

Characterization of groundwater–lake water interactions at Pyhäjärvi, a lake in SW Finland

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This paper presents the first attempt in Finland to collect detailed observations on surface water and groundwater interactions. We focused on a 10-km northeastern shoreline of Pyhäjärvi, where the potential groundwater discharge areas are associated with the esker aquifers. The methods used during 2008 and 2009 involved winter mapping, lake water and lake bed sediment temperature measurements, pH and electrical conductivity measurements, seepage meters, mini-piezometers, and chemical (PCE/TCE) and isotopic tracers. The main aims of this study were to compare the applicability of the methods used and evaluate the results obtained. The results from the various methods correlated and confirmed the groundwater discharge ($4.69 \times 10^{-5} \text{ cm s}^{-1}$ to $4.80 \times 10^{-3} \text{ cm s}^{-1}$) into lake water at the shoreline. Further research is needed to obtain a quantitative estimation of groundwater seepage in Pyhäjärvi.

Introduction

Management of water resources in Finland has traditionally focused on either surface water or groundwater, and interactions between the groundwater and surface water have not been systematically studied. However, it is apparent that nearly all surface water features (lakes, rivers, wetlands) interact with groundwater (Winter *et al.* 1998). Management of one component of the hydrologic system is usually only partially effective, because each hydrologic component is in continuing interaction with other components, and development or contamination of one commonly affects the others (Winter *et al.* 1998). However, some estimations about groundwater component has been used in some river water balance models recently in Finland (e.g. Grizzetti *et al.* 2003, Tattari *et al.* 2009).

Lakes interact with aquifers in three basic ways: some receive groundwater inflow throughout their entire bed, some recharge groundwater throughout their bed, but the majority of lakes probably receive groundwater inflow through part of their bed and recharge groundwater through the other parts (Winter *et al.* 1998). Some lakes may have no interaction with the underlying aquifer. The seepage meter approach (Lee 1977) provided information on both the spatial variability and distribution of seepage flux rates and amounts into lakes (McBride and Plannkuch 1975, Brock *et al.* 1982, Woessner and Sullivan 1984, Mitchell *et al.* 1988, Schafran and Driscoll 1990, Shaw and Prepas 1990a, Schneider *et al.* 2005). In addition to spatial variability, groundwater flow rates vary temporally (Downing and Peterka 1978, Connor and Belanger 1981, Kenoyer and Anderson 1989,

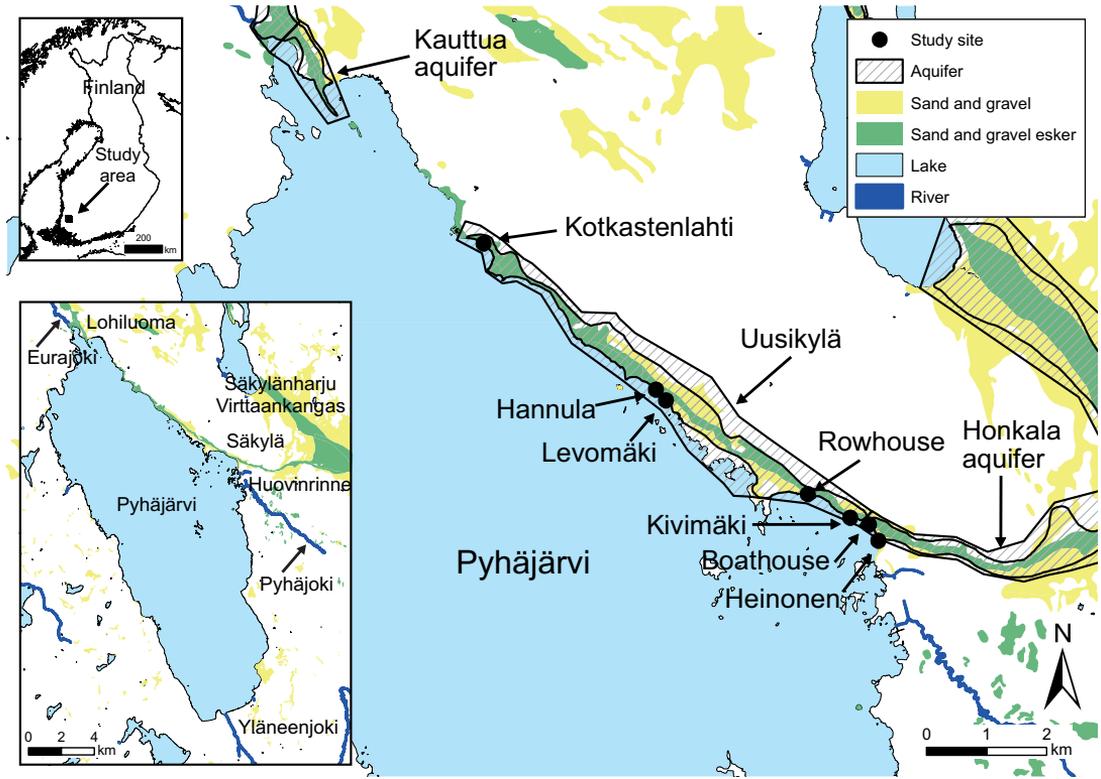


Fig. 1. Location of the study sites; esker deposit and aquifers close to study sites at Pyhäjärvi. (Basemap Database ©National Land Survey of Finland 2007; Quaternary Deposits Database ©Geological Survey of Finland 2008; Groundwater Database ©SYKE 2009).

Shaw and Prepas 1990a, Sebestyen and Schneider 2001). Large lakes are more likely than small lakes to exhibit variation in shoreline substrates, aquifer characteristics, topography or meteorological conditions that influence groundwater seepage processes (Schneider *et al.* 2005).

Pyhäjärvi provides an ideal, large lake system in which it is possible to examine spatial patterns in shoreline groundwater seepage. We focused on a 10-km northeastern shoreline at Pyhäjärvi, because the potential groundwater discharge areas are associated with the esker aquifers (Fig. 1). Pyhäjärvi has been intensively studied for decades and was the object of comprehensive restoration activities both in the catchment and in the lake since the 1990s (Ventelä *et al.* 2007, 2011). However, the previous studies made a general assumption that lake water is derived almost solely from precipitation and surface runoff, with only a minor proportion of groundwater discharge to Pyhäjärvi (e.g. Ekholm *et al.*

1997, Krogurus and Ekholm 2003, Lepistö *et al.* 2008, Ventelä *et al.* 2011).

The goal of this study was to compare the applicability of the methods used and evaluate the results obtained at Pyhäjärvi. The more specific objectives were to (1) identify the locations where groundwater discharges to surface water, (2) measure the directions and rates of groundwater seepage with mini-piezometers and seepage meters, and (3) examine the factors that influence groundwater seepage to Pyhäjärvi.

Research area

Pyhäjärvi (60°54'–61°06'N, 22°09'–22°25'E) is the largest lake in southwestern Finland (Fig. 1). This mesotrophic lake has a surface area of approx. 155 km² and a perimeter length of 88 km. Its drainage basin area is 616 km², which is quite a small area as compared with the

surface area of the lake. Pyhäjärvi is shallow; the mean depth of the lake is 5.5 m. The only depression in the lake is 26 m deep and is located on the west side of the lake. The Yläneenjoki, Pyhäjoki and Eurajoki are the three largest rivers in the drainage basin area (Fig. 1). Two major rivers, Yläneenjoki and Pyhäjoki, discharge into Pyhäjärvi. The Eurajoki, on the north side of the lake, forms the only outflow from the lake.

Pyhäjärvi is used for recreational activities and a commercial fishery, as well as local industrial processes (Ventelä *et al.* 2007). During recent decades, all these activities have become seriously threatened by eutrophication which is associated with high levels of nutrient loading from the catchment (Ventelä *et al.* 2007). Pyhäjärvi has been the target of an intensive restoration programme since 1995, when the Pyhäjärvi Protection Fund (PPF) was created (Mattila *et al.* 2001, Ventelä *et al.* 2001, 2002, Ventelä and Lathrop 2005).

The water intake plant of Lohiluoma in the Kauttua aquifer is located in the northernmost corner of Pyhäjärvi (Fig. 1). Some of the water pumped from the Lohiluoma ($3000 \text{ m}^3 \text{ d}^{-1}$) is artificially recharged, taken from Pyhäjärvi and sprinkled on the top of the aquifer; some is Pyhäjärvi water from bank infiltration; and some is natural groundwater flowing from the north. In the Honkala aquifer the main groundwater flow direction is towards Pyhäjärvi, according to a regional-scale groundwater flow model constructed by Artimo (2002). The water intake plant in Honkala has been closed, due to contamination by tetrachloroethylene (PCE) and trichloroethylene (TCE) from a spill at a dry-cleaning laundry near the Honkala aquifer (Fig. 1) (Artimo 2001).

The glacial sediments of the research area consist of deposits formed during the Late Weichselian deglaciation of the Scandinavian Ice Sheet and include till, sand, gravel, and glacial and postglacial clay deposits (Fig. 1) (Artimo 2002). The Honkala aquifer is composed of esker deposits and is mainly unconfined (Artimo 2002). The Huovinrinne esker is associated with the most prominent glacial landform in southwest Finland, the Säköjärvi–Virtaankangas glaciofluvial complex, referred to as an interlobate esker (Punkari 1980, Saarnisto and Salonen 1995, Mäkinen 2003). The Huovinrinne esker

continues to Uusikylä (Fig. 1) and then to Kauttua along the northeast shore of Pyhäjärvi; this elongated esker is partly under water on the north side of Uusikylä (Fig. 1). All seven study sites are in the shore zone, where the lake is bordered by the esker aquifers. The substrate varies among the seven study sites. The bottom sediments consist of fine sand, silty clay and peat at the Heinonen study site and predominantly of sand and scattered pebbles at the Boathouse, Kivimäki, Levomäki and Hannula study sites (Fig. 1). The substrates are dominated by dense pebbles, cobbles and boulders at the Rowhouse and Kotkastenlahti study sites (Fig. 1). All seven study sites are gently sloping with a slight gradient. The study sites are underlain by sandstone, which is the main rock type on the NE and E shores of Pyhäjärvi.

Methods

Winter mapping of unfrozen and open water shoreline areas

Unfrozen and open water shoreline areas were mapped in March 2009 during one field campaign by skiing on the lake ice along a 15-km stretch of the northeastern shoreline of Pyhäjärvi. The air temperature varied between $-6 \text{ }^\circ\text{C}$ and $-2 \text{ }^\circ\text{C}$ and according to monthly hydrological report of Finnish Environmental Institute (SYKE) the average lake ice thickness was 52 cm on the field campaign. The potential groundwater discharge areas were open water areas without ice cover or the lake ice was considerably weaker and had higher water content than the surrounding lake ice. Unfrozen shore areas were recorded with the Global Positioning System (GPS) and photographed for detailed studies. This preliminary mapping was used as a guide for fieldworks in summer 2009.

Electrical conductivity, pH and lake-water and lakebed temperature measurements

Groundwater discharge zones can be located by searching for anomalies in electrical conductivity (EC) values (Lee 1985, Vanek and

Lee 1991, Harvey *et al.* 1997, Rosenberry and LaBaugh 2008) near the sediment–water interface. Fresh surface waters and more mineralized local groundwaters commonly differ with respect to their EC levels, and a ratio of 2:1 for groundwater to surface water may be sufficient for identification. The EC level of shallow groundwater is typically low in Finland, usually less than 0.5 mS cm^{-1} (Lahermo *et al.* 1990, Korkka-Niemi 2001). The groundwater EC values were approximately 0.2 mS cm^{-1} at the closed Honkala groundwater intake plant (the Finnish Defence Forces monitoring programme of the old municipal waterworks, 2001). The EC values of the lake water were (uppermost 5 m) approximately 0.09 mS cm^{-1} at the lake water observation station (Pyhäjärvi 93 va93) of the Regional Environmental Centre (HERTTA database at www.ymparisto.fi/oiva [In Finnish]).

The pH of groundwater and surface water also usually differ. The pH of lake water is partially dependent on biological activity, which results in more variability in pH values than in groundwater pH values. Other factors affecting lake water pH include the quality of the precipitation and the properties of soil in the watershed and lake bed. Groundwater in Finland is generally slightly acidic due to the bedrock (Lahermo *et al.* 1990), which consists of acidic igneous and metamorphic rocks and only minor fractions of carbonates (Lavapuro *et al.* 2008). According to Lahermo *et al.* (1990) the mean pH value is 6.4 (median = 6.4, $n = 663$) in sand and gravel aquifers in Finland. According to drinking water quality study (Lahermo *et al.* 2002) the arithmetic mean pH value of groundwater is 6.5 in rural shallow household wells dug into sand, gravel or till sediments ($SD = 0.532$, $n = 698$). The mean pH value were 6.5 (median = 6.5, $SD = 0.16$, $n = 20$) measured at the closed Honkala groundwater intake plant near the Pyhäjärvi (the Finnish Defence Forces monitoring programme of the old municipal waterworks, 2001). The pH values of the lake water (uppermost 5 m) varied between 6.8 and 7.8 at the lake water observation station (Pyhäjärvi 93 va93) (HERTTA database at www.ymparisto.fi/oiva [In Finnish]).

Groundwater discharge zones can be located by searching for temperature anomalies in surface water in the vicinity of the sediment–water

interface and in lakebed (Lee 1985, Silliman and Booth 1993, Conant 2004, Rosenberry and LaBaugh 2008). These methods require a discernable difference in temperatures between groundwater and surface water (Lee 1985) and a sufficient flow through the lakebed to allow advective processes to be significant relative to conductive temperature signals (Rosenberry and LaBaugh 2008).

The temperature of groundwater remains relatively constant throughout the year, whereas lake water temperatures vary seasonally and diurnally. In summer, lake water and lakebed temperatures in groundwater discharge zones are relatively colder than lake water and lakebed temperatures outside these zones, and can be used for locating groundwater discharge zones in lakes. The annual range of Pyhäjärvi water temperature is from 0.5 °C to 21 °C (HERTTA database, SYKE, 2009). The mean annual air temperature varies from 4.1 °C to 5.0 °C in the southern Finland (Finnish Meteorological Institute 1991). The groundwater temperature is approximately 6.6 °C at the groundwater monitoring station of the Regional Environmental Centre at the Vaanii water intake plant (HERTTA database at www.ymparisto.fi/oiva [In Finnish]), 25 km north of the municipal of Säskylä.

An YSI model 600XLM-V2-M multiparameter probe (YSI Inc., Yellow Springs, Ohio, USA) was used for measuring *in situ* EC, pH and temperature values near the sediment–water interface and in surface water column along the studied transects. We used an YSI probe also to verify the water type (lake water/groundwater) of the seepage meter collection tests and the water samples. The uppermost lakebed temperatures (10–30 cm below the sediment–water interface) along the transects were measured, using a stainless steel temperature probe (Electronic Temperature Instruments Ltd., Worthing, West Sussex, UK).

Seepage meters

Seepage meters are used for the direct measurement of seepage flux (Lee 1977, Lee and Cherry 1978, Carr and Winter 1980, Shaw and Prepas 1990a, 1990b, Avery 1994, Duff *et al.* 1999, Paulsen *et al.* 2001). The basic concept of the

seepage meter is to cover and isolate part of the sediment–water interface with a chamber open at the base and measure the change in volume of the water contained in a bag attached to the chamber over a measured time interval (Fig. 2). The time when a bag is connected and when it is subsequently disconnected is recorded, as well as the change in volume of the water in the bag.

The seepage flux is the magnitude and direction of water flowing at the interface between the surface water and groundwater system. The convention is that positive seepage flux values indicate the flow of groundwater to the surface waterbody (also referred to as discharge) and negative flux values indicate the flow from the surface waterbody to the aquifer (also referred to as recharge). The seepage fluxes were calculated with the following equation:

$$Q_s = [(V_f - V_0)/A]/t \quad (1)$$

where V_0 is the initial volume of water in the bag (cm^3), V_f is the final volume of water in the bag (cm^3), t is the time elapsed between when the bag was connected and disconnected (s), and A is the sediment surface area covered by the seepage meter (m^2). Seepage flux is measured as the volume of water per unit area per unit time, yielding seepage flux in $\text{cm}^3 \text{ m}^{-2} \text{ s}^{-1}$. Seepage flux velocity (Q_v) is also a useful term because it normalizes the area covered by the seepage meter and allows comparison of results with other studies and other sizes of seepage meters (Rosenberry and LaBaugh 2008); the unit of flux velocity is cm s^{-1} . The seepage flux velocities were calculated with the following equation:

$$Q_v = (V_f - V_0)/tA \quad (2)$$

The seepage meters used in this study were built as shallow plastic drums sealed at one end, based on the classic design of Lee (1977). Four different designs were used at Pyhäjärvi (heights: 30, 30, 26 and 16 cm and bottom radius: 28.7, 16.5, 27.3 and 26.5 cm). The seepage meters were installed in the lake bottom near the shoreline at the six study sites (Fig. 1) in the area overlapping the unfrozen and open water shoreline areas in winter, except for Kotkastenlahti due to coarse substrate. The installation depths of the seepage

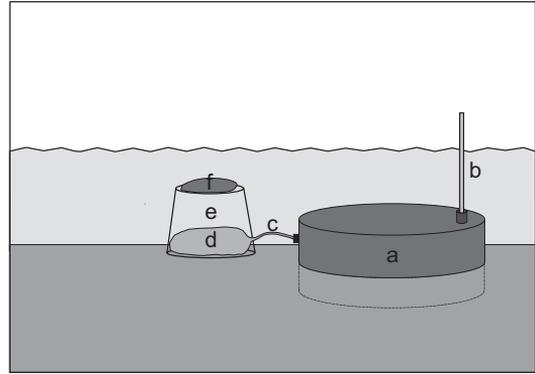


Fig. 2. Basic design of a seepage meter with inverted open chamber (a); top vent with a tube (b), measuring tube with quick release connector to chamber (c), a seepage collection bag (d), installed in a protective housing (e), which is overlaid by extra weight (f) (modified from Lee and Cherry 1978).

meters varied between 10 and 15 cm. The depths of the water varied between 10 and 80 cm at the installation locations and the distances from the shoreline between 0.3 and 7.0 m. After installation the seepage meters were left to equilibrate, because the compression of the sediments and the disturbance of the natural rate of water flow through the sediments during installation (Rosenberry and LaBaugh 2008). The equilibration times used in this study varied from 30 minutes to two days. The collection bags (4-l thin-walled plastic bags) were prefilled with 300 ml of lake water before attaching them to meters to prevent errors caused by the memory effect (Shaw and Prepas 1989, Blanchfield and Ridgeway 1996). The perforated buckets and nests weighted down with stones were used to protect the collection bags from wave action. The collection times varied between 5 min and several hours, depending on seepage flux velocity. A total of 95 measurements were done during autumn 2008 and summer 2009. The proper functionality of the seepage meters was verified by measuring the pH and EC values of water collected from seepage meters at the beginning of a measurement series.

Mini-piezometers

A mini-piezometer is used to measure the hydraulic head in geologic material that is satu-

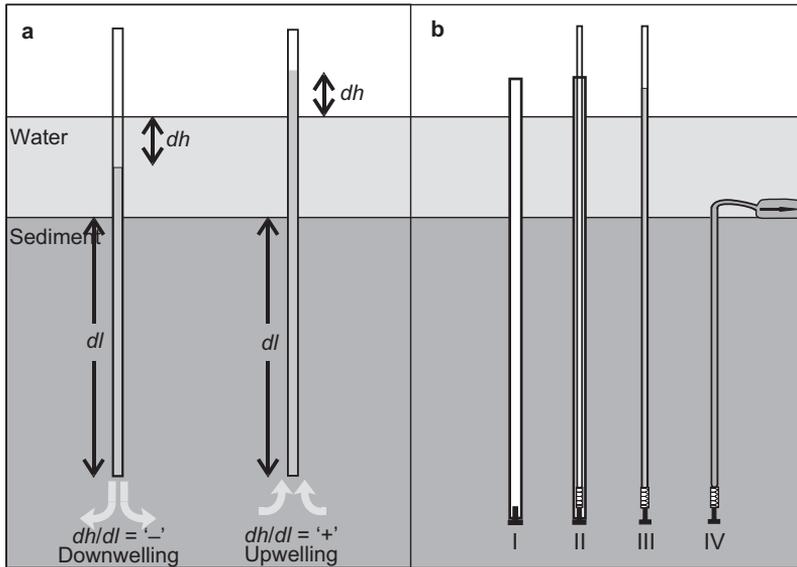


Fig. 3. (a) Vertical hydraulic gradient in a downwelling and upwelling region of the lakebed (modified from Hauer and Lamberti 2006). (b) General features and bolt installation method of a mini-piezometer (modified from Lee and Cherry 1978); (I) casing driven into sediment, (II) plastic tube with screened end inserted in the casing, (III) plastic tube is a piezometer and indicates a differential head with respect to the surface water, (IV) plastic bag attached to the mini-piezometer collects sediment-porewater.

rated under positive pressure (Lee and Cherry 1978). A mini-piezometer provides (1) a comparison between the stage of the surface water and the hydraulic head beneath the surface water at the depth to which the screen at the end of the probe is driven (Lee and Cherry 1978, Winter *et al.* 1998), (2) rapid characterization of the direction and magnitude of the vertical hydraulic gradient (VHG) (Rosenberry and LaBaugh 2008), (3) information on the hydraulic conductivity of the sediments (Baxter *et al.* 2003, Kelly and Murdoch 2003), and (4) a method for collecting water samples (Lee and Cherry 1978, Winter *et al.* 1998, Rosenberry and LaBaugh 2008).

The VHG values between the lake and groundwater were calculated at certain depths in a single point, using the equation:

$$\text{VHG} = dh/dl \quad (3)$$

where dh is the hydraulic head difference between the mini-piezometer and lake stage (cm), and dl is the vertical distance between the lakebed and the midpoint of the mini-piezometer screen perforations (cm) (Fig. 3a). Negative values of VHG indicate loss of water from the lake to groundwater, and positive values indicate groundwater discharge into the lake.

In the present study, the mini-piezometers consisted of a small-diameter translucent, plastic

tube (ID 4 mm, OD 6 mm) ending in a screen and a short (length 5 cm, ID 10 mm, OD 12 mm) perforated section (approximately 14 holes 4 mm in diameter), surrounded by a suitably fine mesh (300 μm). Mini-piezometers were hand driven into the lakebed, using the bolt method described by Lee and Cherry (1978) (Fig. 3b). A total of 41 mini-piezometers were installed along the shorelines and perpendicular to the shorelines at the study sites during autumn 2008 and summer 2009. The middle points of the mini-piezometer screens were installed at depths ranging between 34 cm and 276.5 cm below the surface of the lakebed. Head differences were measured with a ruler outside the tube. Mini-piezometers were used in all seven study locations for measuring head differences, for characterization of the direction and magnitude of the vertical hydraulic gradient, and for collecting water samples for PCE/TCE and isotope analysis. The EC and pH values of the water samples collected were measured to verify the water quality at the depth of the screen.

Isotopic and chemical tracers

The ratios of the oxygen isotopes present in water have been used for decades to distinguish sources of water, including groundwater discharge to sur-

face waterbodies (e.g. Dincer 1968, Krabbenhoft *et al.* 1990, Cey *et al.* 1998). The method works well when the degree of isotopic fractionation is different for different sources of water. In precipitation, the δD and $\delta^{18}O$ values are strongly related following the Global Meteoric Water Line (GMWL) (Craig 1961). Studies of shallow groundwaters in temperate climates have shown that the isotope ratios of oxygen and hydrogen closely follow those in mean annual precipitation (Friedman *et al.* 1964, Fritz *et al.* 1987, Ingraham and Taylor 1991). The isotope composition of shallow groundwaters lies almost exactly in the GMWL line in southern Finland (Karhu 2001). Evaporation tends to enrich the lake water in ^{18}O relative to the composition of the original precipitation (Gat *et al.* 1994, Gat 1996) and shifts the composition away from the GMWL line. Thus, the ratio of lighter to heavier isotopes will change over time in the water and the water vapour.

Isotopic tracers were used to confirm the discharge of groundwater to the lake at all seven study sites in autumn 2008 and summer 2009. A total of 17 isotope samples were collected from the lake water (one sample), mixed water at the shoreline (six samples) and groundwater (10 samples). Groundwater isotope samples were collected from seepage meters and mini-piezometers. All samples were analysed at the Department of Geology, University of Helsinki. The results are given in δ units defined by:

$$\delta^{18}O = [(R_{\text{sample}} - R_{\text{standard}})/R_{\text{standard}}] \times 1000 \quad (4)$$

where R_{sample} and R_{standard} are the $^{18}O/^{16}O$ ratios for the sample and standard, respectively. The isotopic ratios are expressed as a permil (‰) difference from reference, in this study international Vienna Standard Mean Ocean Water (VSMOW).

PCE and its degradation product TCE may be used as tracers in the Honkala aquifer. The presence of PCE in the aquifer is due to a contaminant spill at a dry-cleaning laundry near the esker around 1977 (Artimo 2002). The detectable limit for PCE and TCE is $0.5 \mu\text{g l}^{-1}$. A total of four groundwater samples (three samples collected from mini-piezometers and one sample from well) were collected and analysed at MetropoliLab in Helsinki in 2008.

Results

Winter mapping

A total of 38 different unfrozen areas were detected along and farther from the shoreline (Fig. 4). Three types of area were detected, indicating groundwater discharge of some amount into Pyhäjärvi (Fig. 4): unfrozen shore without open water present (Fig. 4a), unfrozen shore and open water present along the shoreline (Fig. 4b and c) and areas of soft lake ice farther from shore (Fig. 4d). Four additional study sites: Rowhouse, Levomäki, Hannula and Kotkastenlahti, were selected for the study, based on winter mapping of shorelines. In the present study, the Boathouse, Kivimäki, Rowhouse and Kotkastenlahti study sites had unfrozen shores and open water present along the shoreline in March 2009. The Heinonen, Levomäki and Hannula study sites had unfrozen shores without open water present along the shoreline in March 2009.

Electrical conductivity, pH, lake water and lakebed temperature measurements

The EC values of groundwater were usually 1.5–4 times greater than the EC values of lake water. Regardless of season, the lake water EC values varied between 0.080 mS cm^{-1} and 0.093 mS cm^{-1} at the study sites (Table 1). The EC values of groundwater varied spatially between 0.125 mS cm^{-1} and 0.423 mS cm^{-1} (Table 1). The pH value range was between 7.0 and 9.2 in lake water, and varied greatly both temporally and spatially (Table 2). The pH values of groundwater were more constant; they varied between 5.4 and 7.0. The relative variations were smaller in groundwater pH values than in lake water pH values at each study site (Table 2). Anomalous EC and pH values in shoreline water near the sediment-water interface were detected at the Boathouse, Kivimäki, Rowhouse and Kotkastenlahti study sites in autumn 2008 and summer 2009, but not at the Heinonen, Levomäki and Hannula study sites.

The lake water and lake sediment temperature measurements detected a single temperature anomaly at the Kivimäki study site in autumn

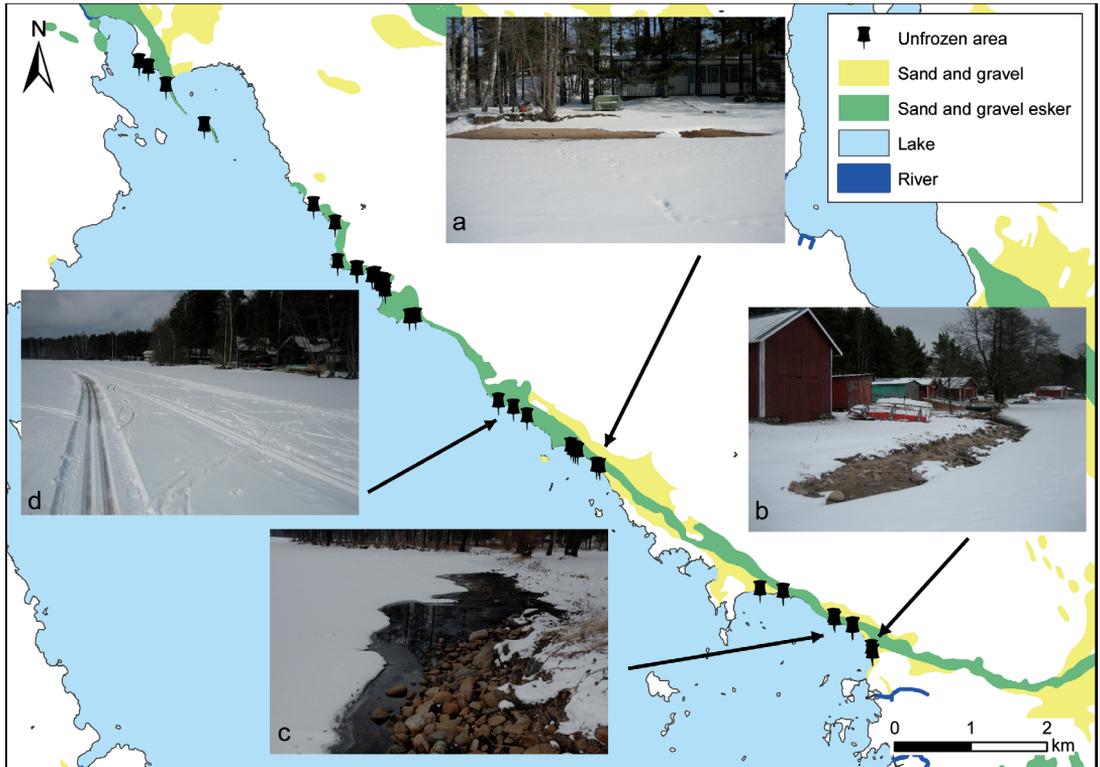


Fig. 4. Locations of potential groundwater discharge areas after winter mapping; (a) unfrozen shoreline without open nearshore water, (b and c) unfrozen shoreline with open nearshore water, (d) weak lake ice farther distance from shoreline.

Table 1. EC values in lake water, groundwater and shoreline water. n = number of measurements.

Location	n	EC _(lake water) (mS cm ⁻¹)	n	EC _(shoreline) (mS cm ⁻¹)	n	EC _(groundwater) (mS cm ⁻¹)
Heinonen	26	0.088–0.92	16	0.089	0	no data
Boathouse	26	0.086–0.091	12	0.100–0.157	27	0.194–0.244
Kivimäki	3	0.089–0.093	10	0.099–0.223	17	0.199–0.253
Rowhouse	5	0.086–0.092	9	0.100–0.200	16	0.315–0.423
Levomäki	1	0.086	5	0.086	11	0.125–0.149
Hannula	1	0.084	2	0.084	6	0.151–0.159
Kotkastenlahti	4	0.080–0.086	3	0.080–0.116	3	0.232–0.241

Table 2. pH values in lake water, groundwater and shoreline water. n = number of measurements.

Location	n	pH _(lake water)	n	pH _(shoreline)	n	pH _(groundwater)
Heinonen	26	7.31–7.54	16	7.41–7.49	0	no data
Boathouse	26	7.33–9.22	12	6.86–8.30	27	6.53–6.95
Kivimäki	3	7.59–8.85	10	6.81–7.61	17	6.34–6.92
Rowhouse	5	7.55–8.31	9	6.40–7.50	16	6.49–6.85
Levomäki	1	8.20	5	7.38–7.90	11	5.37–6.19
Hannula	1	6.99	2	6.99	6	5.95–6.40
Kotkastenlahti	4	7.10–9.20	3	6.83–8.60	3	6.20–6.50

2008. The shoreline water temperatures near the sediment–water interface were 1–4 °C lower than those of lake water, and the lake sediment temperatures were 1–3 °C lower at the shoreline than farther into the lake. The temperature measurements in summer 2009 detected three distinct temperature anomalies in the shoreline water and lake sediment temperatures, indicating groundwater discharge areas in the Boathouse, Kivimäki and Rowhouse study sites. The surface water temperature varied between 16.4 and 18.9 °C, the shoreline water temperatures between 8 and 13.2 °C and the lake sediment temperatures between 7.6 and 16 °C (depths of 15–30 cm) at the Boathouse, Kivimäki and Rowhouse study sites in summer 2009. Lakebed temperatures as low as 7.3 °C were measured at 1 m depth at the Boathouse study site in July 2009. We were not able to measure the lakebed temperatures at the Kotkaskenlahti study site due to coarse substrate. The other three study sites showed no distinctive anomalies in shoreline water or lakebed temperatures.

Seepage meter measurements

A total of 65 seepage flux measurements are in the results (Table 3). A total of 30 seepage

flux measurements were excluded from the total results due to the presence of lake water and measurement errors (the collection bags had lost protective covers or had pinholes). The EC and pH values of collection tests and the water samples collected were monitored with an YSI probe in the fieldwork to verify the presence of groundwater. The average seepage flux rates varied from 4.69×10^{-5} to 4.80×10^{-3} cm s⁻¹ (Table 3) at the Boathouse, Kivimäki and Rowhouse study sites. The highest seepage fluxes were measured in the vicinity of the shoreline and the average groundwater seepage velocity diminished with increasing distance from the shoreline at these three study sites. Seepage meters collected both groundwater and lake water at the Heinonen, Levomäki and Hannula study sites. A coarse substrate prevented installation of the seepage meters at the Kotkaskenlahti study site.

Mini-piezometer measurements

The hydraulic head differences (*dh*) measured between groundwater and lake water indicated upward flow of groundwater at the Heinonen, Boathouse, Kivimäki, Rowhouse and Kotkaskenlahti study sites (Table 4). The head differences measured with 25 mini-piezometers varied

Table 3. Seepage meter data collected in 2008 and 2009. *n* = number of measurements, *l* = distance from shoreline (m), $Q_v(AM)$ = Average seepage flux velocity (arithmetic mean) (cm s⁻¹), $Q_s(AM)$ = Average seepage flux (arithmetic mean) (cm³ m⁻² s⁻¹).

Location	Monitor	<i>n</i>	<i>l</i> (m)	$Q_v(AM)$ (cm s ⁻¹)	Q_{vmin} (cm s ⁻¹)	Q_{vmax} (cm s ⁻¹)	$Q_s(AM)$ (cm ³ m ⁻² s ⁻¹)
Boathouse	SM8	5	1.3	3.89E-04	3.53E-04	4.51E-04	3.89E+00
Boathouse	SM3	6	3.5	2.48E-04	1.84E-04	3.10E-04	2.48E+00
Boathouse	SM5	4	4	1.35E-04	1.23E-04	1.50E-04	1.35E+00
Boathouse	SM6	1	7	4.69E-05	–	–	4.69E-01
Kivimäki	SM28	5	0.3	4.80E-03	4.31E-03	5.23E-03	4.63E+01
Kivimäki	SM29	7	0.3	4.10E-03	3.64E-03	4.54E-03	3.71E+01
Kivimäki*	SM30	4	0.3	7.90E-04	6.43E-04	8.83E-04	7.90E+00
Kivimäki	SM27	7	0.5	4.01E-03	3.35E-03	4.43E-03	4.01E+01
Kivimäki**	SM1	3	1	5.42E-04	2.82E-04	8.46E-04	5.42E+00
Kivimäki	SM24	5	1	2.06E-03	1.52E-03	2.74E-03	2.06E+01
Kivimäki	SM25	3	1	1.82E-03	1.30E-03	2.23E-03	1.82E+01
Kivimäki	SM18	4	1.3	2.48E-03	1.40E-03	3.55E-03	2.48E+01
Rowhouse	SM10	7	0.5	1.23E-03	1.14E-03	1.36E-03	1.23E+01
Rowhouse	SM11	4	1	5.11E-04	4.85E-04	5.28E-03	5.11E+00

* SM30 may have been affected by clay lenses. ** SM1 tests carried out in autumn 2008.

between 1 and 26 cm and the vertical hydraulic gradients between 0.011 and 0.128 at these five study sites (Table 4). Positive vertical hydraulic

gradient values indicate groundwater discharge into the lake. The head differences between the groundwater and lake stage were zero at the Lev-

Table 4. Mini-piezometer data collected at the Heinonen, Boathouse, Kivimäki, Rowhouse and Kotkaste-lahti study sites in autumn 2008 and summer 2009. l = distance from shoreline (m), dl = the vertical distance between the lakebed and the midpoint of the mini-piezometer perforations (cm), dh = head difference (cm), VHVG = vertical hydraulic gradient.

Location	Monitor	Date	l (m)	dl (cm)	dh (cm)	VHVG
Heinonen	MP2	3 Nov. 2008	2	187	10	0.053
Heinonen	MP2	5 Dec. 2008	2	181	6	0.033
Boathouse	MPA	26 May 2009	0.3	229	20	0.087
Boathouse	MPA	29 May 2009	0.3	228	19	0.083
Boathouse	MPA	1 June 2009	0.3	226	18.5	0.082
Boathouse	MPA	2 June 2009	0.3	226	20	0.089
Boathouse	MPB1	26 May 2009	0.3	99	5	0.051
Boathouse	MPB1	29 May 2009	0.3	99	8	0.081
Boathouse	MPB1	1 June 2009	0.3	97	5	0.052
Boathouse	MPB2	26 May 2009	0.3	134	5	0.037
Boathouse	MPB2	29 May 2009	0.3	100.5	6.5	0.065
Boathouse	MPB2	1 June 2009	0.3	128	10	0.078
Boathouse	MPC	26 May 2009	0.3	204.5	21	0.103
Boathouse	MPC	29 May 2009	0.3	200	19.5	0.098
Boathouse	MPC	1 June 2009	0.3	198.5	18.5	0.095
Boathouse	MPC	2 June 2009	0.3	201.5	18.5	0.092
Boathouse	MPD	26 May 2009	2.3	168.5	4.5	0.024
Boathouse	MPD	29 May 2009	2.3	168.5	5.5	0.033
Boathouse	MPD	1 June 2009	2.3	167	5	0.030
Boathouse	MPE	29 May 2009	0.3	202.5	26	0.128
Boathouse	MPE	1 June 2009	0.3	202.5	25	0.123
Boathouse	MPF	29 May 2009	0.3	260.5	25.5	0.098
Boathouse	MPF	1 June 2009	0.3	257.5	25.5	0.100
Boathouse	MPF	2 June 2009	0.3	257.5	26	0.101
Boathouse	MPG	1 June 2009	0.3	276.5	14	0.051
Boathouse	MPG	2 June 2009	0.3	276.5	14	0.051
Boathouse	MPH	1 June 2009	0.5	254.5	18	0.071
Boathouse	MPH	2 June 2009	0.5	249.5	22.5	0.090
Boathouse	MPI	1 June 2009	1.5	193	20.5	0.106
Boathouse	MPI	2 June 2009	1.5	189	23	0.122
Boathouse	MP3	5 Dec. 2008	2.0	132	5.5	0.042
Kivimäki	MP α	1 July 2009	0.2	125.5	6.5	0.052
Kivimäki	MP α	2 July 2009	0.2	125.5	6	0.048
Kivimäki	MP β	1 July 2009	0.2	166.5	6	0.036
Kivimäki	MP β	2 July 2009	0.2	166.5	6.5	0.039
Kivimäki	MP γ	1 July 2009	0.2	216	7	0.032
Kivimäki	MP γ	2 July 2009	0.2	216	9.5	0.044
Kivimäki	MPIII	31 Oct. 2008	shoreline	127	5.3	0.042
Kivimäki	MP4	31 Oct. 2008	0.3	34	2	0.059
Kivimäki	MP5	31 Oct. 2008	0.3	103	3.5	0.034
Kivimäki	MP6	31 Oct. 2008	0.3	152	4.5	0.030
Kivimäki	MP7	31 Oct. 2008	2.3	146	4.5	0.031
Kivimäki	MP8	31 Oct. 2008	4	136	2.5	0.018
Kivimäki	MP9	31 Oct. 2008	shoreline	147	5	0.033
Rowhouse	MPJ	2 June 2009	shoreline	151	2	0.013
Rowhouse	MPM	2 June 2009	0.2	98	8	0.082
Rowhouse	MPM	3 June 2009	0.2	92.5	6	0.065
Kotkaste-lahti	MPP	10 June 2009	0.2	95	1	0.011

Table 5. Oxygen isotope data collected in 2008 and 2009 (after Korkka-Niemi *et al.* 2011).

Location	Date	Monitor	Sample	$\delta^{18}\text{O}$ (‰, VSMOW)
Heinonen	5 Dec. 2008	MP2	Groundwater	-12.14
Heinonen	5 Dec. 2008	Inshore water	Mix	-8.09
Boathouse	5 Dec. 2008	MP3	Groundwater	-12.00
Boathouse	10 June 2009	MPI	Groundwater	-11.97
Boathouse	10 June 2009	MPF	Groundwater	-11.90
Boathouse	5 Dec. 2008	Inshore water	Mix	-8.22
Kivimäki	5 Dec. 2008	SM1	Groundwater	-12.04
Kivimäki	2 July 2009	SM7	Groundwater	-12.07
Kivimäki	5 Dec. 2008	Inshore water	Mix	-8.26
Kivimäki	6 Aug. 2009	Inshore water	Mix	-11.04
Kivimäki	5 Dec. 2008	Lake water	Surface water	-8.05
Rowhouse	10 June 2009	MPN	Groundwater	-11.62
Rowhouse	6 Aug. 2009	Inshore water	Mix	-7.77
Levomäki	17 June 2009	MPV	Groundwater	-11.47
Hannula	17 June 2009	MPÅ	Groundwater	-11.61
Kotkaste-lahti	17 June 2009	MPP	Groundwater	-11.48
Kotkaste-lahti	6 Aug. 2009	Inshore water	Mix	-7.93

omäki and Hannula study sites, indicating that groundwater does not discharge into the lake in the vicinity of the shore zone.

Isotope and chemical tracers

Lake water $\delta^{18}\text{O}$ value was -8.05‰ , whereas inshore water $\delta^{18}\text{O}$ values varied between -7.77‰ and -11.04‰ VSMOW (Table 5). Groundwater $\delta^{18}\text{O}$ values varied between -11.47‰ and -12.14‰ VSMOW (Table 5). The PCE and TCE concentrations in the groundwater samples collected were detectable and the method was useful for identification of different types of water. Detectable concentrations of total PCE and TCE varied between 3 and $108 \mu\text{g l}^{-1}$ in the groundwater samples collected (Table 6). The PCE and TCE concentrations decreased in samples in northwards direction and only minor concentrations of PCE and TCE were present at the Kivimäki study site.

Discussion

Identification of groundwater discharge areas

The groundwater discharge locations in this

study were identified with temperature, EC and pH measurements of lake water near the sediment-water interface, lakebed temperature measurements and winter mapping of shoreline areas at Pyhäjärvi. In comparing the different types of unfrozen shore areas with the results of other methods used in this study, we recognized an association between the type and extension of unfrozen shore area with the amount of discharging groundwater into lake water from the same area. Discharging groundwater to a surface waterbody could be detected with the other methods used in this study, where the shore areas were unfrozen and open water was present in March 2009 (Fig. 4b and c).

In summer the temperature differences between the groundwater and lake water/lake bed were highlighted when the lake water warmed up. Three discharging groundwater areas (Boat-

Table 6. PCE/TCE data collected on 13 November 2008.

Location	Monitor	PCE and TCE ($\mu\text{g l}^{-1}$)	TCE ($\mu\text{g l}^{-1}$)	PCE ($\mu\text{g l}^{-1}$)
Heinonen	MP2	108	13	95
Boathouse	MP3	48	5.8	42
Kivimäki	MP4	13	1.2	12
Kivimäki	Well	3	< 0.5	2.9

house, Kivimäki and Rowhouse) were detected by shoreline water and lake bed temperature in situ measurements in summer 2009. Conant (2004) developed flux based conceptual model with five different types of groundwater discharge behaviours. In comparing the lakebed temperatures and the seepage velocities measured in our study with the conceptual model of Conant (2004), we were able to characterize and categorize groundwater discharge behaviours at the study sites. The sediment temperatures varied between 7.6 and 16 °C at the Boathouse, Kivimäki and Rowhouse study sites, indicating high and low-to-moderate groundwater discharge flow type at these locations according to conceptual model of Conant (2004). The coldest lakebed temperatures (< 8.5 °C) measured were close to temperature of groundwater (6 °C), indicating high discharge flow type at the Kivimäki ($Q_{vmax} = 5.23 \times 10^{-3} \text{ cm s}^{-1}/4.52 \text{ m day}^{-1}$) and Rowhouse study sites ($Q_{vmax} = 5.28 \times 10^{-3} \text{ cm s}^{-1}/4.56 \text{ m day}^{-1}$), where the coldest lakebed temperatures measured were localized at narrow shoreline zone. The coldest shoreline water temperature (8.0 °C) was measured at the Kivimäki study site (lake water 18.9 °C). The areas of coldest shoreline water and lakebed temperatures overlapped both the areas of highest groundwater discharge velocities measured with the seepage meters and the areas of unfrozen and open water shore zones. The shoreline water and lakebed temperatures increased with distance from the shoreline, while the groundwater discharge rates measured by seepage meters decreased with distance from the shoreline. The lakebed temperatures were similar to the lake water temperatures at the Heinonen, Levomäki and Hannula study sites, indicating no discharge flow type according to Conant (2004). The mid-summer shoreline water temperature measurements were a good reconnaissance tool for locating areas of discharging groundwater into a surface waterbody, but not sufficiently reliable to measure seepage in absolute terms. However, the method can still be useful for defining relative differences in seepage fluxes. In autumn the temperature differences between groundwater and lake water were detectable, but distinctly smaller than in summer, and only the Kivimäki study site revealed anomalous temperatures in the shore-

line water and lake bed.

The EC in situ measurements were used both to identify the groundwater discharge areas and to monitor water quality in the seepage meter and mini-piezometer collections to verify the presence of groundwater. In the present study the EC values of groundwater were 1.5–4 times of the magnitude lake water. Lee (1985) used EC values to locate the groundwater discharge areas but stressed that anomalies cannot be ascribed to groundwater flow without additional investigation with existing methods; thus the method should be considered only as a reconnaissance tool (Rosenberry and LaBaugh 2008). The high EC values and low lakebed temperatures were positively correlated between areas of high groundwater discharge measured with seepage meters in this study. The measured EC values of shoreline water as well as groundwater discharge decreased rapidly with distance from the shoreline.

In the present study the pH values of groundwater were approximately one unit lower than in the lake water. The pH values offered one additional method for the identification of groundwater discharge areas. The pH values of groundwater varied between 5.4 and 7.0 and those of lake water between 7.0 and 9.2. The groundwater pH values were more consistent at each study site compared with those of lake water at the same location. These anomalies cannot be ascribed to groundwater discharge without additional confirmation with other methods. The large Isosuo swamp, located on the eastern side of the Levomäki and Hannula study sites, may affect groundwater quality at both locations, especially the low pH values of groundwater measured.

Measured seepage fluxes and hydraulic gradients

In general, the seepage meter results showed an approximately exponential decrease in seepage flux velocities with increasing distance from the shoreline at the Boathouse, Kivimäki and Rowhouse study sites and the highest seepage velocities were concentrated in the narrow areas of the shorelines (Pearson's $r = -0.5650$, $p < 0.01$, $n = 65$) (Fig. 5). When data are collected

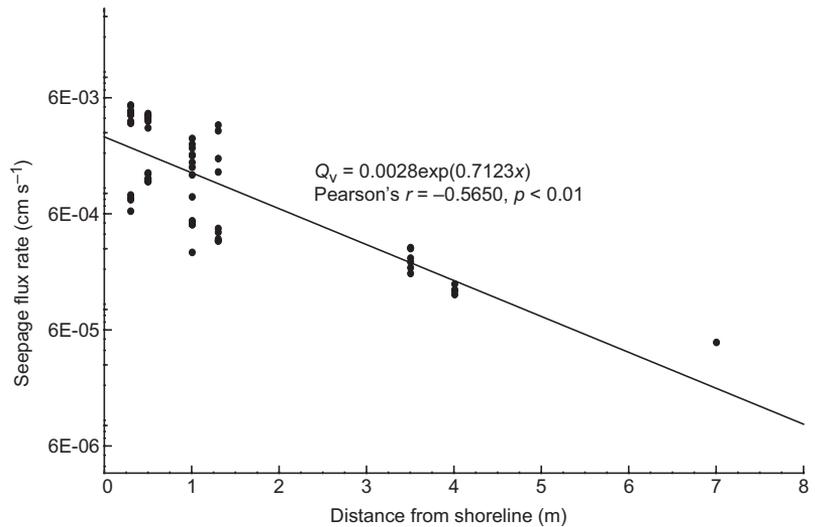


Fig. 5. Relationship of seepage velocity (in logarithmic scale) and distance from shoreline at the Boathouse, Kivimäki and Rowhouse study sites (modified from Korkkaniemi *et al.* 2011).

along transects perpendicular to the shore and the groundwater flux is distributed exponentially with distance from the shore, the exponential curves can fit through the transect data, and the equation for the curve can be used to calculate the water flux for an area deemed to be representative of the transect point measurements (Rosenberry and LaBaugh 2008). The seepage rates measured showed the highest variation in values at short distances from the shoreline. Nearshore discharge was rapid, from 3.89×10^{-4} to 4.80×10^{-3} cm s⁻¹, and the highest average seepage velocity 4.80×10^{-3} cm s⁻¹ was measured at the Kivimäki study site. The range of seepage rates that have been reported from coastal and freshwater settings is several orders of magnitude (Rosenberry and LaBaugh 2008), including seepage rate values varying between 1.15×10^{-7} cm s⁻¹ (Cherkauer and McBride 1988) and 6.0×10^{-2} cm s⁻¹ (Duff *et al.* 1999). The seepage rates measured are moderate to high values, as compared with the wide range of seepage flux rates reported for other lakes (Shaw and Prepas 1990b).

Shaw and Prepas (1990a) determined that seepage rates could vary by more than a factor of 2 when meters are installed only 1 m apart, due to heterogeneity of the substrate. The detected thin clay lenses (5–10 cm) beneath sand, gravel and pebbles can be an interpretive factor for spatial variability in flux across the sediment–water interface measured at the Kivimäki study site (Table 3). Seepage meters collected lake water

with a minor groundwater component at the Heinenon, Hannula and Levomäki study sites. Based on results from temperature measurements, seepage meters, mini-piezometers and winter mapping, groundwater probably discharges to the lake farther from shore at the Levomäki and Hannula study sites.

Measurement errors can be introduced with the design and operation of seepage meters. A seepage meter only measures flux at a point in space and many measurements are required to derive meaningful interpolations. Manual seepage meters also measure only the aggregate seepage over the time period and provide no data on how seepage changes during that time period. High spatial and temporal variability in seepage characteristics can result in poor repeatability of measurements (Kaleris 1998). The most common sources of error, when operating with seepage meters, are insufficient equilibration time, collection bags, weight of an observer, leaks and measurement errors (Rosenberry and LaBaugh 2008). Wave action influences the seepage rates observed; therefore, calm weather conditions were preferred. Correction factors were not used in this study and therefore the seepage values presented could have resulted in underestimation of the actual seepage fluxes. Due to the range of potential sources of error, we recommend that seepage meter measurements be accompanied by other indirect methods (e.g. hydraulic head difference, lakebed temperature

measurements, chemical tracers and isotopes) to verify the direction and likely magnitude of the seepage flux. Despite these limitations, the seepage meter is still the method that is most commonly available for direct measurements of water flow at the interface between aquifers and surface water features.

The range of differences in the hydraulic head, although dependent on the installation depth, is typically from 0 to 10 cm, but head differences as much as 30 cm are not uncommon (Rosenberry and LaBaugh 2008). Differences in the hydraulic head ranged from 0 to 26 cm at Pyhäjärvi, and a maximum head difference of 26 cm occurred at MPF at the Boathouse study site (Table 4). Vertical hydraulic gradients ranged from 0.011 to 0.128. The results from mini-piezometer tests indicated positive upwelling groundwater conditions at the Heinonen, Boathouse, Kivimäki, Rowhouse and Kotkasteenlahti study sites and supported the results from the seepage meter tests at the Boathouse, Kivimäki and Rowhouse study sites. All comparisons of head differences, vertical hydraulic gradient, and horizontal hydraulic gradient were done at approximately the same time, due to temporal variations in groundwater movement. The horizontal gradients indicated that groundwater discharge decreased with distance from the shoreline at the Boathouse and Kivimäki study sites. Positive groundwater upwelling conditions could not be verified with the mini-piezometer tests at the Levomäki and Hannula study sites, where the head differences measured were zero. Based on the measured EC values and isotope analysis of these sampled mini-piezometers at the Levomäki and Hannula study sites, we assumed that all mini-piezometers were installed in the groundwater zone. However, groundwater was not discharging from this layer into the lake, although it may have been discharging at some distances farther from the shoreline. Both the Levomäki and Hannula study sites need further investigations to verify discharging groundwater conditions at these locations. Some sources of error attend the use of a mini-piezometer. An erroneous determination of the depth of screen and erroneous reading of the head difference are common sources of errors. An improper seal between the outer pipe and the sediments can also cause errors.

Verification of groundwater discharge using isotopic and chemical tracers

The isotope samples were taken successfully both from the mini-piezometers and seepage meters. In the literature, cautions are issued regarding collection of water quality samples from seepage meters (Brock *et al.* 1982, Belanger and Mikutel 1985). The residence time of water contained inside the seepage chamber or bag may allow the chemistry of the water to change and samples may not be representative (Rosenberry and LaBaugh 2008). However, in the present study the results from the isotope analysis sampled (the sample collections times between 10 and 15 min) from the seepage meters showed no errors caused by the sampling procedure when the results from the seepage meters and mini-piezometers were compared. The measured $\delta^{18}\text{O}$ values of groundwater ranged from -12.00‰ to -12.14‰ VSMOW in samples taken in late autumn 2008 and values from -11.47‰ to -12.07‰ VSMOW in samples taken in summer 2009. These values are in good agreement with those reported by Karhu (2001) and Kortelainen and Karhu (2004). Karhu (2001) reported a mean $\delta^{18}\text{O}$ value of -11.9‰ (5-year monitoring programme) for shallow groundwater in southwestern Finland. Kortelainen and Karhu (2004) reported a mean $\delta^{18}\text{O}$ value of -11.98‰ for the Junnila monitoring site at Eura, which is located approximately 10 km to the north from the Kotkasteenlahti study site. Kortelainen (2002) reported a mean $\delta^{18}\text{O}$ value of -12.54‰ for groundwater in the Virttaankangas aquifer (6-year monitoring programme). The seasonal variations in the isotopic composition of groundwater in southern Finland span a range of 0.4‰ – 1.0‰ for oxygen (Kortelainen and Karhu 2004). The range of measured oxygen isotope composition of groundwater (0.67‰) between different samples and sampling dates may be the result of seasonal variations. The results of isotopic analysis confirmed the results of all other methods used in this study that groundwater is discharging into Pyhäjärvi and discharging groundwater fluxes can be measured with seepage meters and mini-piezometers.

PCE and TCE concentrations (3 – $108 \mu\text{g l}^{-1}$) in four samples from discharging groundwa-

ter confirm the computer modelling estimation (Artimo 2002) that PCE concentrations in contaminated groundwater discharging into the lake would be between 10 and 30 $\mu\text{g l}^{-1}$ over most of the region where the contaminated Honkala aquifer intersect the lake in 2010. Groundwater discharge into the Pyhäjärvi is the main process for the natural attenuation of PCE via mixing into lake water and evaporation to the atmosphere (Artimo 2002). The maximum allowable PCE and TCE concentration in drinking water according to the Finnish Ministry of Social Affairs and Health (2000) is 10 $\mu\text{g l}^{-1}$, which will be reached in groundwater by the year 2030 at the latest according to Artimo (2002). PCE and TCE will not be useful as tracers farther northwest from the Kivimäki study site, because their expected concentrations will be under the detectable limit (0.5 $\mu\text{g l}^{-1}$).

Conclusions

The overall goal of this study was to examine the groundwater seepage to Pyhäjärvi on a 10-km northeastern shoreline and the factors that influence it. The more specific goals were to (1) identify the locations where groundwater discharges to surface waterbody in the study area, (2) determine the directions and rates of groundwater seepage with seepage meters and mini-piezometers, and (3) compare and evaluate both the applicability of the methods used and the results of the methods.

Winter mapping is a useful method for the selection of possible groundwater discharge areas for detailed study at Pyhäjärvi. However, the optimal time for mapping these discharge areas is short and good weather conditions must prevail to make detection possible. When the results from winter mapping were compared with the results of other methods used in this study, we were able to confirm moderate to high groundwater discharge to unfrozen and open water shoreline areas.

In the present study, the summer was the best season for the temperature-based methods because of the distinct temperature differences between the surface water (16.4–18.9 °C) and the groundwater (6 °C). Midsummer shoreline

water and lakebed temperature measurements were good reconnaissance tools for locating areas of discharging groundwater into the surface waterbody. The in situ EC measurements could be used to identify groundwater discharge areas. The EC values of groundwater were 1.5–4 times greater than those of lake water, depending on the study site. The pH values of groundwater were approximately one unit lower than those of lake water but these anomalies cannot be ascribed to groundwater discharge without additional confirmation with other methods. The relatively high EC values, low pH values and low lakebed and shoreline water temperatures were positively correlated with areas of moderate to high groundwater seepage velocities as measured with seepage meters.

Seepage meters could be used to measure direct groundwater seepage flux in Pyhäjärvi. Nearshore groundwater discharge was rapid (3.89×10^{-4} – 4.80×10^{-3} cm s^{-1}), and velocity declined approximately exponentially with distance from the shoreline at the Boathouse, Kivimäki and Rowhouse study sites. Additionally, the seepage meters were used successfully for sampling isotope and PCE/TCE samples. The mini-piezometer was a useful device for characterization of the direction and magnitude of the vertical hydraulic gradient and collecting groundwater samples at Pyhäjärvi. Head differences and vertical hydraulic gradients confirmed upwelling groundwater conditions at five study sites and that groundwater discharge decreased with increasing distance from the shoreline at three study sites. The coarse and stony substrate limited the use of seepage meters and mini-piezometers at some study sites.

The results of isotope analysis confirmed that groundwater is discharging into Pyhäjärvi and verified the proper working of seepage meters and mini-piezometers. PCE and its degradation product TCE could be used as tracers in the Honkala aquifer. PCE/TCE constituents were useful tracers only in a limited contaminated part of the study area.

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