discharge to the coastal ocean, Limnol. Oceanogr., 41, 1347–1353, 1996.

Colman, J. A. and Armstrong, D. E.: Vertical eddy diffusivity determined with $^{222}$Rn in the benthic boundary layer of ice-covered lakes, Limnol. Oceanogr., 32, 577–590, 1987.

Corbett, R., Burnett, W. C., Cable, P. H., and Clark, S. B.: Radon tracing of groundwater input into Par Pond, Savannah River Site, J. Hydrol., 203, 209–227, 1997.

Corbett, R., Dillon, K., Burnett, W. C., Chanton, J., and Rutkowki, C.: Patterns of groundwater discharge into Florida Bay, Limnol. Oceanogr., 44, 1045–1055, 1999.

Corbett, R., Dillon, K., Burnett, W. C., and Chanton, J.: Estimating the groundwater contribution into Florida Bay via natural tracers, Rn-222 and CH-4, Limnol. Oceanogr., 45, 1546–1557, 2000.

Crusius, J., Koopmans, D., Bratton, J. F., Charette, M. A., Kroeger, K. D., Henderson, P., Ryckman, L., Halloran, K., and Colman, J. A.: Submarine groundwater discharge to a small estuary estimated from radon and salinity measurements and a box model, Biogeosciences, 2, 141–157, 2005, http://www.biogeosciences.net/2/141/2005/.

Dulaiova, H., Peterson, R., Burnett, W. C., and Lane-Smith, D.: A multi-detector continuous monitor for assessment of $^{222}$Rn in the coastal ocean, J. Radioanal. Nucl. Chem., 263, 361–365, 2005.

Durridge Co.: RAD7 radon detector – Owner's manual, available at http://www.durridge.com/Manuals.htm, 2000.

Durridge Co.: RAD7 RAD-$H_20$ – Radon in water accessory – Owner's manual, available at http://www.durridge.com/Manuals.htm, 2001a.

Durridge Co.: RAD AQUA – Continuous radon-in-water monitoring system, available at http://www.durridge.com/Manuals.htm, 2001b.

Hoehn, E. and von Gunten, H.: Radon in groundwater: a tool to assess infiltration from surface waters to aquifers, Water Resour. Res., 25, 1795–1803, 1989.

HGK, Arbeitsgruppe Hydrogeologische Kartierung und Grundwasserbewirtschaftung Rhein-Neckar-Raum: Hydrogeologische Kartierung und Grundwasserbewirtschaftung Rhein-Neckar-Raum, Analyse des Ist-Zustandes, Technical report, Ministerium für Ernährung, Landwirtschaft, Umwelt und Forsten Baden-Württemberg, Hessischer Minister für Landesentwicklung, Umwelt, Landwirtschaft und Forsten, Ministerium für Landwirtschaft, Weinbau und Forsten Rheinland-Pfalz, 1999.

Imboden, D. and Emerson, S.: Natural radon and phosphorus as limnologic tracers: Horizontal and vertical eddy diffusion in Greifensee, Limnol. Oceanogr., 29, 831–844, 1984.

Kawabata, H., Narita, H., Harada, K., Tsunogai, S., and Kusakabe, M.: Air-sea gas transfer velocity in stormy winter estimated from radon deficiency, J. Oceanogr., 59, 651–661, 2003.

Kluge, T.: Radon als Tracer in aquatischen Systemen, Diploma thesis, Univ. Heidelberg, 2005.

Krabbenhoft, D. P., Bowser, C. J., Anderson, M. P., and Valley, J. W.: Estimating groundwater exchange with lakes 1. The stable isotope mass balance method, Water Resour. Res., 26, 2445–2453, 1990.

Lambert, M. J. and Burnett, W. C.: Submarine groundwater discharge estimates at a Florida coastal site based on continuous radon measurements, Biogeochem., 66, 55–73, 2003.

Laukenmann, S.: Transport und Austausch redoxsensitiver Elemente zwischen Freiwasser und Sediment in einem eutrophen Hartwassersee (Willersinnweiher/Ludwigshafen), unter besonderer Berücksichtigung des geochemischen Verhaltens von Uran, Ph.D. thesis., Univ. Heidelberg, 2002.

Lee, D. R.: A device for measuring seepage flux in lakes and estuaries, Limnol. Oceanogr., 22, 140–147, 1977.

Levin, I., Born, M., Cuntz, M., Langendörfer, U., Mantsch, S., Naegler, T., Schmidt, M., Verlagin, A., Verclas S., and Wagenbach, D.: Observations of atmospheric variability and soil exhalation rate of radon-222 at a Russian forest site, Tellus, 54B, 462–475, 2002.

Mathieu, G. G., Biscaye, P. E., and Lupton, R. A.: System for measurement of Rn-222 at low levels in natural waters, Health Phys., 55, 982–992, 1988.

Peeters, F., Wüest, A., Piepke, G., and Imboden, D.: Horizontal mixing in lakes, J. Geophys. Res., 101, 18 361–18 375, 1996.

Roether, W. and Kromer, B.: Field determination of air-sea gas exchange by continuous measurement of radon-222, Pure Appl. Geophys., 116, 476–485, 1978.

Sandler, B.: Die Wirkung von Sanierungs- und Restaurierungsmaßnahmen auf die Nährstoffströme und die biotische Dynamik eines anthropogenen Gewässers am Beispiel des Willersinnweihers/Ludwigshafen, Ph.D. thesis, Univ. Heidelberg, 2000.

Schmid, J.: Calcitfällung und Phosphor-Kopräzipitation im Phosphorhaushalt eines eutrophen Hartwassersees mit anoxischem Hypolimnion (Willersinnweiher, Ludwigshafen am Rhein), Ph.D. thesis., Univ. Heidelberg, 2002.

Schubert, M., Knoeller, K., Treutler, H.-C., Weiss, H., Dehnert, J.: $^{222}$Rn as a tracer for the estimation of infiltration of surface waters into aquifers, in: Radionuclides in the environment, edited by: Povinec, P. P. and Sanchez-Cabeza, J. A., Radioactivity in the environment, Vol. 8, Elsevier, Amsterdam, 326–334, 2006.

Schwartz, M. C.: Significant groundwater input to a coastal plain estuary: assessment from excess radon, Estuar. Coast. Shelf S., 56, 31–42, 2003.

Trettin, R., Gläßer, W., Lerche, I., Seelig, U., and Treutler, H.-C.: Flooding of lignite mines: isotope variations and processes in a system influenced by saline groundwater, Isot. Environ. Health S., 42, 159–179, 2006.

Tuccimei, P., Salvati, R., Capelli, G., Delitala, M. C., and Primavera, P.: Groundwater fluxes into a submerged sinkhole area, Central Italy, using radon and water chemistry, Appl. Geochem., 20, 1831–1847, 2005.

Upstill-Goddard, R. C., Watson, A. J., Liss, P. S., and Liddicoat, M.: Gas transfer velocities in lakes measured with $SF_6$, Tellus, 42B, 364–377, 1990.

von Rohden, C. and Ilmberger, J.: Tracer experiment with sulfur hexafluoride to quantify the vertical transport in a meromictic pit lake, Aquat. Sci., 63, 417–431, 2001.

von Rohden, C., Wunderle, K., and Ilmberger, J.: Parameterisation of the vertical transport in a small thermally stratified lake, Aquat. Sci., 69, 129–137, doi:10.1007/s00027-006-0827-4, 2007.

Wanninkhof, R.: Relationship Between Wind Speed and Gas Exchange Over the Ocean, J. Geophys. Res., 97, 7373–7382, 1992.
Weigel, F.: Radon, Chemiker-Zeitung, 102, 287–298, 1978.
Wollschläger, U.: Kopplung zwischen Oberflächenwasser und Grundwasser: Modellierung und Analyse von Umwelttracern, Ph.D. thesis., Univ. Heidelberg, 2003.
Wollschläger, U., Ilmberger, J., Isenbeck-Schröter, M., Kreuzer, A., von Rhoden, C., Roth, K. and Schäfer, W.: Coupling of groundwater and surface water at Lake Willersinnweiher: Groundwater modeling and tracer studies, Aquat. Sci., 69, 138–152, doi:10.1007/s00027-006-0825-6, 2007.
Yehdegho, B., Rozanski, K., Zojer, H., and Stichler, W.: Interaction of dredging lakes with the adjacent groundwater field: an isotope study, J. Hydrol., 192, 247–270, 1997.