Investigating submarine groundwater discharge in the Gulf of Bothnia using Ra isotopes

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Introduction

Forsmark, on the southern shore of the Gulf of Bothnia, Baltic Sea (Figure 1), applied to host an underground repository for radioactive waste. **Submarine groundwater discharge (SGD)** may influence the risk assessment, thus it should be characterized and quantified while assessing the safety of coastal repositories. Yet, in Sweden overall, coastal loads to the Baltic Sea Drainage Basin from small unmonitored catchment areas remains uncertain [1].

^{223,224}Ra ($t_{1/2}$=11.43, 3.66 d) isotopes can be used to quantify submarine groundwater discharge (SGD) because they:

- are usually enriched in groundwater (in dissolved and particle-adsorbed form) with respect to seawater
- behave conservatively once released into a sufficiently saline marine environment
- have $t_{1/2}$s ≠ ^{226,228}Ra, which allows estimation of SGD and residence time of coastal water on many time scales.

Besides SGD, Ra is **enriched** in coastal seawater by:

- riverine discharge in dissolved form and by desorption from suspended sediments
- diffusion from seabed sediments.

and **removed** by:

- exchange with open sea waters
- radioactive decay [2].

This method of SGD flux calculation involves determining Ra supply & removal, assuming excess Ra as SGD-sourced.

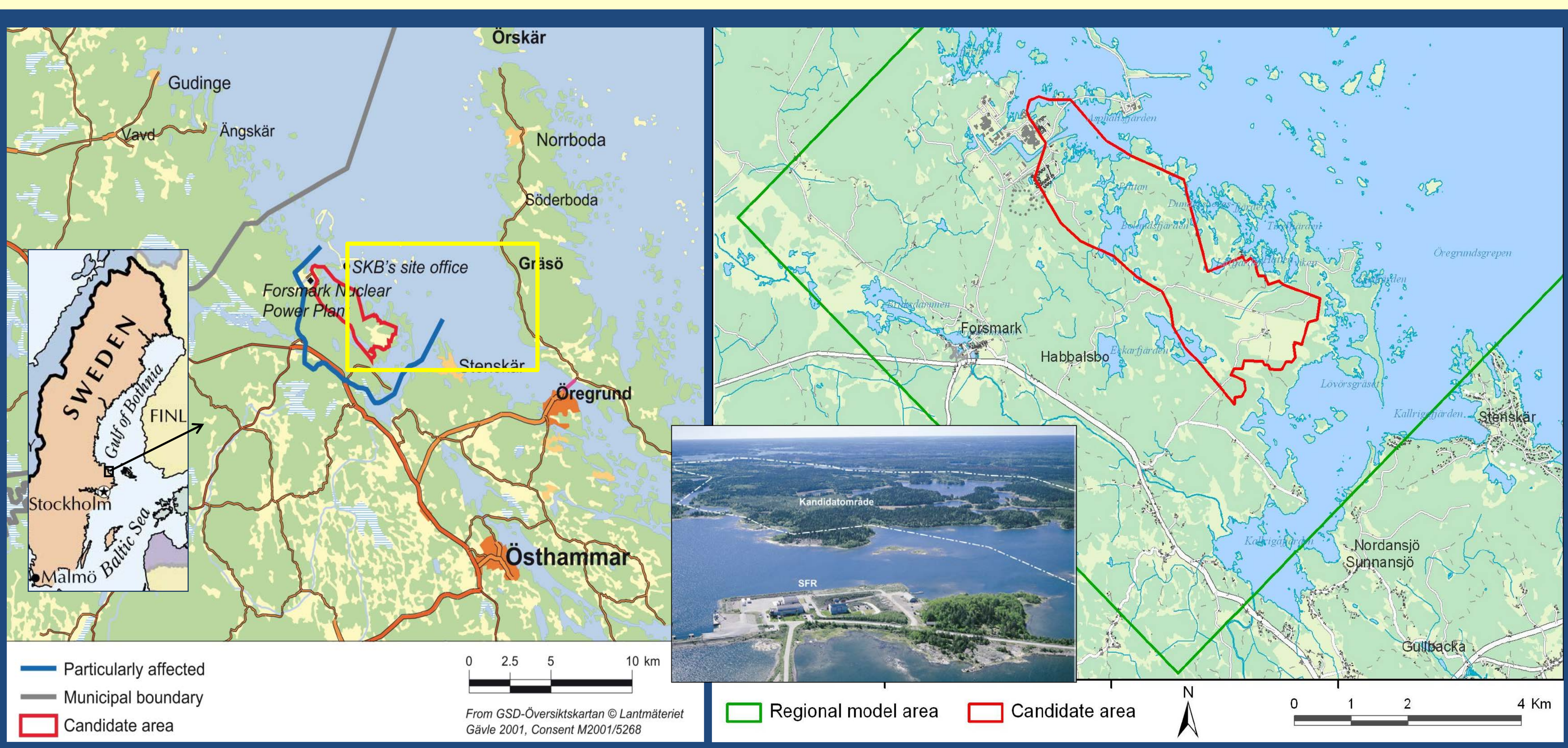

Figure 1: Views of the study site, multiple scales, yellow square shows location of region enlarged in Figure 5.

Site background

The bedrock is dominated by meta-granite (~1.87 Ga). Regional deformation zones occur mainly as steeply dipping fracture zones which strike WNW-ENE & NW-SE. In the SE of the candidate volume, fracture zones dip gently to the SE. A shallow bedrock aquifer horiz. fractures/sheet joints rests in the upper (0-150 m) of the bedrock. Here meteoric water mixes with groundwater then is transported laterally to the NE and discharged into the Baltic Sea (Figure 2) [3].

The Geological Survey of Sweden (SGU) mapped Quaternary sediments deposited on the seabed outside Forsmark. These include glacial and post-glacial clays, sometimes overlain by gyttja [4]. Additionally, sediment echo sounding by the SGU revealed an esker of coarse-grained & permeable sediment running along the seabed [5]. These sediments are notable because the clays may supply Ra, while the esker may provide a transport pathway for SGD.

In November 2013,three sites along the shore were sampled for short-lived Ra isotope measurements. Relatively high ^{224}Ra activities **(35.9 ± 1.8, 44.5± 3.8, and 16.6± 1.1 dpm/100L)** prompted a larger coastal sampling.

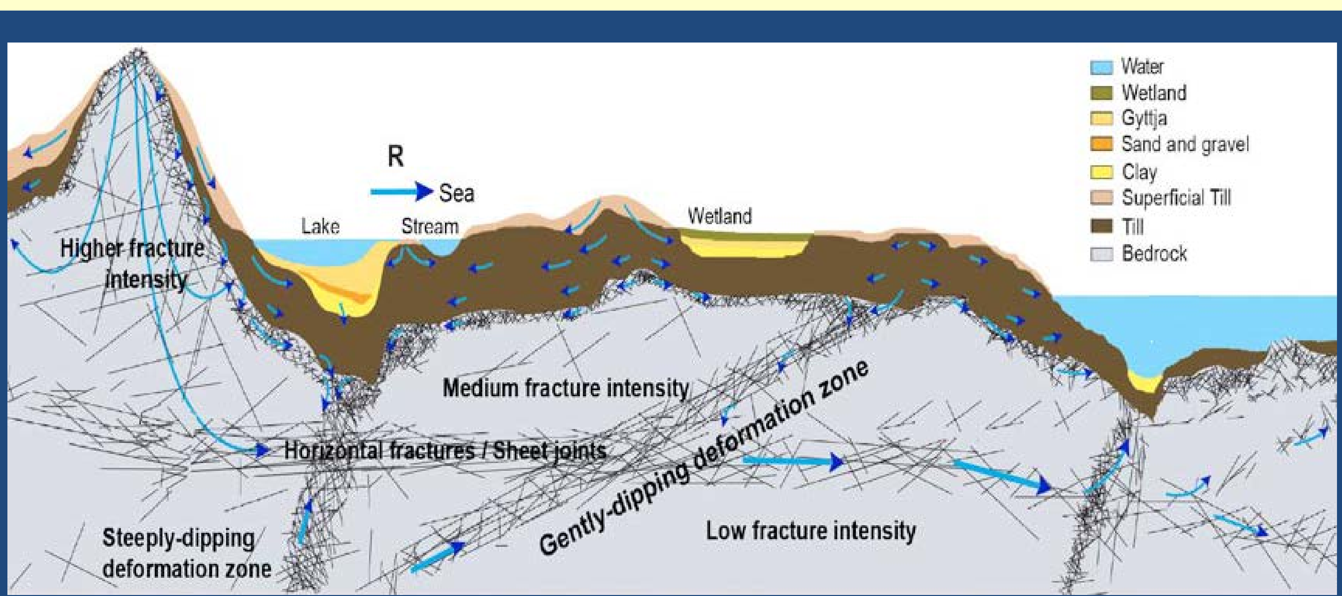

Figure 2: Cartoon cross-section fracture zones and water migration paths. Note intersection of shallow bedrock aquifer with steeply dipping fractures [3].

The goal:

To collect data on short-lived Ra-isotopes and begin to quantify SGD in the Forsmark area. Here we present results of the offshore distribution of ^{223,224}Ra.

Sampling and methods

In late April 2014, water samples were collected from:

- Three, five-point transects along the coast (1 m depth); at two sites, samples (50 L) also collected from 10 m depth
- Four points Kallrigafjärden (bay) (50 L)
- Two near-shore sites (50 L)
- Five shallow wells (20 L) and two deeper fractures (50 L)

At each station, salinity profiles were collected using a YSI 556 multiparameter probe while water was pumped through a 0.5 µm cartridge filter. Samples of 250 mL each were collected for $\delta^{18}O$ analyses. The 50 L samples were passed through a column of MnO_2-impregnated fibers to extract Ra. The fibers were measured for short-lived Ra isotopes ($^{224, 223}$Ra) using a Radium Delay Coincidence Counter (RaDeCC) [6]. ^{224}Ra activities were measured within three days of collection and ^{223}Ra after seven days. The $\delta^{18}O$ compositions were determined using a Picaro Isotope Water Analyser L2140-i with cavity ring down spectroscope; data was normalized so that the difference between SLAP and SMOW was -55.5‰.

Results

Nearshore $^{224, 223}$Ra activities are comparable to those observed in 11-2013, **38.58±2.66, 3.74±0.31 and 27.11±1.79, 2.78±0.18** dpm 100L^{-1}, respectively. Figure 3 shows $^{224, 223}$Ra activities as a function of distance offshore for the offshore transects and Kallrigsfjärden bay. In Kallrigsfjärden, $^{224, 223}$Ra activities are relatively high, and salinities low (min. 1.71 ppt). In the offshore transects $^{224, 223}$Ra concentrations decrease with distance offshore (Figure 4). These follow a trend which appears to break beyond ~4.5 – 5.5 km from the coast. This interpretation is supported by the corresponding $\delta^{18}O$ figures. Salinity ranges **4.63 - 4.82 ppt**, decreases slightly with distance from shore, and increases slighly with depth (Figure 5). Two samples collected from 10 m depth have $^{224, 223}$Ra activities of 4.9±0.5, 0.2±0.03 and 4.5±0.4, 0.1±0.02 dpm 100L^{-1} which correspond to 1 m depth samples with 4.8±0.4, 0.4±0.06 and 4.5±0.4, 0.1±0.02 dpm 100L^{-1}, respectively. Average $^{224, 223}$Ra activities of the soil tubes were 181.3±12.7 and 6.4±0.9 dpm 100L^{-1}, respectively.

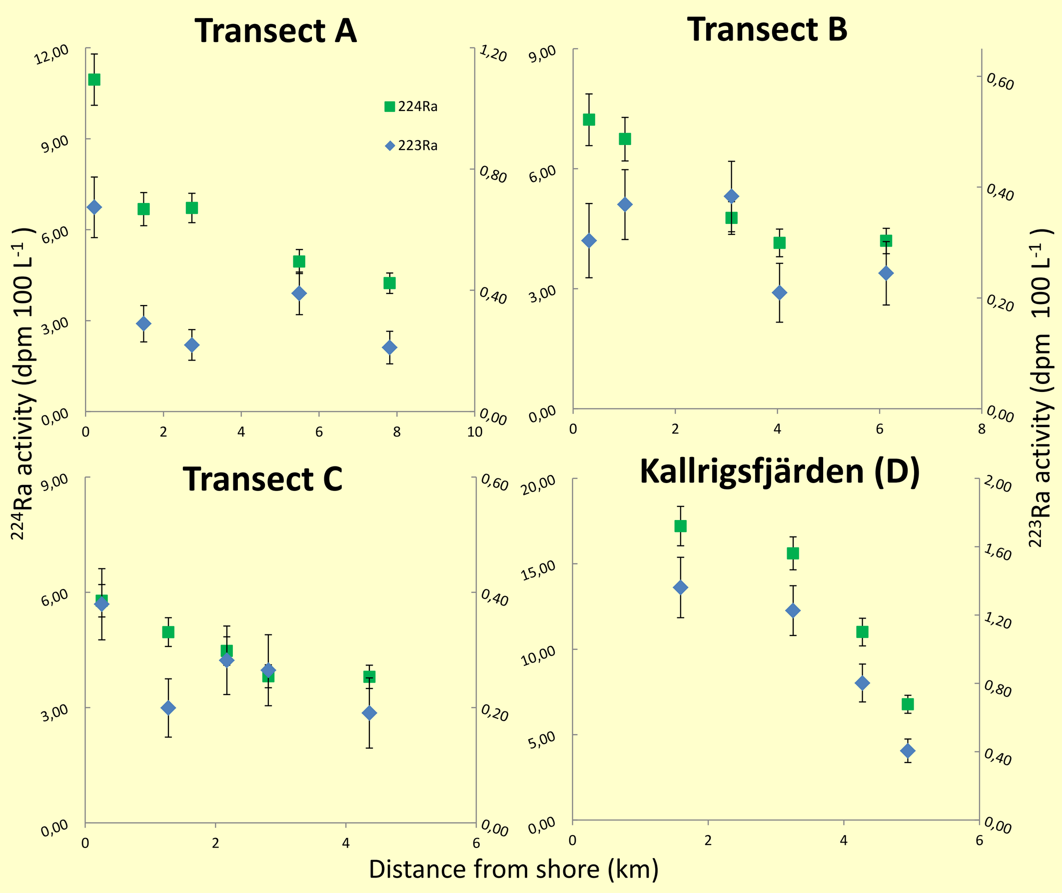

Figure 4: ^{224}Ra and ^{223}Ra activities & uncertainties as functions of distance from shore for each transect. A) **10.9±0.8 - 4.2±0.3** and **0.7±0.1 - 0.2±0.05**, B) **7.2±0.6 - 4.2±0.3** and **0.3±0.07 - 0.2±0.06**, C) **5.8±0.4 - 3.8±0.3** and **0.4±0.06 - 0.2±0.06** dpm 100L^{-1}, and D) **17.2±1.2 - 6.8±0.5** and **1.36±0.18 - 0.41±0.07** dpm 100L^{-1}, respectively.

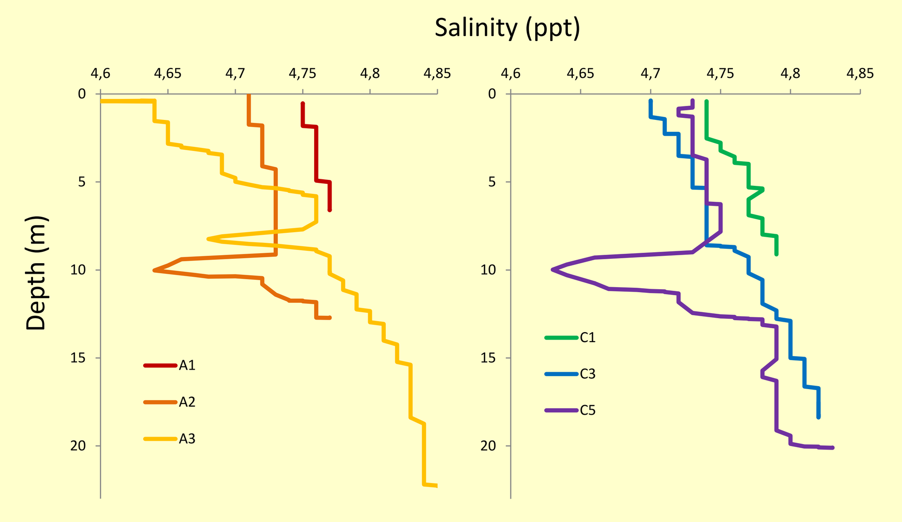

Figure 5: Salinity profiles from three stations in each transect A and C.

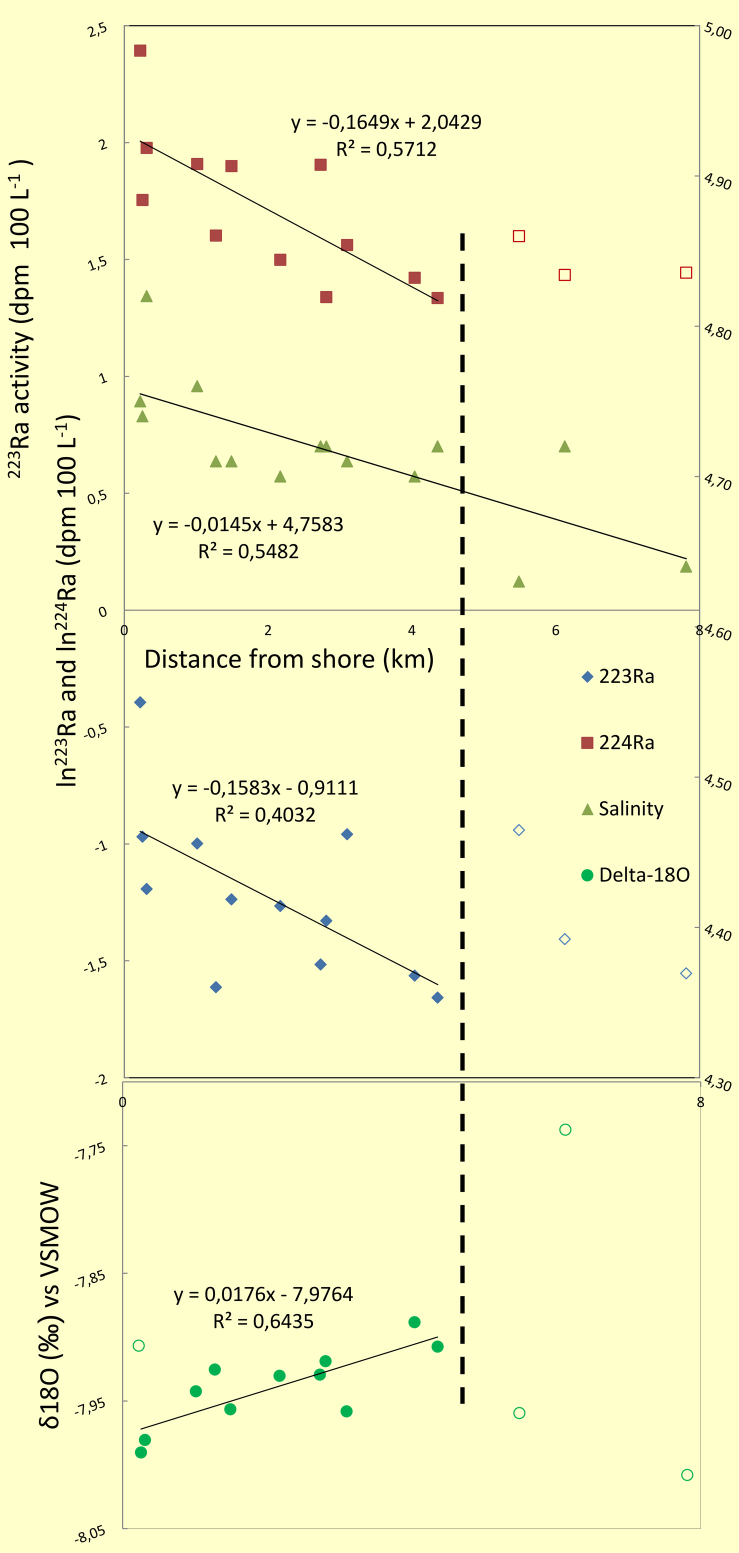

Figure 4: Natural log of ^{223,224}Ra transects A,B, and C compiled plotted alongside salinity (1m) and and $\delta^{18}O$ as a function of distance offshore for each transect.

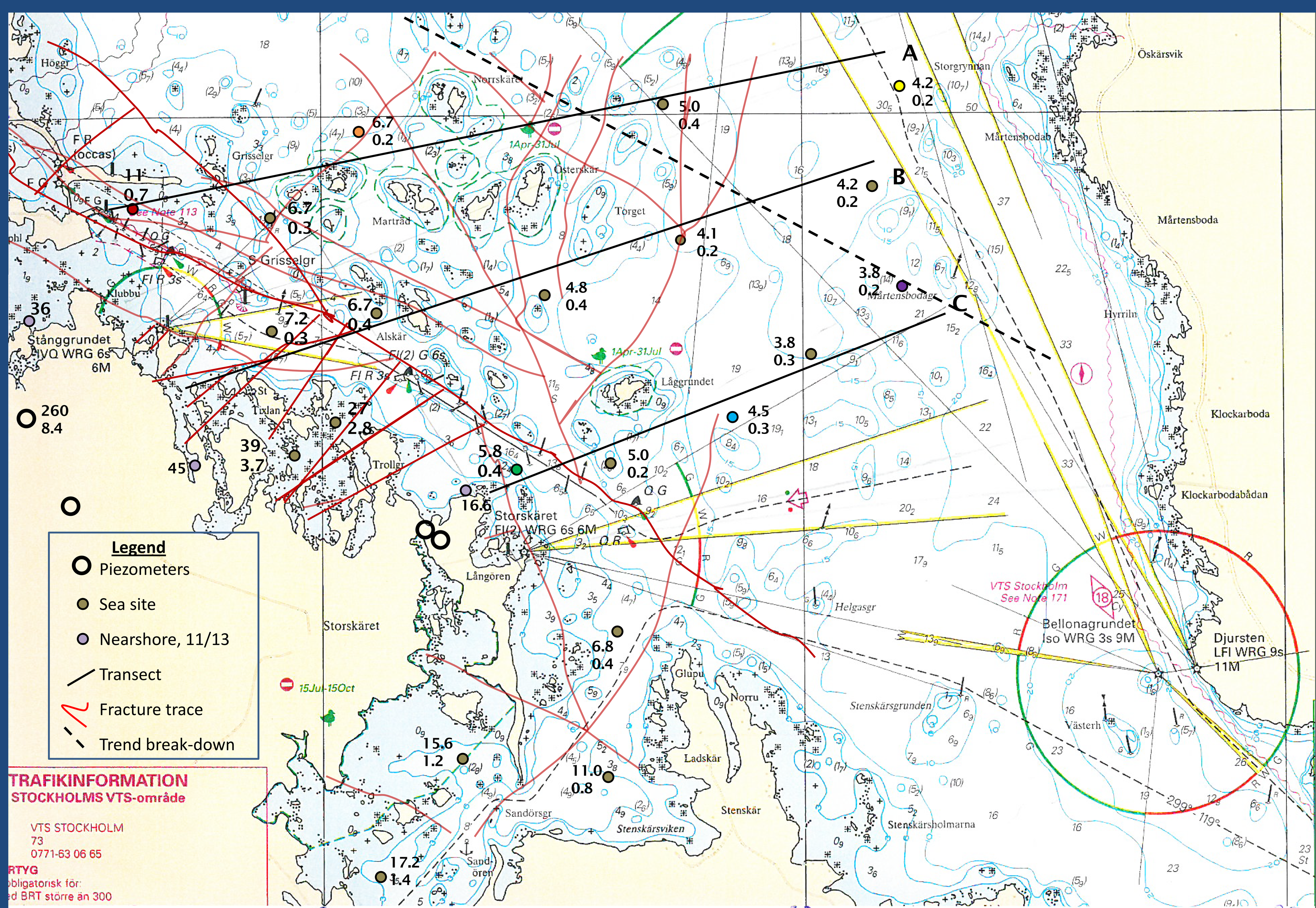

Figure 5: Map showing locations of transects and Kallrigsfjärden, sampling stations and respective ^{224}Ra (top) and ^{223}Ra (bottom) activities (dpm 100 L^{-1}), fracture networks, and line corresponding to breakdown of Ra trend.

Conclusions

- ^{223,224}Ra activity profiles show a trend reminiscent of that seen off the South Atlantic Bight [7].
- Activities are higher in the bay, but possibly due to desorption of Ra from sediments contributed by a nearby stream.
- Ra activities are higher in groundwater: *could* support a flux of Ra to sea.
- Cannot discriminate whether Ra activities are due to SGD or to diffusion from clay sediments on the seabed.

Future work

The authors will progress to estimate SGD in the area by:

- Continuing to analyze samples from these sites
  - $\delta^{18}O$, $\delta^{2}H$, Sr, and major cations
  - $^{226, 228}$Ra (minimum influence from bottom sediments)
- Collecting sediment cores for diffusion experiments to measure influence from sediments
- Measuring Rn activities to distinguish influence from springs along shore

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