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Discussions

Confronting vicinity of the surface water and sea shore in a shallow glaciogenic aquifer in southern Finland

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Abstract

The chemical characterization of groundwater in shallow, unconfined, low-lying coastal aquifer in Santala, southern Finland was identified by utilising the integrations of multivariate statistical approaches; principal component analysis (PCA) and hierarchical cluster analysis (HCA), with the stable isotope δ^2H and δ^{18}O, hydrogeochemistry and field monitoring data. HCA classified groundwater samples into 6 distinct groups that reveal the factors controlling temporal and spatial variations of groundwater geochemistry such as geology, anthropogenic sources from human activities, climate and surface water. High temporal variation of groundwater chemistry corresponds directly to precipitation. With increase in precipitation, $KMnO_4$ consumption, EC, alkalinity and Ca concentrations also increase in most wells, while Fe, Al, Mn and SO_4 are occasionally increased during spring after the snowmelt in specific geologic condition. The continued increase of NO_3 and metals concentrations in groundwater indicates the potential contamination risk to aquifer. Stable isotopes of δ^{18}O and δ^2H indicate groundwater recharges directly from the meteoric water with insignificant contribution from lake water, and no seawater intrusion into the aquifer. Groundwater geochemistry suggests local seawater intrusion being temporary able to take place in the sulphate reduction zone along the fresh and seawater mixed zone in the low-lying coastal area but contribution of seawater was very low. The influence of lake water can be observed from the increases of $KMnO_4$ consumption in wells nearby the lake.

1 Introduction

Shallow permeable aquifers located next to sea shore may face more risks and challenges in water consumption and management compared with the inland water intake. Aquifers along the coastline are vulnerable especially to the seawater intrusion either due to the sea level rise or storm surge (e.g. Oude Essink, 1999, 2001; Barlow, 2003; Pulido-Leboeuf, 2004; Oude Essink et al., 2010; Rasmussen et al., 2013; Luoma et al.,

2013) – phenomena those will presumable be accelerated by the changing climate (IPCC, 2000, 2007; Nicholls et al., 2007). Besides this, increasing water demand for population and industries as well as changing land use practises as a result of human activities e.g. exceeded water intake, gravel excavation pits, car parking, and ground-

5   water contamination risk areas would expose shallow aquifers for contamination.

Under the changing climate, a potential increase of precipitations in the winter, spring and autumn and increase of evapotranspiration in summer owing to an increase of temperature is expected (Scibek and Allen, 2006; Scibek et al., 2007; Jyrkama and Sykes, 2007; Okkonen and Kløve, 2011; Okkonen, 2011). The change of climate would po-

10   tentially affect not only the groundwater, but also the surface water. For the sustainable groundwater resources management and land use planning, it is important to understand the hydrogeological processes and the interactions between groundwater and surface water, factors controlling groundwater quality.

Approximately 300 of total classified 6000 Finnish shallow groundwater areas (ex-

15   cluding Ahvenmaa) locate 100 m or less from the shore of the Baltic Sea (Hertta database-SYKE, 2013). The Baltic Sea is one of the largest brackish water areas in the world (EEA, 1999). Although it contains low salinity, e.g. varies from 3.0–7.0‰ in the Gulf of Finland (Alenius et al., 1998; UNEP, 2005; Fagerlund, 2008) which not pose a high risk on the seawater intrusion compare to the other parts of the world

20   (e.g. Oude Essink, 1999, 2001; Barlow, 2003; Pablo Pulido-Leboeuf, 2004; Oude Essink et al., 2010; Rasmussen et al., 2013), the salinity concentration is, however, in the level higher than drinking water standard, and in the long term, it can damage the infrastructure such as pipeline network.

Seasonal variations in groundwater quality are reported in many unconfined shallow

25   aquifers in Finland, e.g. a lower dissolved concentration in groundwater during the snowmelt period indicates effect of the snowmelt (Backman et al., 1999; Korkka-Niemi, 2001; Okkonen, 2011). So far there is still no clear understanding of the surface water and groundwater interactions and also the impacts of brackish seawater intrusion on the shallow, low-lying coastal aquifers. Main ion chemistry, as well as stable isotopic

composition of oxygen-18 (δ^{18}O) and hydrogen (δ^2H or deuterium), could be used to assess these interactions as noticed before by, e.g. Allen (2004), Harbison (2007), Kortelainen (2007), and Mongelli (2013).

A shallow glaciogenic aquifer in the Hanko area in southern of Finland, our case study area, confronts these issues as aimed to maintain water quality within a drinking water standard in a long term. The objective of this study was to investigate the factors controlling groundwater quality, the main water type and chemical evolution of groundwater in the low lying, porous coastline aquifer in southern Finland. The interaction between the Baltic Sea and the lake and groundwater, and the temporal and spatial variations of groundwater and surface water geochemistry in different recharge periods was studied. The data used consist of field investigation and hydrogeochemical data. The stable isotope ratios of δ^{18}O and δ^2H were used to determine the origin of groundwater. The multivariate statistical methods: hierarchical cluster analysis (HCA) and the principal component analysis (PCA) (Cloutier et al., 2008) in conjunction with the conventional groundwater classification method by Piper diagram were used to analyse and classify the geochemistry of water samples.

2 The study area

The study area is located in Santala in the northern coast of Hanko peninsula, southern Finland at approximately 59°53″ N 23°10″ E (Fig. 1), and covers approximately 17 km^2 of the main aquifer. This aquifer is an important source of drinking water and the water supply for Hanko town (with the population of 9282 in 2013) and local industries. The aquifer consists of porous gravels and sands of an ice-marginal end deposit, and is located in the low-lying coastal area bounded by the Baltic Sea, and a lake in the middle of the aquifer. Topographic landform varies 10–14 m a.s.l. along the northern ridge of the First Salpausselkä Quaternary ice-marginal formation (Fig. 1) and decreases its slope into less than two metres along the northern coastline. While in the south and south east the elevation is gradually decrease to 5–7 m a.s.l. The southern part is partly

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covered by wetlands and peatlands. In the east the area is covered by sand dune terrains from the Aeolian deposit. The lake and wetlands in the middle of the aquifer are located in the depression area as a part of the First Salpausselkä formation and sand dunes terrain. Lake surface area is about $1.8\,km^2$, and it is shallow with the average depth approximately 1–2 m. In many parts of the aquifer, groundwater level is close to the ground surface and water intake areas are located along the coastline, where groundwater level may often fall below the sea level. The groundwater level has previously been observed to have a rapid response to the rise of sea level as well as the recharge from the snowmelt and rainfall (Backman et al., 2007; Luoma et al., 2013). Additionally, the existing of human activities in the area, e.g. gravel excavation pits, industries or the Highway (Fig. 1), could pose more contamination risk to the groundwater quality.

The Hanko area belongs to the temperate coniferous-mixed forest climate zone with cold, wet winters. During the study period, the mean annual temperature stayed at approximately $+6\,°C$, which agrees the long-term mean annual temperature in the reference period of 1981–2010 (FMI, 2013). Meteorological parameters (max, min and mean temperatures, precipitation and snow) both in 2012 and during the period of 1981–2010 measured at the Tvärminne weather station, approx. 5 to 8 km south eastern of the study area are given in Fig. 2. The lowest daily temperatures are generally recorded in January and February, and those of the highest during July and August (Fig. 2). Year 2012 was exceptionally rainy as the cumulative amount of annual precipitation was 893 mm. This is 41 % higher than the long-term mean value of 634 mm (1981–2010).

2.1 Geology and hydrogeology of the study area

The stratigraphy in Hanko area is underlain by the basement of the Precambrian crystalline igneous and metamorphic rock and covered by the Quaternary deposit. The Precambrian bedrock consists mainly of granite, quartz diorite and granodiorites, formed a sharp contact with the Quaternary deposit with some outcrops in the area (Kielosto

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et al., 1996). Aquifer in the study area is in First Salpausselkä ice-marginal formation deposited during the Weichselian and Holocence deglaciation of the Scandinavian Ice Sheet (Fyle, 1991; Saarnisto and Saarinen, 2001). The First Salpausselkä ice-marginal formation consists of the glacial till, gravel, sand and clay; and the postglacial littoral gravel, sand and clay (Fig. 1).

The primary ice-marginal formation in Hanko was formed in deep water with a low narrow ridge (Fyle, 1991). When the ice sheet withdrew from the area, this in deep water sitting formation was covered by fine-grained sediments, silt and clay layers, of the Ancylus Lake and Littorina Sea. The sea level has been regressive since the glacial time caused by the isostatic land uplift. The primary deposit of the First Salpausselkä formation was exposed to sea waves and also for wind (Kielosto et al., 1996). The well sorted gravels indicate rework materials from the high energy of wave and storm activities are found in a large area in Santala, the study area, while the fine sand from Aeolian deposits covers a large area in the east and southeast of the lake (Fig. 1) (Fyle, 1991; Kielosto et al., 1996).

The shallow aquifer in Hanko is unconfined with the thickness of the Quaternary deposit varying from less than one meter to 75 m, with the average thickness about 25 m. The sediments are generally thick in the western side, in NE–SW direction conforms the First Salpausselkä formation, and decrease eastward to less than one meter in the eastern coast area. Groundwater table in Santala is between 2–7 m below ground surface in inland area, and less than 2 m below ground surface in the coastal area, where groundwater discharges into the Baltic Sea. The results of well testing and soil sample analysis show that the hydraulic conductivity of aquifer varies from 0.3–4.8 m d^{-1} in silty sand and fine sand, and up to 100 m d^{-1} in sand and gravel (Luoma and Pullinen, 2011). Groundwater recharge occurs mainly twice a year during spring (late March–early April) and late autumn (November–early December) from the infiltration of snowmelt and rainfalls (Luoma et al., 2013). Groundwater flows mainly northward into the coastal area and also towards south-southeast into the wetlands and peatlands as well as towards the Baltic Sea in the east.

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3   Materials and Methods

3.1   Sampling, field investigation and well monitoring

The field investigation and water sampling were carried out four times during winter, spring, summer, and autumn 2012. Water samples were collected from 15 sites in the Santala area, including water sample from 12 groundwater observation wells (Table 1), a lake, the Baltic Sea, and water intake well (Fig. 1). In a total 26 samples were taken for chemical analysis and 59 samples for the analysis of stable isotope compositions of oxygen and hydrogen. In addition, water chemical analysis results of 6 water samples taken during spring 2010 as well as groundwater monitoring data (depth (D), temperature (T), electrical conductivity (EC)) and field measurements data including D, T, EC, pH, dissolved oxygen (O_2), redox potential (Eh), and dissolved carbon dioxide (CO_2) of 12 wells in Hanko area performed during 2009–2010 were used to support the interpretation.

The field investigation consists of the down-hole profile logging and field measurement of water samples during the water sampling. The profile logging was performed in order to investigate the vertical distribution of physical and chemical parameters of groundwater in the well screen interval of the observation wells, and the water sampling depths were selected based on the results of these logs. The measurement was performed by using either WTW P4 instrument (during the winter) or YSI Professional Plus (IP-67) – a multi-parameter recording device, for the measurement of EC, pH, T, O_2 and Eh. The CO_2 was measured by titration. The pumping rate, time, and amount of water pumped before sampling were recorded. Also the groundwater tables were measured before and after the pumping.

Well monitoring was carried out for 8 groundwater observation wells during March to November 2012 and the Sandösträsket Lake during September to November 2012. The measurement was done by installing the Schlumberger Mini-Diver data logger and pressure transducer at the same sampling depths. Three wells locate nearby the northern coastline area were monitored for T, D and EC. Lake water and 5 wells in the

mainland, which situate far from the coastline and the water intake well, were monitored for T and D. All data were measured hourly. Time series of climate data (air temperature and precipitation as rainfall or snow thickness) and sea level of the same period in the Hanko received from the Finish Meteorological Institute (FMI, 2013) were used for comparison.

The groundwater from the observation wells were pumped for a minimum of 20 min until all parameters readings (e.g. T, EC) were constant before sampling. Groundwater from the water intake well was taken from tap by allowing water to flow out until all parameters (e.g. T, EC) were constant. The surface water samples (seawater and lake water) were taken with Limnos water sampler from 1 m below the water surface. The number and sampling depth of groundwater samples of each well was selected from the profile-logged results. If there were no variations in the profile, a sample was taken from the middle of the screen interval, at least 2 m below the top screen.

3.2 Analytical methods and quality control

For cation analysis, water samples were filtrated through 0.45 μm membrane filters and collected into 100 mL high density polyethylene (HDPE) bottles. The samples were acidified and analysed in the Labtium Ltd accredited laboratory in Espoo. Filtered and acidified water samples were analysed for major and minor elements (Ag, Al, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, I, K, Li, Mn, Mo, Ni, P, Pb, Rb, Sb, Se, Sr, Th, Tl, U, V, Ca, Fe, Mg, Na, Si and S) by using the inductively coupled plasma atomic emission and mass spectrometry (ICP-AES and ICP-MS) techniques. Alkalinity (as HCO_3), EC, pH, and $KMnO_4$ consumption were measured from unfiltered and unpreserved samples immediately upon the samples arrived to the laboratory. They were analysed also for anion (Br, Cl, F, NO_3, PO_4 and SO_4) by using ion chromatography (IC) technique. To ensure the quality of the equipment and chemical analysis, a zero-sample is prepared from the distilled water and four duplicate pairs of groundwater samples were also analysed. The zero and duplicated samples analysis results indicated no field contamination. The charge balance error (BE) (Drever, 1997) was calculated for all

water samples. 26 of total 34 water samples have BE < ±5 %, and 8 samples have BE between ±6 % and ±10 %. The chemical analysis and quality control results from Labtium Ltd indicate high proportions of anions in some water samples and the BE cutoff at ±10 % was acceptable. Many studies such as Appelo and Postma (2005) and Allen (2004) recommended BE cutoff at < ±5 % for good water analysis, Drever (1997) and Harbison (2007) used BE cutoff at ±10 % due to the presence of high organic acid concentration in fresh groundwater samples from the shallow coastal aquifer.

3.3 Stable isotopes oxygen and hydrogen analysis

The stable isotopic δ^{18}O and δ^2H are commonly used in many studies concerning groundwater flow and the interaction of groundwater and surface water. These traditional water isotopes are effective tracers to be used to identify water origin as well as processes of mixing isotopically distinct water masses and evaporation in surface water reservoirs (Gonfiantini, 1986; Richter and Kreitler, 1991; Taylor and Howard, 1996; Clark and Fritz, 1997; Kendall and McDonnell, 1998; Allen, 2004; Faure and Mensing, 2005). Both the recent regional isotopic composition of atmospheric precipitation and groundwater are well known in southern Finland (Kortelainen, 2007; Kortelainen and Karhu, 2004). Based on monthly isotopic values of oxygen and hydrogen in Finnish precipitation, the national scale meteoric water line (δ^2H = $7.67\delta^{18}$O + 5.79 ‰) was established by Kortelainen in 2007. The line agrees well to local lines derived to the southern coast of Finland (Kortelainen, 2007, 2009) and can be thereby applied to this study. In temperate climates as in Finland, the isotopic composition of the local mean annual precipitation closely follows that of local groundwater and most of the seasonal isotopic variations in rainfall are smoothed out during the infiltration process (Kortelainen, 2007; Kortelainen and Karhu, 2004). Stable isotope method has been widely applied in Finland, especially to evaluate both natural groundwater surface water interaction (Rautio and Korkka-Niemi, 2011) and artificially enhanced surface water infiltration into the aquifers (Kortelainen and Karhu, 2006; Hendriksson, 2012).

For the stable isotopes δ^{18}O and δ^2H analysis, the filtered water samples ($< 45\,\mu$m) about 60 mL were collected in the HDPE bottle and were kept in the dark cooler and out of the sunlight during transported to the Research Laboratory, Geological Survey of Finland, Espoo. Isotopic composition of δ^{18}O and δ^2H were analysed by laser based

5   cavity ring-down spectroscopy (CRDS), using a Picarro isotopic water analyser. In the CRDS method the absolute abundances of water molecules with different combinations of stable isotopes of δ^{18}O and δ^2H are measured from a vaporized sample in an optic chamber. The isotopic composition of water is reported using δ-notation per mill (‰) relative to the VSMOW (Vienna Standard Mean Ocean Water) standard. The δ-value is

10   defined as $\delta = ((R_{SA} - R_{VSMOW})/R_{VSMOW}) \times 1000$, where SA is the sample and VSMOW is the standard, $\delta = \delta^{18}$O and $R = {}^{18}$O$/{}^{16}$O for oxygen, and $\delta = \delta^2$H and $R = {}^2$H$/$H for hydrogen. The repeatability of analyses was < 0.1 ‰ for δ^{18}O and < 0.5 ‰ for δ^2H.

3.4   Statistical analysis

Data correlations and statistical analysis were performed by using the IBM SPSS

15   Statistics program to determine the variables that represent the controlling factors behind geochemistry of all water samples taken during April 2012 and August 2012. The correlations between variables were carried out for all raw data by using the Pearson correlation coefficient to identify the relationships between variables among the water sampling sites.

20   The multivariate statistical approaches, Principal Component Analysis (PCA) and Hierarchical Cluster Analysis (HCA) were used to group the water samples or variables analysed. PCA is a factor analysis method that used to extract components representing the information contained in the data that explain the pattern of correlations and differences within a group of variables (Korkka-Niemi, 2001; IBM SPSS Statistics,

25   2013). The number of the factors or components was extracted by the PCA with the Varimax rotation based on Kaiser Normalization.

HCA is a classification method that reveal natural groupings or clusters within a data set by reorganizing the data into homogenous groups and link the two most similar clusters until all of the variables are joined in a complete classification tree (Korkka-Niemi, 2001; IBM SPSS Statistics, 2013). The results of HCA are presented in a dendrogram which constructed using the Ward's method (Ward, 1963) with the Euclidean distance as a measure of similarity between the samples. Ward's method is one of the most widespread hierarchical clustering methods used in the classification of the hydrogeochemical data by using the minimum variance to evaluate the distances between the clusters (Güler et al., 2002; Cloutier et al., 2008; Templ et al., 2008).

The data used for PCA and HCA consists of stable isotope δ^2H and δ^{18}O, chemical parameters, pH and EC. The dataset include variables having normal or close to normal distributions from the chemical parameters (alkalinity, B, Ba, Ca, I, Li, Mg, Na, Rb, S, Si, Sr), and δ^2H, δ^{18}O, pH and EC. The variables having non-normal distribution (Al, Cl, Cu, Mn, K, $KMnO_4$, Mo, Ni, NO_3, SO_4, U and Zn) were log-transformed prior to statistical analysis. The variables below or close to the detection limit (Ag, Be, Bi, Br, Cd, Co, Cr, F, Fe, P, PO_4, Se, Th and Tl) were excluded from the analysis. Data was standardized by subtracting the sample mean from each variable and dividing by the standard deviation (Z score standardization) prior to multivariate analysis to ensure that each variable is weighted equally (Güler et al., 2002; Cloutier et al., 2008; Templ et al., 2008).

3.5 Data processing

Data analysis was performed in Microsoft Excel for data storage, cross-plots and basic calculation. GWChart (USGS, 2012) for Piper Diagram, a plot of the relative concentration of major dissolved species of anions and cations, in order to identify the water types (Piper, 1944; Appelo and Postma, 2005). ArcGIS/ArcMap program was used for spatial analysis and mapping of the spatial variations of chemical concentrations in water and data from field measurement. For comparison to the chemical analysis results of the water samples, median values of groundwater compositions of Finnish shallow aquifers

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(Lahermo et al., 2002) and the precipitation (rainfall and snowmelt) collected from the Tvärminne weather station (Vuorenmaa et al., 1999; Järvinen and Vänni, 1996, 1997), recommended values for drinking water for Finnish household wells from the Ministry of Social Affairs and Health (STM, 2001) were used as references.

4   Results

4.1   Water levels

Lake level monitoring during September–November 2012, groundwater levels from monitoring wells during March–November 2012 and daily precipitation and temperature data from the Tvärminne weather station in Hanko area (FMI, 2013) is presented in Fig. 3. Groundwater levels show that the main recharge took places twice a year; during spring (early April) suddenly after snowmelt period (Fig. 3b) where groundwater reaches its highest level, and during late autumn (October–December). Groundwater levels reach the lowest levels during winter and late summer due to snow cover and evapotranspiration, respectively. Mean groundwater levels range from 8.4 m a.s.l. in Obs8 to 12.3 m a.s.l. in Obs1 for the wells in the mainland, and from 0.4 m a.s.l. in Obs11 to 0.5 m a.s.l. Obs10 for the wells in the coastal area. Time series of lake level data during September–November 2012 varies from 11.83–11.98 m a.s.l. (Fig. 3b). The previous measurements of lake levels during this study were 11.75 m a.s.l. in November 2009 and 12.08 m a.s.l. in January 2012. The variation of lake levels during the year is less than 0.5 m. Obs2 locates approximately 170 m west of the lake shoreline. Groundwater level at Obs2 during March–November 2012 varies between 11.13 and 11.79 m a.s.l. which is lower than lake level. The fluctuation of groundwater level in Obs2 has a positive correlation with the lake level with the Pearson correlation coefficient of 0.94, ($\rho < 0.01$).

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4.2 Geochemistry of water

Statistics summary of groundwater and surface waters geochemistry is presented in Table 2. The EC profile logging in Obs8 and Obs6 showed higher values in the top section than in the bottom section of the observation well. The partition samples were taken from Obs8 and Obs6 during summer for chemical and stable isotopic analysis, and during autumn for stable isotope analysis. Figure 4 presents the profile log results of Obs6 and the sampling depths. The bottom samples could not be taken from a deeper depth because of the packer got stuck into the observation well in certain depths.

4.3 Stable isotopes of oxygen and hydrogen

The δ^2H and δ^{18}O values of groundwater vary spatially and temporally with the range between -86.90 to -76.50‰ VSMOW and -12.30 to -10.62‰ VSMOW, with mean values of -80.39 ± 2.52 (1σ)‰ and -11.37 ± 0.36 (1σ)‰ VSMOW, respectively. The mean δ^2H and δ^{18}O values for the lake water are -55.57 ± 9.65‰ and -6.85 ± 2.08‰, respectively, and for the seawater are -57.65 ± 2.35‰ and -7.55 ± 0.38‰, respectively. Characteristics of the δ^2H and δ^{18}O data from 59 water samples in Santala are shown in Table 3.

5 Discussions

5.1 Origin of groundwater

The stable isotopes δ^2H and δ^{18}O were used to differentiate the origin of groundwater from the meteoric origin and the surface water. The δ^2H and δ^{18}O of groundwater from this study are consistent with Kortelainen and Karhu (2004) and Kortelainen (2007), where the mean δ^2H and δ^{18}O values of the groundwater from southern Finland ($n = 37$) are reported at -82.0 ± 0.9‰ and -11.55 ± 0.14‰ VSMOW,

respectively. The stable isotopes δ^2H and δ^{18}O of groundwater in Santala are also consistent with the mean δ^2H and δ^{18}O values of the precipitation from southern Finland ($n = 60$) is -82.0 ± 23.6‰ and -11.54 ± 3.1‰ VSMOW, respectively. These values are distinct from the surface water where the mean δ^2H and δ^{18}O values of lake water are -55.57 ± 9.6‰ and -6.85 ± 2.1‰, respectively, and mean of seawater are -57.65 ± 2.4‰ and -7.55 ± 0.4‰, respectively (Table 3).

Most of the δ^2H and δ^{18}O values of groundwater from Santala fall closely on the Finnish local meteoric water line (Fig. 5, LMWL) indicating direct recharge from the precipitation (snowmelt and rainfall) with no indications of evaporation nor contribution from the surface water. Due to evaporation, the δ^2H and δ^{18}O values of lake and seawater deviate to the right from the LMWL. The higher the evaporation effect is the further right from the local groundwater and the LMWL (Gonfiantini, 1986). In surface waters, high seasonal variations are also recorded (Fig. 5). The variations are larger in lake water than in seawater. Both lake and sea water show the highest evaporation effect in the summer samples.

Only sign of possible mixing of groundwater and surface water was observed from sample taken in spring 11 April 2013 in Obs2, approx. 170 m west of the lake shoreline, where a slightly heavier δ^2H and δ^{18}O values were recorded that fall with a small deviation from the LMWL. Water monitoring data imply the water flow direction from lake into Obs2. The KMnO$_4$ consumptions from April 2012 and August 2012 samples from Obs2 are high relative to the surrounding wells and close to the concentration from lake water, which indicates the influence of lake water into this well.

Majority of the groundwater samples (except from Obs8, Obs7 and Obs2) show a narrow variations with low standard deviations of 1.6 and 0.23‰ for the δ^2H and δ^{18}O, respectively (Fig. 5, insert figure). This may indicate the same source of groundwater recharge with a short percolation time. The fluctuation of groundwater levels in many observation wells (Fig. 3) indicates a rapid response of groundwater level to the precipitations during snowmelt and heavy rains events, which reflect the short percolation time. This is consistent with many wells that contain quite thin vadose zone

thickness (Table 1) and/or high hydraulic conductivity of aquifer materials. However, the observation wells next to a gravel excavation pit (Obs8, Obs7) and a lake shoreline (Obs2) show high seasonal variation with high standard deviations of 3.21 and 0.49‰ for the δ^2H and $\delta^{18}O$, respectively. This may indicate the influences of the gravel excavation pit and lake water.

Median concentrations of major ions in groundwater are low and are close to median concentrations of precipitation (Vuorimaa et al., 1999; Järvinen and Vänni, 1996, 1997) and lower than the median values of shallow groundwater in Finland (Lahermo et al., 2002). The concentrations of minor ions and trace elements (F, Fe, Ag, Be, Bi, Br, Cd, Co, Cr, P, PO_4, Se, Th and Tl) in many wells are below or close to the detection limit. However median values of metals and trace elements such as Fe, As, Cd, Cr, Ni, Pb, Sb, Se, I, Li and PO_4 from groundwater samples are higher than Lahermo et al. (2002) have observed (Table 2).

Piper diagram in Fig. 6 shows the compositions of groundwater in Santala are mainly Ca-HCO_3 type with no cation exchange take place, and only groundwater from Obs6_top and _mixed samples fall closer to Na-Cl type. Groundwater Ca-HCO_3 type, with low dissolved concentrations, low pH, alkalinity, Ca and Mg, is a common groundwater type for glaciated areas in Finland (e.g. Backman et al., 1999; Soveri et al., 2001; Korkka-Niemi, 2001; Lahermo et al., 2002; Backman, 2004). Allen (2004) reported that Ca-HCO_3 type groundwater indicates a young groundwater from the direct percolation or a short resident time, or there is no substrate e.g. clay minerals present for cation exchange. This agreed with the results of low dissolved concentrations found in many groundwater samples.

Chloride concentrations of all groundwater samples vary between 1.5 and 28.3 mg L^{-1}, and have great differences from seawater sample which has Cl concentration of 2690 mg L^{-1}. Elevated Na and Cl concentrations found in Obs6_top sample are much higher than concentrations in Obs6_bottom sample and nearby wells, and they have no correlations with the other variables. Obs6 situates about 50 m in the downstream direction from the Highway running through the middle of the aquifer. Elevated

concentrations of Na and Cl in this well indicate the influences of de-icing chemicals (NaCl) in the Obs6 top section, while the Obs6 bottom part contains overall low dissolved concentrations same as the nearby wells. The use of salt for de-icing purposes in Finland increased in 1987 with the maximum peak in 1990 when 157 000 ton of salt

5 was applied along the highways in Southern Finland, many of them running on top of the First Salpausselkä formation including Hanko area. The use of de-icing road salt has been tried to reduce since 1993 (Gustafsson and Nystén, 2000). The aquifer materials at Obs6 well location consist of the interbedded layers of sand and fine-grained sediments (Luoma et al., 2013). The fine-grained layers could prevent the distributions

10 of Na and Cl in groundwater to the top section.

No intrusion of seawater was observed in Obs10 and Obs11 which locate nearby the coastline and water intake well. Chloride concentration of seawater in this study is $2690\,mg\,L^{-1}$, is consistent with other studies e.g. Alenius et al. (1998) and Fagerlund (2008). The Cl contributions to groundwater in Obs10 and Obs11 calculated from

15 Cl concentrations of water samples, seawater and fresh water (Appelo and Postma, 2005) were less than 0.5 %, which are very small contributions and could have other origin than direct seawater intrusion, e.g. atmospheric fallout. However, from the piper diagram the sulphate reduction may possibly be observed in Obs11, approx. 60 m from the coastline. Obs11 locates in the low-lying area with well head elevation of 0.5 m a.s.l.

20 It contains the highest SO_4 concentration of $38.3\,mg\,L^{-1}$, with CO_2 and O_2 concentrations of 29.0 and $4.1\,mg\,L^{-1}$, respectively. Based on geochemistry of three water samples from this well, SO_4 shows a strong negative correlation with HCO_3 ($r = 0.98$). Seawater has a high SO_4 concentration and when it intrudes into the aquifer, especially into the anoxic low-lying coastal aquifer, it may result in the sulphate reduction (Ander-

25 sen, 2001; Appelo and Postma, 2005) and have a negative correlation with HCO_3. However, the reduction of sulphate while the enrichment of bicarbonate in the mixing zone suggests bacterial reactions (Magaritz and Luzier, 1985). The monitoring data shows groundwater level is occasionally lower than seawater either due to overpumping or sea level rise, which can cause the temporally seawater intrusion into the aquifer.

5.2 Spatial distribution of groundwater geochemistry

5.2.1 Correlation between variables

Correlation coefficient was calculated between in order to identify the relationships between variables among the water sampling sites. Stable isotopes δ^2H, δ^{18}O and KMnO$_4$ consumption seem to have negative correlation with Si and no correlation with the other variables (Table 4). Other variables have statistically significant correlation with two or more analysed variables.

5.2.2 Principal Component Analysis (PCA)

Table 4 shows statistically significant correlations between certain variables. Therefore, principal component analysis (PCA) was worth to do (Table 5) in order to group the variables. After the Varimax rotation with Kaiser Normalization four principal components having eigenvalues higher than one were chosen because they explained over 80 % of the total variance observed (Cloutier et al., 2008; IBM SPSS Statistics, 2013). The factor score of each principal component was calculated for each samples as showed in Table 6.

PC1 is characterized by highly positive loading in SO$_4$, K, Ba, Cu, Mo, Mg, I, Mn, Li, and Si, explaining 43.8 % of the total variance. The highest scores are found in Obs10, Obs9 and Obs11, also a moderate to low positive scores are observed in Obs1 and Obs2_0412 samples. PC1 mainly indicates the natural groundwater quality. PC1 consists of wells that penetrated into the primary deposit of the First Salpausselkä formation (Luoma et al., 2013) and Obs10 and Obs11 locate further in the discharge area. The primary deposit of the First Salpausselkä formation consists of the glacial till, gravel, sand and clay of the glaciogenic sediments from the erosion of the pre-existing Precambrian bedrock, e.g. granite, quartz diorite and granodiorites. PC1 reveals the influence of natural origin on groundwater quality from the dissolution of minerals bearing in the aquifer material such as sulphide or K-feldspar or clay minerals (Lahermo et al.,

2002; Shand and Edmunds, 2008). Shotyk et al. (2009) reported high concentrations of S, Si, Ba, Mg, Mn, Li and Rb in the groundwater from weathering of the dominant silicate minerals in sediments of discharge area in two natural artesian flows in Ontario, Canada.

PC2 has the highest loadings for Ca, alkalinity, pH, EC, NO_3 and Sr explaining 18.0 % of the total variance. It has highly positive loading in Obs8 and Obs10. A low positive loading was observed also in Obs6_0812top sample. The variables in PC2 have strong to very strong positive correlation among each other, especially Ca, alkalinity, Sr and EC have the Pearson correlation coefficients among each other between 0.99 and 0.92 ($\rho < 0.01$) (Table 4). This is related to the solution of carbonate in the groundwater. Obs8 and Obs10 contain the highest concentrations of Ca, Sr and EC, and distinct from the other wells in the study area. Obs8 locates next to the gravel pit and the concrete factory and Obs10 locates in the downstream direction from Obs8. The excess Sr associates with Ca and HCO_3 (alkalinity) compared with the surrounding wells indicates the potentially contamination of groundwater from human activity as same as NO_3 which has high and highest concentrations in Obs8 and Obs10, respectively. Although NO_3 could come from the dissolutions of organic matter under the reducing condition, the increases of NO_3, EC and other dissolved ions concentrations in Obs10 since 2010 during the sampling periods, may indicate a potential contamination from human activity taking place in this area.

PC3 explains 11.6 % of total variance and has the high positive loading in $KMnO_4$ consumption and the stable isotope δ^2H and $\delta^{18}O$. The highest scores are found in lake water and Obs2_0412 samples, and moderate scores in Obs2_0812 and Obs10. PC3 reveals the influences of surface water into groundwater. The presence of $KMnO_4$ consumption in groundwater indicates an influence of surface water or dissolved organic matter (Korkka-Niemi, 2001; Lahermo et al., 2002). In many wells, the concentrations of major ions and some other dissolved ions in groundwater are higher in spring samples than summer samples, while $KMnO_4$ consumptions are higher in summer than in spring. Based only on $KMnO_4$ consumptions, the influences of surface water

should be more observed in summer. However, the δ^2H and δ^{18}O reveal the possible influence of surface water from lake water only in Obs2_0412 sample, and show no influences of surface water in Obs2_0812 and Obs10. Obs11 is locating closer to the seashore than Obs10. Based on its location Obs11 should present more influences

5 from seawater than Obs10. However, PC3 extracted the information existing in Obs10 which contains higher $KMnO_4$ consumption than Obs11 and contributed higher factor score loading for Obs10 than in Obs11. PC3 indicates influences of surface water or surface water induced dissolved organic matter on groundwater quality at Obs10.

PC4 has highly positive loadings in Na and Cl, and is explaining 7.1 % of the total

10 variance. The highest scores are found in Obs6_mixed and _top samples, while the Obs6_bottom sample shows a negative score. The moderate to low positive scores are found also in Obs9, Obs10 and Obs11. PC4 indicates the impact of human activities on groundwater quality such as the influences of road de-icing salt in the Obs6 top section, which locates about 50 m downstream direction of the Highway. Median concentrations

15 of Na and Cl in Obs9, Obs10 and Obs11 (7.3 and 7.9 mg L^{-1}, respectively) are higher than the background Lahermo et al. (2002, 4.18 and 4.40 mg L^{-1}, respectively). Sodium and Cl of those 3 wells show positive correlation with Ca, alkalinity, Sr and EC, the main factor components found in PC2. PC1 reveals the natural impact on the groundwater quality in Obs9, Obs10 and Obs11 while PC4 may indicate some influences of human

20 activity on these wells.

Overall PCA is a useful tool to extract the main components that represent both natural and human activities impacts on the water quality either in regional or site specific scales and Obs10 has shown the most vulnerable well that has been impacted by both natural and human activities.

25 ### 5.2.3 Hierarchical cluster analysis (HCA)

The result of HCA is presented based on the sample locations in the dendrogram as showed in Fig. 7. The clusters were classified based on the linkage distance on

the dendrogram, indicating the level of similarity of variables between clusters. The shorter the linkage distance, the higher similarity of variables between clusters. Based on the linkage distance at 9, the water samples were classified into 6 clusters (C1–C6). Clusters C1, C2 and C3 have a linkage distance between the clusters at 10, which is shorter than the linkage distance of C4, C5 and C6 (at 16). This indicates C1, C2 and C3 have the greater similarity between the clusters. C1 consists of Obs3, Obs4, Obs5, Obs6_bottom, and Obs7. C2 consists of Obs6_top and Obs6_mixed samples. C3 consists of Obs1, Obs2 and lake water. C4 consists of all samples from Obs8. C5 consists of Obs10, and C6 consists of Obs9 and Obs11. The median values of the main variables in each cluster (excluded lake water) are summarized in Table 7.

C1, C2 and C3 contain lower median values of variables compare with C4, C5 and C6. The main differences between C1, C2 and C3 are that C2 contains the highest Na and Cl concentrations than the others, and C3 contains the highest concentrations of Al and $KMnO_4$ consumption. These clusters consist of wells that locate in the groundwater recharge area in the middle of the aquifer and could be grouped to the same cluster. However, with the classification based on the linkage distance at 9, C2 could emphasize the influences of de-icing road salt take place in Obs6 top section and C3 for the influence of lake water into groundwater into Obs2 and Obs1. C2 and C3 are agreed with PC4 and PC3, respectively, except PC3 shows no principal component loading for Obs1.

C4, C5 and C6 consist of wells locating closer to the groundwater discharge and human activities areas. Some dissolved ions in groundwater found in C4 are quite low and close to C1, C2 and C3, but elevated concentrations of Ca, HCO_3, Sr, EC, pH and NO_3 has linked it to C5 which indicates the impact of human activities from the gravel excavation pit and the concrete factory. C5 and C6 contain higher dissolved solute concentrations than the other clusters and are linked together due to they share the same or similar natural origin and probably also some influences from human activities. C4 and C5 are consistent well with PC2, and PC5 and PC6 are consistent with PC1.

5.3 Temporal variability of groundwater geochemistry

Temporal variations of dissolved solute concentrations in groundwater were examined for all groundwater samples between different seasons: spring (April) 2010, spring (April) 2012 and summer (August) 2012. Only data from 4 observation wells (Obs1, Obs8, Obs10 and Obs11) were available for all 3 seasons. The annual precipitation of 2012 was exceptionally high, about 41 % higher than the mean annual precipitation of 1981–2010 and it is the highest precipitation since 1963 (FMI, 2013). The annual precipitations of April 2011–April 2012 (876 mm) is 322 mm higher than the April 2009–April 2010 (554 mm). In 2012 groundwater reached the highest level in April 2012 right after snowmelt, however, the amount of precipitations continued to increase during June to the end of the year due to the increase of heavy rain events during that period (Fig. 2). Percentage changes of selected dissolved concentrations between seasons in different observation well locations are presented in Table 8. The positive number indicates increasing of dissolved concentrations. Table 8 indicates the variations in amount of precipitations or timing of recharge in different seasons having been affected on groundwater quality. However the aquifer area has received mostly the same amount of precipitation at a time, the dissolved concentrations in groundwater have varied spatially in different well locations. Overall, the concentrations of Ca, HCO_3, EC, and $KMnO_4$ consumptions increase in most wells with increase of precipitation. Obs6 and Obs4 show very high variations of $KMnO_4$ consumptions relative to the other wells. The increase of $KMnO_4$ consumptions implies influences of surface water or dissolved organic matter (Korkka-Niemi, 2001; Lahermo et al., 2002).

Obs10 has higher EC and dissolved ion concentrations than the other wells, and EC, Ca, HCO_3, and $KMnO_4$ consumptions have increased since April 2010, while Cl concentration has decreased. Obs10 contains the highest NO_3 concentration, however the high increased rate of NO_3 since April 2010 is observed in Obs11, downstream from Obs10. In addition, Obs10 showed increase of metals concentrations in groundwater with increase of precipitations since April 2010. Okkonen and Kløve (2011) and

Korkka-Niemi (2001) reported a decrease of NO_3 in groundwater during the spring with more recharge from snowmelt. On the opposite, high NO_3 concentrations in spring and autumn are found due to increased nitrogen leaching when runoff increase from forest (Lepistö, 1996) and drained peatlands (Kløve, 2001).

5   The concentrations of Fe, Al, Mn, and SO_4 are occasionally high during spring right after the snowmelt (both in April 2010 and April 2012). However, their concentrations are highly variable and often associated with low pH observed in Obs1, Obs2, Obs4 and Obs11. Iron, Mn and SO_4 are redox-sensitive elements in groundwater and being soluble under the reducing condition (Hem, 1985; Shand and Edmunds, 2008). High
10   concentrations of Al can be associated with clay minerals or organic matter and it has high positive correlation with $KMnO_4$ with the Pearson correlation coefficient of 0.79, ($\rho < 0.01$).

Based on the future climate scenarios A1B and B1, the precipitation in Hanko area is expected to increase 12–26 % and sea level rise up to 0.51 m above mean sea
15   level, compare with the current situation (Luoma et al., 2013), by the end of 2100. The potential increase of precipitation during autumn and winter in the future can cause more fresh water enter to the aquifer and possibly more influences of seawater intrusion due to the sea level rise and storm surges. Based on the observed seasonal variation, the increase of some dissolved concentrations as mentioned earlier and changes of
20   groundwater geochemistry of wells along the coastline can be more expected.

6   Conclusions

Geochemistry of groundwater in the shallow, unconfined, low-lying coastal aquifer in Santala has spatial and temporal variability depending on the geological, anthropogenic, seawater and climate factors. The integrations of multivariate statistical HCA
25   and PCA approaches with the stable isotope δ^2H and $\delta^{18}O$, geochemistry methods, and field investigation data, are successfully used to identify the recharge origin of groundwater in Santala, groundwater classification and potentially contaminated

groundwater in the aquifer area. Groundwater quality evolution based on the future climate scenarios, could be assessed, as well.

The stable isotopes δ^2H and δ^{18}O suggest the shallow, unconfined, low-lying coastal aquifer in Santala recharges directly from meteoric water (snowmelt and rainfall) with small or insignificant contributions from Baltic Sea and lake. Groundwater is mainly Ca-HCO$_3$ type, with low dissolved concentrations, low pH, alkalinity, Ca and Mg concentrations indicating young groundwater from the direct percolation or having short residence time. No significant differences in groundwater geochemistry were observed compared to inland aquifers. Variables SO$_4$, K, Ba, Cu, Mo, Mg, I, Mn, Li, and Si (PC1) indicate the natural water quality in the aquifer, variables Ca, alkalinity, pH, EC, NO$_3$ and Sr (PC2) potential contamination (e.g. gravel pit and the concrete factory), KMnO$_4$ consumption and the stable isotope δ^2H and δ^{18}O (PC3) influences of surface water having higher content of organic matter and Na and Cl (PC4) sea water intrusion or the effect of salt used for road de-icing purpose.

Based on the HCA, the groundwater observation sites can be divided into the two main groups: wells that locate in the groundwater recharge area (C1, C2, C3) in the middle of the aquifer and wells locating closer to the groundwater discharge and human activities areas (C4, C5, C6). The influence of lake water can be observed from the increases of KMnO$_4$ consumption in wells nearby the lake. The geochemistry of groundwater suggests the sulphate reduction in the mixed zone between fresh and seawater indicating local seawater intrusion may temporary take place in the low-lying coastal area but contribution of seawater was very low. The concentrations of Ca, HCO$_3$, EC, and KMnO$_4$ consumptions increase in most monitored wells with increase of precipitation. The concentrations of Fe, Al, Mn, and SO$_4$ are occasionally higher soon after the snowmelt. Based on the future climate scenarios the precipitation in Hanko area is expected to increase and sea level rise up. The potential increase of precipitation during autumn and winter can cause more amount of fresh water enter to the aquifer and possibly some seawater intrusion due to the sea level rise and storm surges. Based on the observed seasonal variation, the increase of some dissolved elements and thus

changes in groundwater geochemistry along the coastline can be expected more in the future.

The groundwater water samples could be classified into the six distinct clusters under the 2 main groups based on the similarity of water geochemistry. The influence of lake water can be observed from the increases of $KMnO_4$ consumption in wells nearby the lake. While both the δ^2H and δ^{18}O, and PCA show no evidence of seawater intrusion in the aquifer, the geochemistry of groundwater suggests the sulphate reduction in the mixed zone between fresh and seawater indicating local seawater intrusion may temporary take place in the low-lying coastal area but contribution of seawater was very low.

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Table 1. Observation well information of the sampling depth, screen depth, and vadose zone thickness during the sampling periods. The negative vadose zone thickness means that well was flooded during the measurement.

| Well | Sampling depth from ground surface (m) | Depth of diver from ground surface (m) | Screen interval from ground surface (m) | Screen length (m) | Well depth (m) | Vadose zone thickness in winter (m) | Sediments between ground surface and screen bottom (%) | | |
							Silt and fine sand	Sand	Gravel
Obs1	8	7	5.8–17.8	12	17.8	1.80	35	48	17
Obs2	10	8	3.5–18.5	15	28.5	1.62	63	38	
Obs3	10	no diver	5.4–32.4	27	36.4	3.89	14		86
Obs4	10	no diver	7.2–25.2	18	36.2	4.95	16	25	59
Obs5	8	no diver	4.8–10.8	6	10.8	5.90			100
Obs6	9	8	4.0–49.0	45	49.0	1.39	56	11	33
Obs6	7	partition sample – top							
Obs6	12	partition sample – bottom							
Obs7	7	8	5.7–8.7	3	8.7	2.95	88	4	8
Obs8	9	13	6.9–31.9	25	31.9	6.58	20	22	58
Obs8	7.7	partition sample – top							
Obs8	13.7	partition sample – bottom							
Obs9	10	8	6.0–14	8	17.0	2.79		45	55
Obs10	6	4	1.4–8.4	7	8.4	1.61	11	64	24
Obs11	6	5	1.7–8.7	7	8.7	−0.15	39	32	29
Obs12	4	no diver	1.0–5.0	4	5.0	1.88	31	38	31

Table 2. Characteristics of groundwater in Santala area, Hanko (number of samples, $n = 32$) compare with data from lake water, seawater, the median values from Lahermo et al. (2002) and precipitation from Vuorenmaa et al. (1999) and Järvinen and Vänni (1997, 1996), and safe drinking water (STM, 2001).

Variable	Unit	Min.	Median	Mean	Max.	Lake water ($n = 1$)	Seawater ($n = 1$)	Lahermo et al. (2002) ($n = 739$)	Precip. ($n = 36$)	Drinking water
Temperature	°C	5.70	7.00	7.39	10.40	8.50	7.80	6.80		
Eh	mV	161	255	258	354	279	230			
O_2	$mg\,L^{-1}$	0.84	5.19	6.08	11.7	7.70	13.1	60.9		
CO_2	$mg\,L^{-1}$	1.00	12.0	12.1	36.0	6.33	1.00	34.0		
EC	$mS\,m^{-1}$	3.75	13.4	13.6	30.2	4.51	826		2.82	250
pH		5.50	6.55	6.61	8.00	6.20	8.41		4.50	6.5–9.5
Na	$mg\,L^{-1}$	1.76	3.10	5.11	14.7	3.04	1360	4.18	0.66	
K	$mg\,L^{-1}$	0.31	0.86	1.39	4.84	0.24	59.2	2.78	0.16	
Ca	$mg\,L^{-1}$	2.48	9.94	17.0	47.0	3.38	69.3	11.4	0.40	
Mg	$mg\,L^{-1}$	0.64	1.54	1.85	4.63	1.01	162	2.38	0.12	
Cl	$mg\,L^{-1}$	1.50	3.50	6.50	28.3	3.60	2690	4.46	1.10	100
SO_4	$mg\,L^{-1}$	5.30	9.50	13.2	38.3	7.40	356	10.4	0.90	250
NO_3	$mg\,L^{-1}$	< 0.2	0.75	1.97	7.00	1.50	< 20	3.19	0.53	50.0
Alkalinity	$mmol\,L^{-1}$	0.10	0.45	0.85	2.43	0.07	1.42	0.54		
HCO_3	$mg\,L^{-1}$	6.10	27.5	52.1	148	4.27	86.6	32.9		
$KMnO_4$	$mg\,L^{-1}$	0.32	1.80	2.56	10.0	21.0	26.0	4.50		20.0
Al	$\mu g\,L^{-1}$	1.02	9.89	37.7	227	12.0	9.98	101		200
Mn	$\mu g\,L^{-1}$	0.06	2.14	15.1	80.8	3.18	7.07	4.36		100
Fe	$mg\,L^{-1}$	< 0.03	0.08	0.15	0.31	0.06	< 0.03	< 0.03		0.40
As	$\mu g\,L^{-1}$	< 0.05	0.26	0.39	1.12	0.37	0.65	0.14		10.0
B	$\mu g\,L^{-1}$	4.46	10.4	10.8	24.3	8.40	579	13.8		1000
Cd	$\mu g\,L^{-1}$	< 0.02	0.03	0.04	0.12	0.03	< 0.02	< 0.02		5.00
Cr	$\mu g\,L^{-1}$	< 0.20	0.39	0.47	1.15	< 0.20	< 0.20	0.20		50
Cu	$\mu g\,L^{-1}$	< 0.10	0.66	1.23	4.39	0.37	0.85	2.49		2000
Ni	$\mu g\,L^{-1}$	< 0.05	1.28	2.19	10.6	< 0.05	0.95	0.84		20.0
Pb	$\mu g\,L^{-1}$	< 0.05	0.09	0.10	0.17	0.38	0.06	0.04		10.0
Sb	$\mu g\,L^{-1}$	0.02	0.04	0.04	0.13	0.11	0.11	0.03		5.00
Se	$\mu g\,L^{-1}$	< 0.50	1.57	1.61	1.87	< 0.50	25.1	< 0.5		10.0
F	$mg\,L^{-1}$	< 0.10	0.24	0.31	0.73	< 0.10	< 1.00	< 0.1		1.40
Ba	$\mu g\,L^{-1}$	0.85	3.51	5.04	16.7	2.80	19.6	18.1		
Br	$mg\,L^{-1}$	< 0.10	< 0.10	< 0.10	< 0.10	< 0.10	19.1	9.92		
I	$\mu g\,L^{-1}$	< 2.00	4.68	6.44	19.8	2.62		2.12		
Li	$\mu g\,L^{-1}$	< 0.10	1.47	1.98	6.94	1.41	25.3	0.77		
PO_4	$mg\,L^{-1}$	< 0.02	0.03	0.04	0.13	< 0.02	0.10	< 0.02		
Si	$mg\,L^{-1}$	3.64	6.13	6.57	9.85	0.10	0.37			
Sr	$\mu g\,L^{-1}$	13.0	42.7	62.6	163	11.9	1300	59.9		
Zn	$\mu g\,L^{-1}$	0.69	4.20	7.20	53.4	12.2	7.75	10.4		

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Table 3. Statistics summary of stable isotopes δ^2H and δ^{18}O data from 15 sample sites from groundwater, water intake well (VO), lake water, and seawater in Santala during 2012 compare to the mean values of groundwater (GW1) and precipitation (P1) data from southern Finland (Kortelainen, 2007). n = number of samples, Z (m) = sampling depth (m below ground surface), VDT (m) = median of vadose zone thickness (m) from four measurements in 2012, and std. = standard deviation.

Sampling site	n	Z (m)	VDT (m)	δ^{18}O (‰ VSMOW)				δ^2H (‰ VSMOW)			
				Min.	Mean	Max.	Std.	Min.	Mean	Max.	Std.
VO	4	6	1	−11.5	−11.4	−11.3	0.1	−81.2	−80.6	−80.0	0.5
Obs11	4	6	0	−11.2	−11.1	−11.0	0.1	−78.8	−78.2	−77.9	0.4
Obs10	4	6	1.9	−11.5	−11.2	−11.0	0.3	−80.6	−79.0	−77.5	1.6
Obs9	4	10	2.7	−11.4	−11.2	−11.0	0.2	−80.6	−79.7	−78.6	0.8
Obs8	8	9	6.6	−12.3	−11.6	−11.1	0.4	−86.9	−82.7	−78.9	3.0
Obs7	4	7	2.9	−11.9	−11.8	−11.1	0.4	−84.6	−82.1	−78.4	2.7
Obs6	8	8	1.4	−11.7	−11.4	−11.0	0.2	−82.7	−80.7	−78.5	1.6
Obs5	2	8	5.7	−11.7	−11.6	−11.5	0.1	−82.7	−81.8	−80.9	1.3
Obs4	3	10	4.9	−11.4	−11.4	−11.2	0.1	−80.7	−80.0	−79.0	0.9
Obs3	2	10	3.7	−11.1	−11.0	−11.0	0.1	−77.9	−77.9	−77.9	0.0
Obs2	4	10	1.6	−12.2	−11.6	−10.6	0.8	−86.1	−81.6	−76.9	4.7
Obs1	4	8	1.8	−11.3	−11.0	−10.8	0.2	−78.8	−77.6	−76.5	0.9
Obs12	1	4	1.9	−11.5	−11.5	−11.5		−79.9	−79.9	−79.9	
Lake water	3	1	1	−8.5	−7.5	−4.5	2.1	−63.9	−55.6	−45.0	9.6
Seawater	4	1	1	−7.9	−7.6	−7.1	0.4	−60.6	−57.7	−55.3	2.4
GW1				−13.9		−11.6		−100.3		−82.0	
P1				−14.6	−11.5	−9.1	0.3	−106.8	−82.0	−65.7	3.4

Table 4. Matrix of Pearson correlation coefficients of geochemical variables from all samples (number of samples, $n = 25$).

Variable	δ^2H	$\delta^{18}O$	KMnO₄	SO₄	K	Mo	Mg	I	Ba	Mn	Cu	Si	Li	Ca	Alk.	Sr	EC	pH	NO₃	Na
$\delta^{18}O$	0.99**	1.0																		
KMnO₄	0.77**	0.79**	1.0																	
SO₄	0.0	0.0	0.0	1.0																
K	−0.1	−0.1	0.0	0.95**	1.0															
Mo	−0.1	−0.1	0.0	0.87**	0.92**	1.0														
Mg	−0.1	−0.1	−0.1	0.64**	0.64**	0.53**	1.0													
I	−0.1	−0.1	0.0	0.65**	0.75**	0.65**	0.86**	1.0												
Ba	0.1	0.0	0.1	0.86**	0.86**	0.86**	0.61**	0.70**	1.0											
Mn	0.1	0.0	0.0	0.79**	0.78**	0.76**	0.46*	0.60**	0.76**	1.0										
Cu	0.0	0.0	0.2	0.85**	0.89**	0.90**	0.42*	0.56*	0.88**	0.61**	1.0									
Si	−0.4	−0.46*	−0.50*	0.40*	0.3	0.2	0.50*	0.42*	0.44*	0.44*	0.2	1.0								
Li	0.0	0.0	0.1	0.92**	0.92**	0.88**	0.63**	0.63**	0.72**	0.67**	0.83**	0.1	1.0							
Ca	−0.2	−0.2	−0.1	0.56**	0.64**	0.72**	0.48*	0.46*	0.42*	0.3	0.64**	−0.1	0.77**	1.0						
Alk.	−0.2	−0.2	−0.1	0.52**	0.61**	0.69**	0.52**	0.49*	0.4	0.2	0.59**	−0.1	0.74**	0.99**	1.0					
Sr	−0.2	−0.2	−0.1	0.57**	0.66**	0.72**	0.54**	0.52**	0.43*	0.3	0.61**	0.0	0.77**	0.99**	0.99**	1.0				
EC	−0.2	−0.2	−0.1	0.68**	0.74**	0.74**	0.63**	0.57**	0.55**	0.4	0.67**	0.1	0.81**	0.95**	0.92**	0.95**	1.0			
pH	−0.2	−0.2	−0.3	0.2	0.3	0.3	0.3	0.2	−0.1	−0.1	0.2	−0.3	0.50*	0.80**	0.83**	0.78**	0.68**	1.0		
NO₃	−0.1	−0.1	0.2	0.50*	0.59**	0.69**	0.2	0.3	0.47*	0.2	0.75**	−0.3	0.67**	0.86**	0.82**	0.81**	0.77**	0.56**	1.0	
Na	−0.1	−0.1	−0.1	0.4	0.3	0.1	0.40*	0.3	0.3	0.3	0.1	0.3	0.1	0.0	−0.1	0.0	0.3	−0.2	−0.1	1.0
Cl	−0.1	−0.1	−0.1	0.0	0.0	−0.1	0.1	0.0	0.1	0.0	−0.1	0.1	−0.1	−0.2	−0.2	−0.1	0.1	−0.3	−0.2	0.91**

**: correlation is significant at the 0.01 level (italic) and *: correlation is significant at the 0.05 level.

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Table 5. Principal component loadings from factor analysis and explained variances of geo-chemical variables and stable isotopes δ^2H and δ^{18}O of water samples. Factor loadings > 0.7 are marked as italic.

Parameter	Principal component (PC)			
	1	2	3	4
δ^2H	−0.08	−0.17	*0.86*	−0.05
δ^{18}O	−0.13	−0.14	*0.86*	−0.05
KMnO$_4$ (mg L^{-1})	0.19	0.12	*0.82*	0.03
SO$_4$ (mg L^{-1})	*0.86*	0.27	0.11	0.23
K (mg L^{-1})	*0.83*	0.40	−0.11	0.20
Mo (µg L^{-1})	*0.78*	0.47	−0.15	−0.13
Mg (mg L^{-1})	*0.75*	0.24	−0.07	0.28
I (µg L^{-1})	*0.75*	0.25	0.00	0.20
Ba (µg L^{-1})	*0.82*	0.17	0.23	0.26
Mn (µg L^{-1})	*0.75*	−0.27	0.40	0.26
Cu (µg L^{-1})	*0.81*	0.34	0.37	−0.10
Si (mg L^{-1})	*0.69*	−0.40	−0.43	0.19
Li (µg L^{-1})	*0.69*	0.59	0.16	0.06
Ca (mg L^{-1})	0.35	*0.93*	−0.04	−0.03
Alkalinity (mmol L^{-1})	0.38	*0.90*	−0.07	−0.09
Sr (µg L^{-1})	0.41	*0.88*	−0.05	−0.01
EC (mS m^{-1} at 25 °C)	0.46	*0.84*	−0.05	0.26
pH	0.04	*0.84*	−0.27	−0.26
NO$_3$ (mg L^{-1})	0.20	*0.79*	0.37	−0.14
Na (mg L^{-1})	0.20	−0.06	−0.05	*0.94*
Cl (mg L^{-1})	0.12	−0.19	−0.01	*0.95*
Explained variance	12.3	5.1	3.2	2.0
% of Explained variance	43.8	18.0	11.6	7.1
Cumulative % of variance	43.8	61.8	73.4	80.5

Table 6. Factor score from principal component loading contribute to each case (Observation wells and seasons, where 0412 and 0812 represent samples taken during April 2012 and August 2012, respectively). Positive factor scores are marked as italic.

Case	Principal component (PC)			
	1	2	3	4
Lake water_0812	−1.33	−0.06	*4.06*	−0.24
Obs1_0412	*0.84*	−1.22	−0.03	−0.86
Obs1_0812	*0.34*	−1.12	−0.15	−0.57
Obs2_0412	*0.25*	−1.00	*1.13*	−0.71
Obs2_0812	−0.12	−0.83	*0.75*	−0.35
Obs3_0412	−0.16	−0.62	−0.66	−0.89
Obs4_0412	−0.44	−0.40	−0.86	−1.13
Obs4_0812	−0.48	−0.47	−0.18	−0.76
Obs5_0412	−0.41	−0.58	−0.80	−0.85
Obs6_0812top	−1.34	*0.24*	−0.58	*1.87*
Obs6_0412mixed	−0.79	−0.16	−0.59	*2.43*
Obs6_0812mixed	−0.93	−0.28	*0.15*	*2.41*
Obs6_0812bottom	−0.66	−0.39	−0.36	−0.16
Obs7_0412	−0.97	−0.43	−0.87	−0.33
Obs7_0812	−1.14	−0.33	−0.20	−0.11
Obs8_0412mixed	−0.35	*1.60*	−0.04	−0.67
Obs8_0812top	−0.53	*1.85*	−0.59	−0.91
Obs8_0812mixed	−0.32	*1.22*	−0.16	−0.64
Obs8_0812bottom	−0.46	*1.89*	−0.74	−0.49
Obs9_0412	*1.80*	−0.79	−0.54	*0.37*
Obs9_0812	*1.27*	−0.76	0.01	*0.75*
Obs10_0412	*2.02*	*1.52*	*0.64*	*0.60*
Obs10_0812	*1.48*	*1.74*	*0.74*	*0.26*
Obs11_0412	*1.36*	−0.54	−0.13	*0.35*
Obs11_0812	*1.05*	−0.10	−0.01	*0.62*

Table 7. Median values of variables in each cluster from groundwater samples (number of samples, $n = 24$) taken during spring (April) and summer (August) 2012.

Cluster	1	2	3	4	5	6
Number of samples	7	3	4	4	2	4
Eh (mV)	292	312	273	*201*	267	219
O_2 (mg L^{-1})	11.3	8.77	5.89	5.79	*1.20*	2.68
CO_2 (mg L^{-1})	3.0	13.0	18.0	1.5	16.0	*31.0*
pH	6.6	6.2	*5.7*	7.8	7.1	6.6
EC (mS m^{-1} at 25 °C)	4.74	12.9	4.79	22.7	*29.9*	15.3
Alkalinity (mmol L^{-1})	0.22	0.23	0.16	2.06	*2.17*	0.84
$KMnO_4$ (mg L^{-1})	0.6	1.8	4.90	1.5	*5.0*	1.9
K (mg L^{-1})	0.55	0.59	0.66	0.94	*4.19*	1.88
Ca (mg L^{-1})	4.28	8.00	3.33	38.5	*45.2*	14.1
Mg (mg L^{-1})	0.93	1.40	0.85	1.96	2.82	*3.56*
Na (mg L^{-1})	2.46	*12.7*	2.94	2.41	6.63	7.89
Cl (mg L^{-1})	2.78	*25.0*	3.21	1.95	5.48	8.47
SO_4 (mg L^{-1})	7.18	8.40	8.60	8.22	*29.3*	17.0
NO_3 (mg L^{-1})	< 0.20	0.30	0.90	2.95	*5.40*	0.39
Si (mg L^{-1})	5.37	5.70	7.15	4.73	6.52	*8.87*
F (mg L^{-1})	< 0.10	< 0.10	0.11	< 0.10	0.40	*0.53*
Sr (µg L^{-1})	15.5	29.9	20.9	131.0	*144.5*	64.6
I (µg L^{-1})	3.69	3.27	3.14	5.68	*14.5*	10.3
Li (µg L^{-1})	0.61	0.16	0.14	2.05	*6.10*	2.61
Mn (µg L^{-1})	0.25	1.22	17.3	0.15	*48.5*	21.6
Cu (µg L^{-1})	< 0.10	< 0.10	0.52	0.44	*3.55*	0.79
Al (µg L^{-1})	8.02	13.8	*121.3*	3.95	1.68	35.8
As (µg L^{-1})	0.10	< 0.05	0.10	0.47	*0.79*	0.18
B (µg L^{-1})	11.3	5.69	9.43	5.93	*14.0*	15.0
Ba (µg L^{-1})	1.30	3.14	5.01	1.64	*14.7*	7.25
Cr (µg L^{-1})	< 0.20	< 0.20	0.25	*0.60*	0.29	< 0.20
Mo (µg L^{-1})	0.14	0.09	0.28	0.45	*2.08*	0.55

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Table 8. Changes of selected dissolved concentrations (%) between seasons in different observation well locations. Numbers in the first row (1 to 11) represent observation well numbers (Obs1 to Obs11). Positive numbers are marked as italic, indicates increasing of dissolved concentrations.

| Variable	Apr 2010 to Apr 2012				Apr 2010 to Aug 2012				Apr 2012 to Aug 2012								
	1	8	10	11	1	8	10	11	1	8	10	11	2	6	4	9	7
pH	9	4	3	16	5	4	3	17	–3	0	0	1	–2	–2	–1	–3	–2
EC	23	67	4	–9	13	–3	39	7	–8	–17	2	7	–4	–12	5	–3	1
$KMnO_4$	–47	23	–26	–57	–73	46	15	33	–49	19	56	54	14	556	200	28	66
Alkalinity	21	102	7	46	36	63	14	61	12	–19	6	10	40	10	27	–7	13
Ca	8	75	1	–12	–2	48	9	1	–9	–15	8	15	–5	–19	4	–12	–4
Mg	10	16	33	–4	–1	6	7	0	–10	–9	–20	4	–10	–17	0	–12	–5
K	–1	2	2	–16	–43	–34	–26	–25	–42	–35	–27	–11	–27	–36	–21	–31	–15
Na	9	–14	2	–4	5	–4	–39	–9	–3	12	–40	–5	–4	–14	0	–5	0
Cl	–6	–31	–25	–3	–11	–8	–35	10	–5	34	–13	13	–22	–12	13	19	3
NO_3	–24	–29	–42	45	–31	–34	6	100	–9	–8	84	38	2	–32	–73	0	–52
SO_4	22	–9	19	–42	4	–7	–5	–48	–14	2	–20	–10	3	–8	6	–6	–6
Al	107	–72	–45	–58	–2	–85	–71	–88	–53	–47	–46	–72	–33	–32	100	–53	–62
Mn	–33	–93	283	–60	–66	–53	–23	–80	–49	586	–80	–50	–41	150	800	–59	60
Sr	22	85	–9	–11	–16	26	–13	–8	–31	–32	–5	4	–32	–34	–13	–23	–11
F	200	0	0	22	0	–50	–40	0	–67	–50	–40	–18	–55	–77	0	–35	–58
Fe	29	–67	0	0	–58	–67	0	0	–68	0	0	0	–20	0	0	–67	0
P	0	0	116	0	360	0	180	0	360	0	30	0	0	0	460	0	0
Si	15	–6	17	–11	8	–1	13	–13	–6	5	–4	–3	3	2	1	–6	1
Cd	60	0	100	–42	60	0	50	–58	0	0	–25	–29	25	50	100	–17	0
Co	34	488	367	–48	–8	–37	167	–81	–31	–89	–43	–64	–37	–56	–97	–46	–97
Cr	0	–42	220	100	0	72	150	0	0	195	–22	–50	67	0	0	0	0
Cu	–38	–42	16	–42	–51	–59	54	–28	–22	–31	34	24	–26	0	240	72	0
Ni	36	–37	587	–42	–40	120	158	–78	–56	251	–62	–62	–84	–88	–85	–50	–77
Pb	–58	260	0	0	–58	0	140	340	0	–72	140	340	–63	143	340	260	100
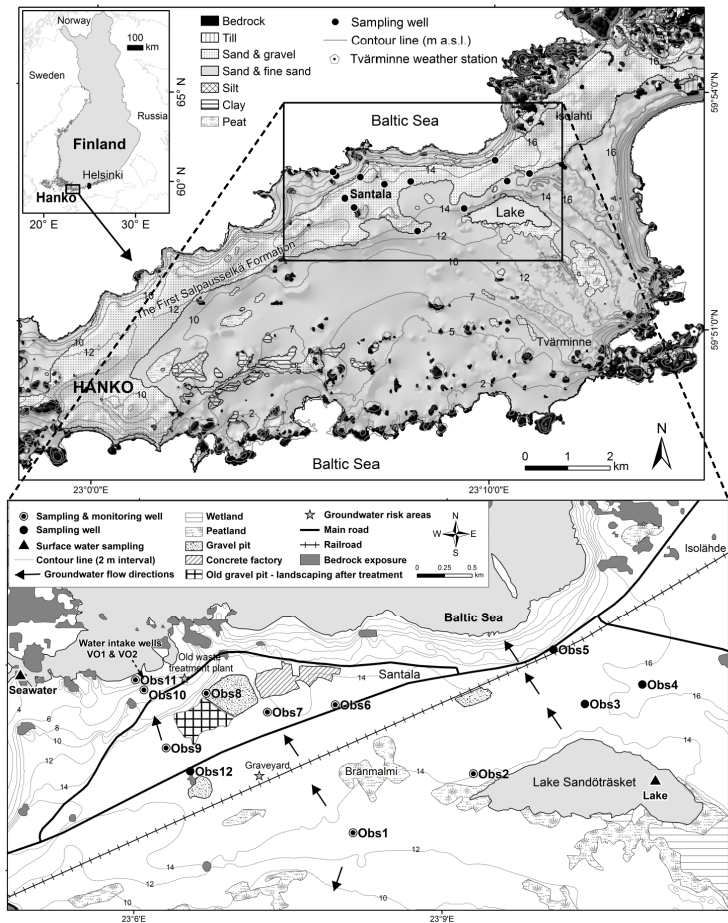
Figure 1. Location and Quaternary geological deposit map of the study area in the eastern Baltic Sea region.
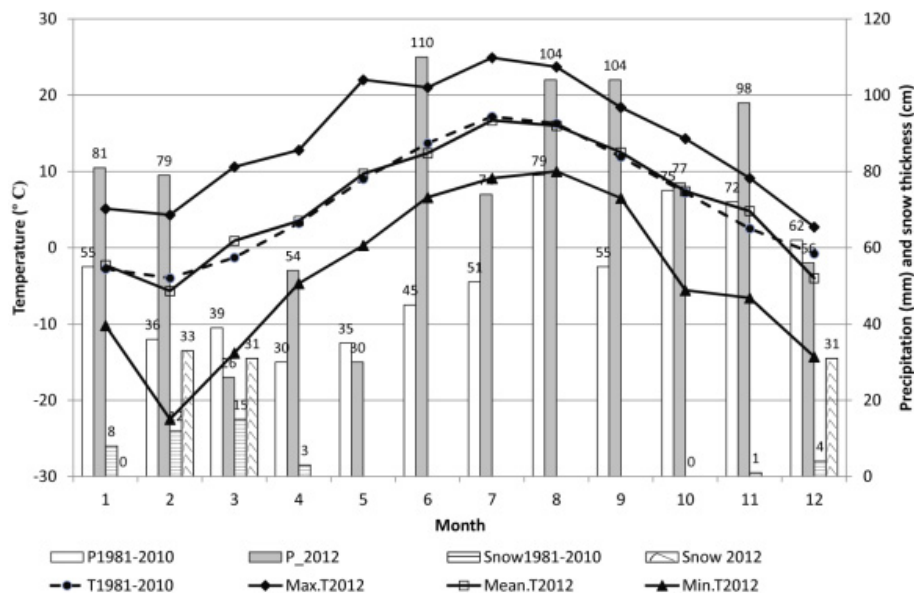
Figure 2. Monthly maximum, mean, minimum temperatures (solid lines, in °C), monthly precipitation (gray bar, in mm, as rainfall and snow) and snow thickness (wavy bar, in cm) in the Tvärminne weather station in Hanko during January–December 2012. Mean monthly temperature (dash line, in °C), monthly precipitations (white bar, in mm, as rainfall and snow) and snow thickness (straight line bar, in cm) of years 1981–2010, are plotted for comparison. Data are received from FMI (2013).
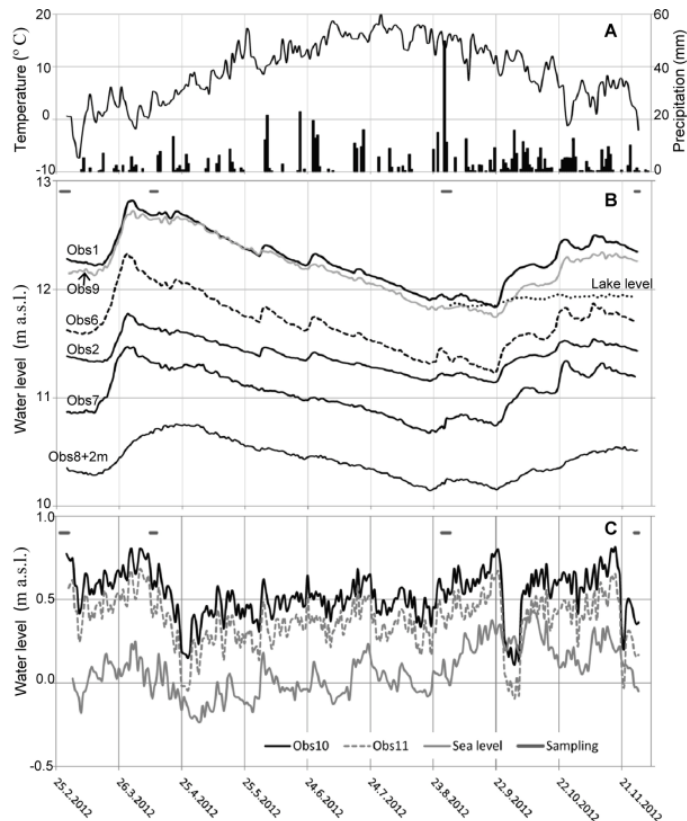
Figure 3. (A) Daily temperature (line) and precipitation (bar) during March–November 2012 from the Tvärminne weather station, Hanko (FMI, 2013). **(B)** Groundwater levels from monitoring wells during the same periods for wells locate in the mainland and lake level during September–November 2012. **(C)** Groundwater levels from wells locate nearby the coastline and sea level data. Groundwater level of Obs8 was shifted up 2 m in order to get the all data fit in one figure. The sample periods present on the top of **(B)** and **(C)**.

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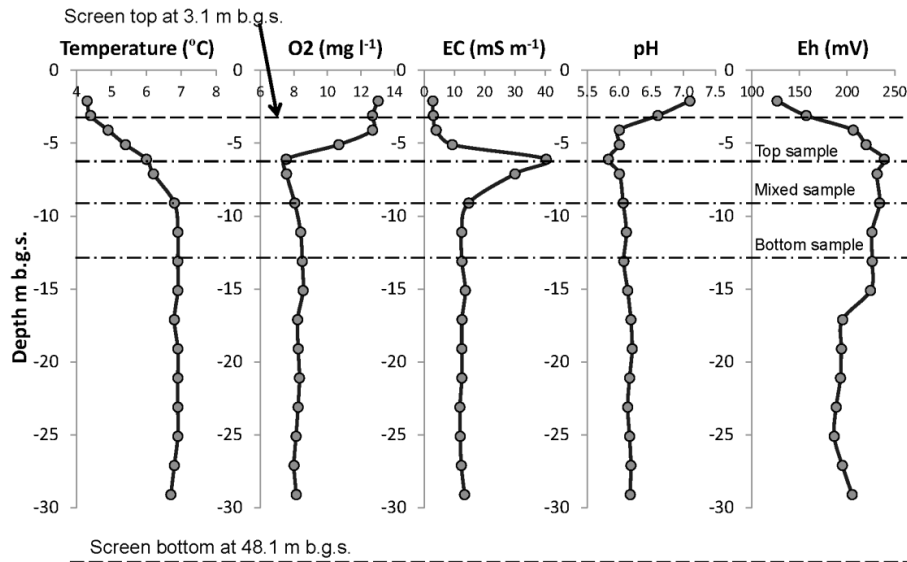
Figure 4. Depth profile of parameters: temperature, dissolved oxygen (O_2), electrical conductivity (EC), pH and redox potential (Eh), in the well screen section of Obs6 (50 m from the highway) during spring 2012. m b.g.s = metre below ground surface.

Confronting vicinity of the surface water and sea shore

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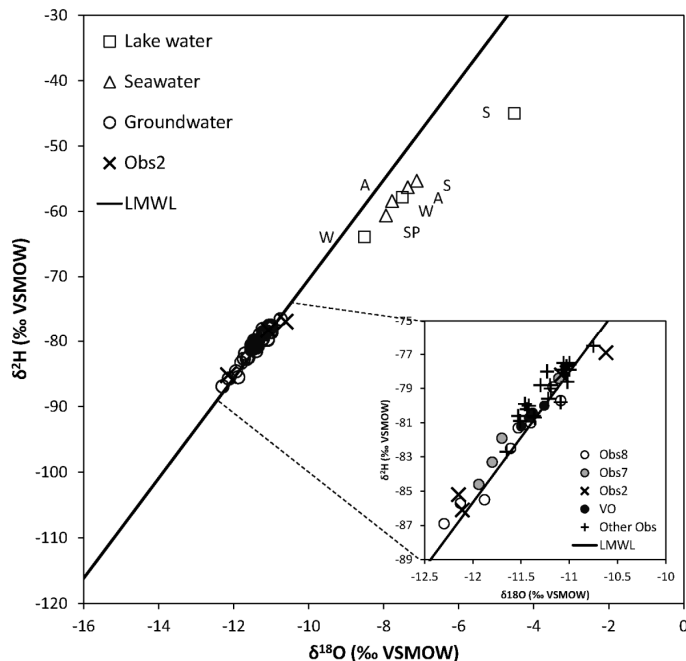
Figure 5. Plot of the δ^2H and δ^{18}O data for lake water, seawater, groundwater from the observation wells (Obs), and water intake well (VO) from Santala during 2012 with the Finnish local meteoric water line (LMWL) for comparison. Labels at surface water indicate the sampling period: S = summer, SP = spring, A = autumn and W = winter. Locations of water samplings are showed in Fig. 1.

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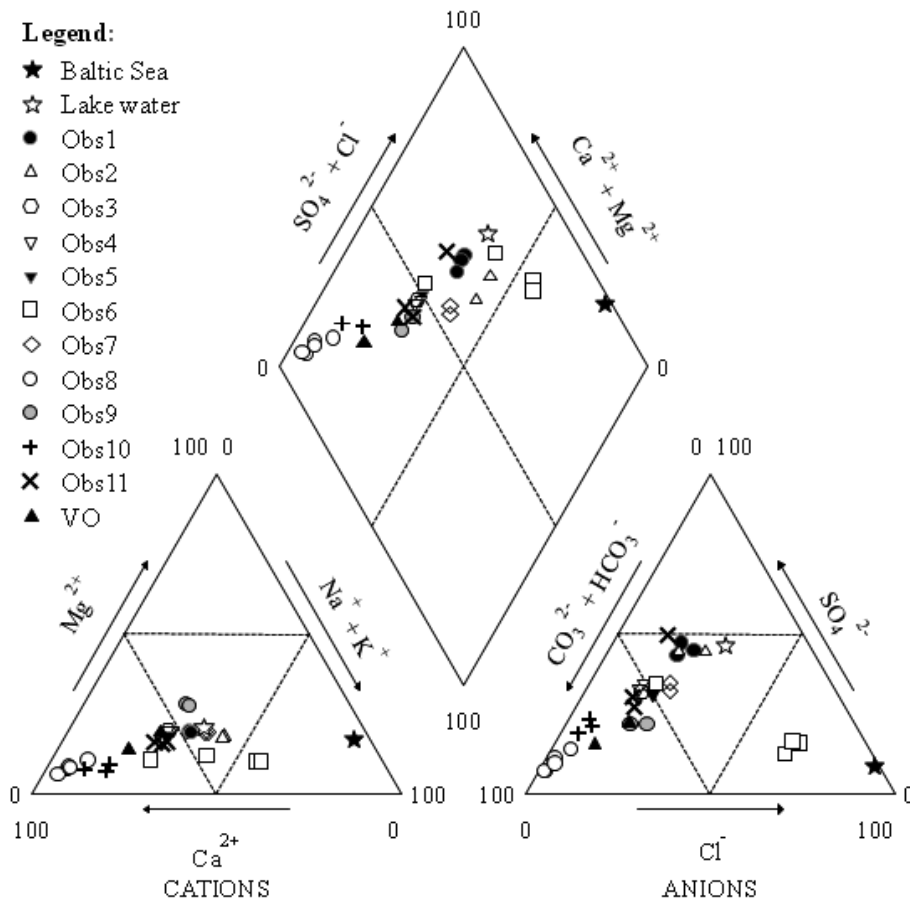Abstract | Introduction

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Figure 6. Piper diagram showing the major ion chemistry of groundwater and surface water samples from the Santala, Hanko.

Figure 7. Hierarchical clustering results (dendrogram) of water samples ($n = 25$) taken during spring (April) and summer (August) 2012.