Morphological and chemical features of submarine groundwater discharge zones in the south-eastern part of the Baltic Sea

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The complex study of the bottom sediments and near-bottom water layer of the Gdansk Deep revealed the Ca maximum anomaly, marking the discharge of the Oxford-Titonian aquifer. The discharge zone is associated with gas-saturated sediments, which is caused by a common pathway of aqueous and gaseous fluids upraise along tectonic faults. The detailed survey of a seabed topography and acoustic survey of the sediments upper layer helped to specify the area of gas-saturated sediments and the size of the pockmarks. *KEYWORDS:* Submarine groundwater discharge; pockmark; Gdansk Deep; ground water; bottom sediments.

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1. Introduction

The Gdansk Deep is the end body of groundwater flow from the adjacent part of the landmass [*Uścinowicz et al.,* 2011]. The system of faults and palaeochannels contributes to groundwater discharge through the glacial and fluvio-glacial sediments to the surface [*Otmas et al.,* 2006; *Petrov,* 2010; *Triponis,* 1973; *Zagorodnyh et al.,* 2011]. Usually, fault zones are associated with gassy (methane-saturated) sediments [*Blazhchishin,* 1998; *Sviridov,* 1990; *Sviridov and Emelyanov,* 2000]. Interacting with the Holocene silt, groundwater creates geochemical anomalies in sediments, manifested with a redistribution of chemical elements. Such geochemical anomaly was found at the eastern slope of the Gdansk Deep in the area of gas (methane) saturated sediments and pockmarks [*Krek et al.,* 2020]. The aim of this study was to investigate submarine groundwater discharge on the bottom of the Gdansk Deep from Jurassic and Cretaceous aquifers.

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2. Brief Description of Aquifers and Submarine Groundwater Discharge Zones

According to previous estimates the groundwater discharge into the Baltic Sea is considerably less than the surface water runoff, namely about 1.5–2% of total surface runoff [*Grigyalis,* 1991; *Gudelis and Emelyanov,* 1976; *Zektser and Kudelin,* 1965] and up to 9% of ionic surface runoff [*Tersiev,* 1992].

The intensity of groundwater discharge for different aquifers varies considerably, depending on geological structure, fault zones, seismic activity, and seabed topography [*Lidzbarski,* 2011]. Such discharge can occur either at a coastal cliff or at a submarine coastal slope.

Quaternary aquifers include unconfined groundwater of super-moraine sediments and low-pressure aquifers of inter-moraine fluvio-glacial sediments. These waters tend to discharge in a bigger volume

Figure 1. Drainage discharge of Quaternary groundwaters at the beach near the village of Sinyavino (western coast of the Sambia Peninsula, Kaliningrad oblast).

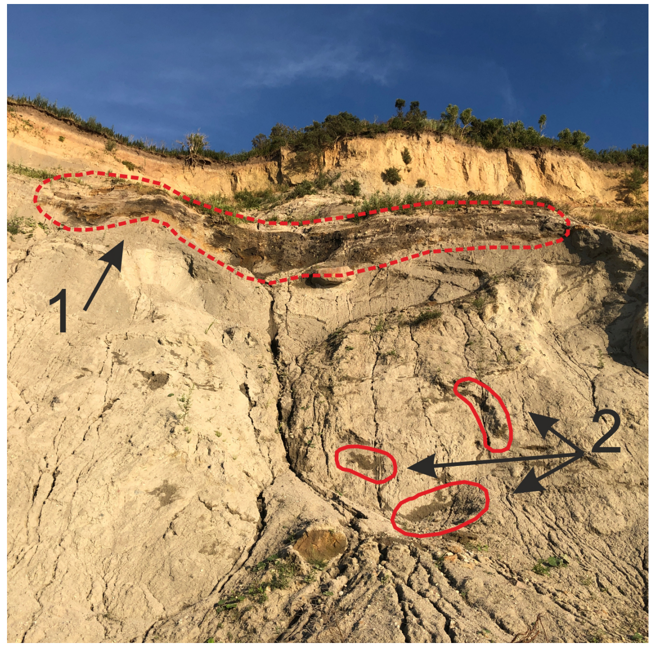

Figure 2. Drainage discharge of Quaternary groundwaters at the beach near the village of Sinyavino (western coast of the Sambia Peninsula, Kaliningrad oblast).

into river valleys, ravines, etc., reaching the sea with surface runoff. The Quaternary aquifers discharge is often drained (Figure 1), which is related to the shore protection activities performed earlier. The Quaternary waters may appear at the coastal cliff (Figure 2) and submarine coastal slope in case of an absence of an erosion network on the shore. In this case, Quaternary water discharge is of low intensity.

The Neogene sediments in the Kaliningrad oblast are poorly represented, the confined to them aquifer is closely associated with the Quaternary sediments and is drained by rivers or discharged at an abrasion coastal cliff (city of Svetlogorsk–Filino village).

Based on the peculiarities of the hydrogeological structure [*Kondratas,* 1970; *Zagorodnykh,* 2011], the submarine coastal slope of the Sambia Peninsula is the most likely place for the submarine water discharge from the zone, where the active water exchange of Paleogene, Cretaceous, Jurassic aquifers takes place.

Two horizons are distinguished in the Paleogene aquifer system: the upper (Oligocene-Pliocene), and the lower (Paleocene-Eocene) [*Zagorodnykh,* 2011].

The Oligocene-Pliocene aquifer occurs on the underwater shore slope only in the western part of the Sambia Peninsula (sporadically, at depths from 0 to 5–10 m). The weak water pressure, low water-holding capacity and high thickness of overlying mobile sands do not allow the tracing of drainage water discharge on the seabed.

The Paleocene-Eocene aquifer is widespread, particularly on the northern underwater shore slope of the Sambia Peninsula, where it emerges at depths of 12–15 to 25–30 m [*Zagorodnykh,* 2011]. The special feature of Paleogene waters is the high iron content. As a result of previous research in the area of the Kupalny Cape, anomalies were found in the distribution of some characteristics of seawater, namely the decrease in concentrations of chlorine ion, sodium ion, the increase in concentration of sulphate ion, decrease in water hardness, etc. [*Mikhnevich et al.,* 2013; *Tupeiko,* 2012]. In general, the waters of the Paleogene horizons are fresh, calcium hydrocarbonate with salinity of 0.2–0.8 g/l [*Zagorodnykh,* 2011].

Two aquifers are also identified in the Cretaceous sediments. *The Upper Cretaceous (Campanian-Masstrichtian) aquifer* is widespread both throughout the Sambia Peninsula, and on the underwater shore slope west of the Curonian Spit. According to *Lidzbarski* [2011], the extensive aquifer up to 100–150 m thick, can be traced in the Creta-

Table 1. Sampling Points List

Station	Latitude, N	Longitude, E	Depth, m	Type of sample
37030	55°22.706′	19°43.650′	92.8	
37031	55°21.877′	19°44.661′	92.6	
37032	55°21.172′	19°45.552′	92.6	
37033	55°20.583′	19°46.287′	91.6	
37035	55°19.283′	19°47.900′	87.0	
37036	55°18.625′	19°48.763′	81.0	
37042	54°57.857′	19°26.917′	98.7	
37043	54°57.186′	19°26.863′	98.3	Bottom sediments
37044	54°56.629′	19°27.038′	98.4	
37045	54°56.067′	19°27.016′	98.8	
37046	54°55.498′	19°26.937′	99.1	
37047	54°55.040′	19°27.143′	99.0	
37048	54°54.492′	19°27.053′	99.0	
37049	54°45.501′	19°29.170′	93.9	
37050	54°44.862′	19°29.246′	92.3	
37051	54°44.506′	19°29.439′	91.4	
37052	54°44.106′	19°29.316′	90.8	
37053	54°43.657′	19°29.412′	89.6	
37054	54°43.323′	19°29.397′	89.3	
37055	54°42.957′	19°29.432′	88.3	
37056	55°19.898′	19°47.044′	89.0	
37057	55°18.669′	19°48.627′	82.0	
47004	55°20.872′	19°46.710′	98.1	Near-bottom water, CTD
51115	55°21.877′	19°44.661′	114.0	Water (layers: near-bottom, 2 m above bottom, 5 m above bottom, cold intermediate layer, upper mixed layer), CTD

ceous sediments of the Gulf of Gdansk. Groundwater up to a depth of 100 m is fresh, sodium hydrocarbonate with mineralisation of 0.3–0.8 g/l, less frequently calcium hydrocarbonate. Salinity increases to 0.94–1.11 g/l at depths of 100–150 m, waters tend to be a transitional type from hydrocarbonate to chloride; the saline waters are sodium chloride-hydrocarbonate and located at the Sambia Peninsula.

The Aptian-Cenomanian aquifer is ubiquitous. These waters are of the chloride or hydrocarbonate sodium type with a mineralisation of 2–4 g/l.

Two aquifers are formed in the Jurassic sediments. *The Oxford-Tithonian aquifer* is confined to the upper part of the Jurassic section. The water is saline (from 12 to 17 g/l), of a sodium chloride type. *The Gettang-Bathonian aquifer* is confined to the lower part of the Jurassic section. The aquifer contains high-pressure water. The water is sodium chloride, its salinity is 13–19 g/l. The Jurassic aquifers discharge into the Baltic Sea in the northern part of the Gulf of Gdansk; the outcrops of water-bearing sediments are located at depths from 50 to 100 m.

3. Materials and Methods

The research included the geoacoustic survey of the seabed surface and upper sediment layer, near-bottom water and bottom sediments sampling, and CTD sounding (Figure 3, Table 1).

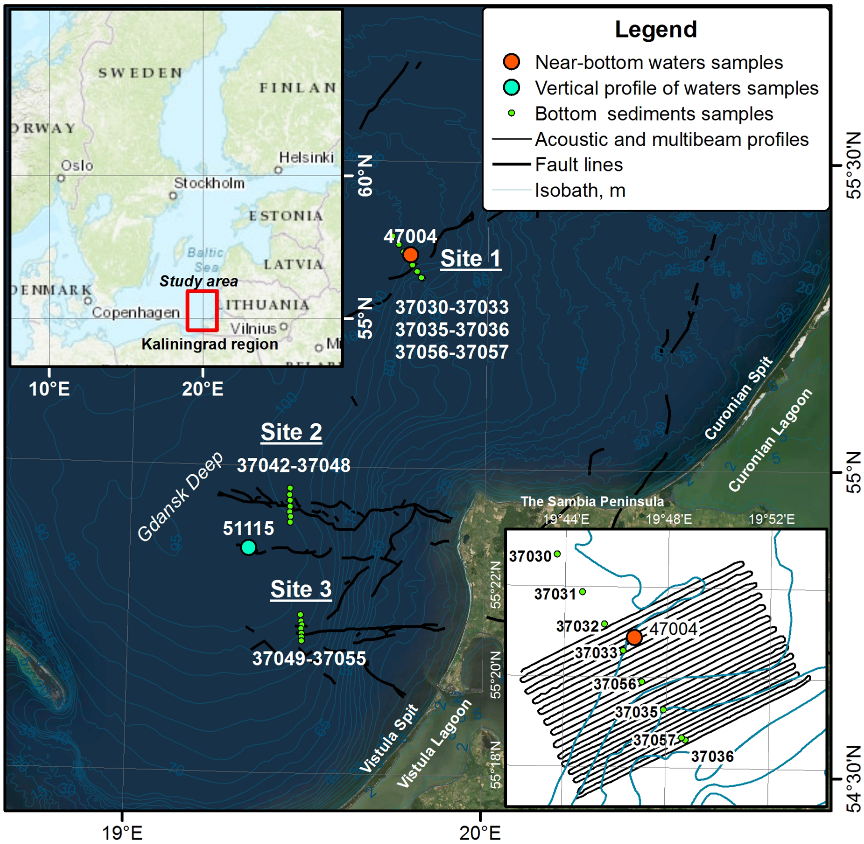

Figure 3. Sampling and geoacoustic survey scheme. Isobaths each 10 m.

Detailed survey of the bottom topography. The bottom topography survey was performed by the SeaBat 8111 multibeam echo sounder (frequency 100 kHz, beam width 1.5°). Data recording and processing was performed in the PDS 2000 software package, where all necessary offsets were included.

The calibration was carried out in the PDS 2000 software in the automatic mode. The resulting corrections were made during the survey. The sound velocity profile in water was applied into the PDS 2000 program before the survey. Profiling was performed with Idronaut Ocean Seven 316 Plus multichannel hydrophysical sounder.

The GRID terrain model and morphological analysis were implemented in the ArcGIS software.

Geoacoustic survey of the upper sediment layer. The vertical structure of the upper sediment layer was studied in conjunction with the bathymetric survey via the high-resolution EdgeTech 3300-HM profiler. The operating frequencies of the profiler were 2–6 kHz, the pulse length was 40 ms, and the sending frequency was 0.12–0.30 Hz.

CTD sounding. The CTD sounding was performed on 25–27.06.2020 with the Cast-Away probe from surface to bottom (the probe did not reach the bottom ∼ 0.3 m).

Sampling. Bottom sediment sampling was performed by the Box Corer during the 37th cruise of the *R/V Akademik Nikolaj Strakhov* [*Krek et al.,* 2019] at three key sites corresponding to tectonic fault zones (see Figure 3). Near-bottom water samplings and CTD soundings were performed during the 47th cruise of the *R/V Akademik Nikolaj Strakhov* [*Dorokhov et al.,* 2021] and the 51th cruise of the *R/V Akademik Sergey Vavilov.* Water samples were taken using a hermetic gravity corer (undisturbed near-bottom water) and Niskin

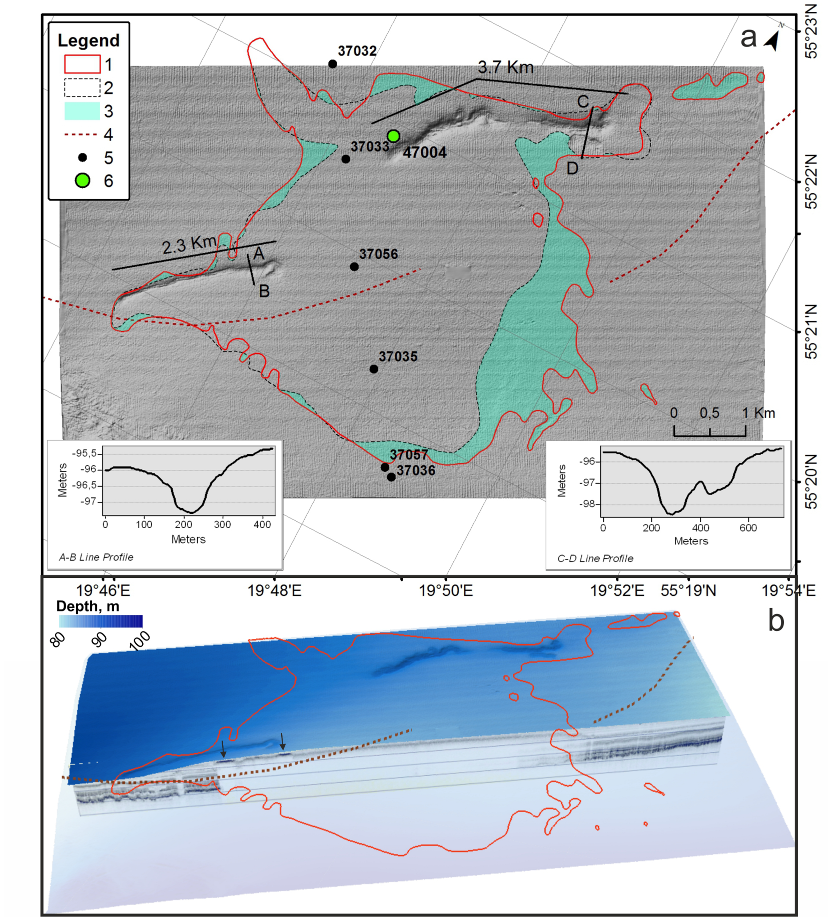

Figure 4. Bottom surface topography in the area of the gas-saturated sediments (a) and geoacoustic profile of the gas-saturated sediments combined with the bathymetry (b). 1 – gas-saturated sediment boundary based on the profiler survey (2017), 2 – gas-saturated sediment boundary based on [*Blazhchishin*, 1998; *Pimenov et al.*, 2010; *Ulyanova et al.*, 2012], 3 – previously unknown area of the gas-saturated sediments, 4 – tectonic faults according to Otmas et al., 2006, 5 – bottom sediments samples, 6 – near-bottom water sample.

water bottles 2–5 m above the bottom). Station 47004 was located in the gas-saturated sediments, in close proximity to the fault where the Jurassic (Oxford-Tithonian aquifer) groundwater discharge was assumed (see Figure 4), background station 51115 was located outside of the gas-saturated sediments area.

3.1. Laboratory Analyses

Determination of Ca in seawater. The determination of the calcium ions concentrations was made via a water sample (100 ml) neutralization with a 0.1 mol/dm^3 HCl solution. The solution was colored in pink and then another 1 cm^3 of HCl was added to the solution and boiled for 5 min to remove CO_2. The solution had to cool down and then 2 cm^3 of NaOH with a concentration of 2 mol/dm^3 was added and the pH was adjusted from 12 to 13. The 1 cm^3 of $C_{21}H_{14}N_2O_7S$ solution with the 0.025% mass fraction of $C_{21}H_{14}N_2O_7S$ was added as indicator. Subsequently, the obtained solution was titrated using $C_{10}H_{16}N_2O_8$ (EDTA) with a concentration of 0.05 mol/dm^3. Method accuracy is 0.02 mg, with a confidence level of 0.95. The lower limit of sensitivity of the method is 1 mg.

Determination of K, Na, Ca, Mg in bottom sediments. K, Na, Ca, and Mg were selected as marker elements for groundwater discharge in sediments [*Krek et al.*, 2020]. The con-

Table 2. Calibration Definition Values for K, Na, Ca, Mg

	Concentration, %			
	K	Na	Ca	Mg
Values obtained from standard samples	1.50	1.18	1.39	1.00
Certified values	1.83 ± 0.36	1.45 ± 0.28	1.32 ± 0.23	1.20 ± 0.21

centrations of the elements in the sediment samples were determined by atomic absorption analysis. Sample decomposition was carried out by the method described in [*Khandros and Shaidurov,* 1980]. All elements were determined on a Varian AA240FS flame atomic absorption spectrometer.

A sample (0.25 g) in a platinum crucible was placed in a muffle furnace and heated to 500°C. Then 5 ml of HF and 1 ml of $HClO_4$ were added to the ash residue and then the ixture was completely dried. 5 ml of HCl were added to the dried residue and heated until the salts were completely dissolved. The obtained solution was transferred into a 50 cm^3 volumetric flask, brought to volume with deionized water and stirred. Then the content of elements in the obtained solutions was determined on the atomic absorption spectrometer by measuring their atomic absorption (atomic vapour optical density) using the calibrations for solutions with known concentrations of elements to be determined. To control the correctness of analysis parallel samples of tested samples were used as well as a standard soil sample of known content of elements being determined.

All results of the instrumental measurements were checked against the reference values to determine possible errors. The limits of determination and error range are given in Table 2.

4. Results

Bottom topography and bottom sediments of upper layer structure. Based on the results of the multibeam bathymetric survey in the Gdansk Deep silt zone, the boundaries of pockmarks, that are located within the geoacoustic anomaly associated with gas-saturated sediments, were specified. The pockmarks were elongated along the slope up to 2.3–3.7 km in length and up to 3 m in relative depth (Figure 4). The boundaries of the geoacoustic anomaly were extended eastward and has an area of 23 km^2 (4 km^2 larger than previous square) [*Blazhchishin,* 1998; *Pimenov et al.,* 2010; *Ulyanova et al.,* 2012].

CTD sounding and Ca content in seawater. Calcium is the most illustrative element for comparing the ionic composition of seawater and waters of the Oxford-Tithonian horizon, since it is significantly predominant in groundwater [*Mikhnevich et al.,* 2019].

The water column of the Gdansk Deep in summer had ordinary stratification: the upper mixed layer (UML) separated from the cold intermediate layer (CIL) by a seasonal thermocline and the bottom layer separated from the CIL by a permanent halocline coinciding with the pycnocline (Figure 5). The permanent halocline was located at depths of 65 m.

The vertical distribution of Ca in the water column at the background station (51115) shows maximum values in the near-bottom layer, further along the vertical profile, the Ca concentrations were approximately at the same level UML, where the significant decrease occurred (Figure 5). The maximum concentration of Ca ions (155 mg/l) was recorded in the near-bottom layer at station 47004.

K, Na, Ca, Mg in bottom sediments. The studies of the fault zones showed that the maximum values of groundwater discharge marker elements (K, Na, Ca, Mg) were observed in the site 1. The higher average values in the site 1 were recorded for Na and Ca (Table 3) while in the site 2 the higher average values were obtained for K and Mg.

5. Discussion

Bottom topography and bottom sediments upper layer structure. The elongated shape of the pockmarks implies they are aging and inactive [*Ulyanova et al.,* 2012] due to the processes

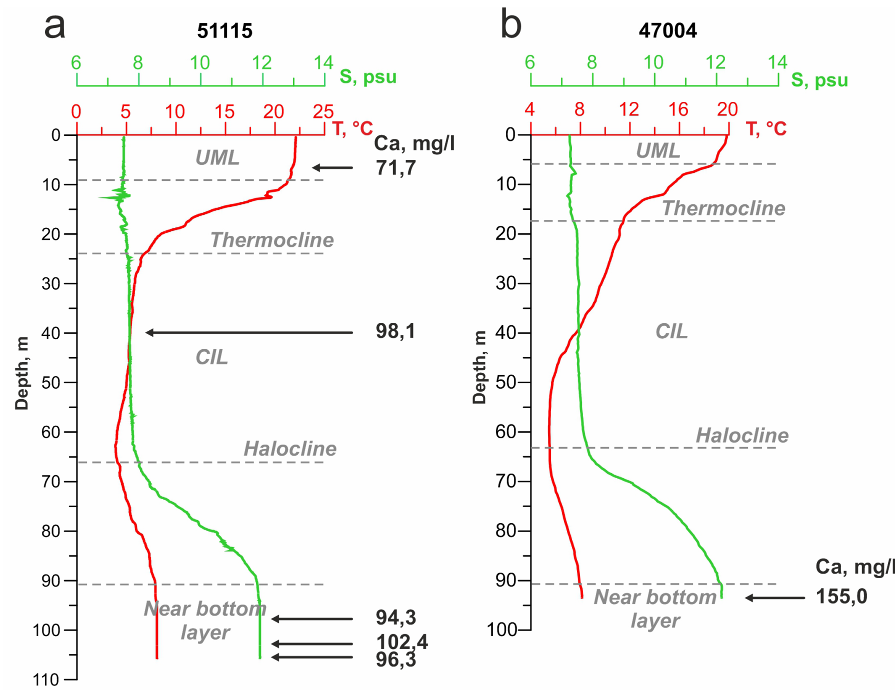

Figure 5. Vertical distribution of temperature (T) and salinity (S) at stations 51115 – background (a) and 47004 – gas-saturated sediments (b). Black numbers indicate the Ca concentration in the water.

Table 3. Average Concentrations of K, Na, Ca, Mg in the Bottom Sediments Upper (0–5 cm) Layer

Site		Concentration, mg/kg			
		K	Na	Ca	Mg
1	Min	1.54	1.69	0.84	1.00
	Max	2.50	2.73	3.02	1.43
	Mean	2.26	2.24	1.38	1.25
	Sigma	0.35	0.35	0.70	0.15
2	Min	2.26	1.88	1.00	1.17
	Max	2.44	2.21	1.31	1.50
	Mean	2.37	2.08	1.19	1.36
	Sigma	0.07	0.12	0.13	0.11
3	Min	2.03	1.70	0.92	1.00
	Max	2.43	2.33	1.21	1.40
	Mean	2.27	2.03	1.09	1.23
	Sigma	0.15	0.23	0.10	0.13

associated with a change in recharge channel position [*Cathles et al.,* 2010; *Hovland,* 2002]. The area where the pockmarks are distributed is the one with the silt thickness of more than 10–15 m [*Dearman,* 1991]. The pockmarks and geoacoustic anomaly in the northern site are located over a fault system according to *Otmas et al.* [2006]. According to *Blazhchishin* [1998], the faults in the zone of acoustic anomalies within the north-eastern part of the Gdansk Deep are connected with an Upper Cretaceous structural scarp. The roots of the anomalies extend into the Triassic-Jurassic deposits, in which supply channels are recorded, often ending as “gas pillows” in Holocene sediments.

The groundwater intrusion through fractured channels along with gas fluid has been detected in the northern section [*Krek et al.,* 2020]. In addition to gas seeps, groundwater discharges may be observed in the pockmarks too [*Bussmann and Suess,* 1998; *Idczak et al.,* 2020; *Schlüter et al.,* 2004; *Whiticar,* 2002]. It can be assumed, that the pockmarks in the study area may be considered as indicators of such “double” discharge. According to the geological map of Pre-Quaternary sediments [*Petrov,* 2010], the Oxford-Tithonian aquifer can be expected to be discharged in the northern site (Figure 6). The ionic composition of groundwater is dominated by Ca and Na in comparison with seawater [*Grigyalis and Kondratas,* 1983]. Mg is at approximately the same level as in seawater (Table 4), from which an additional source can be inferred [*Krek et al.,* 2020].

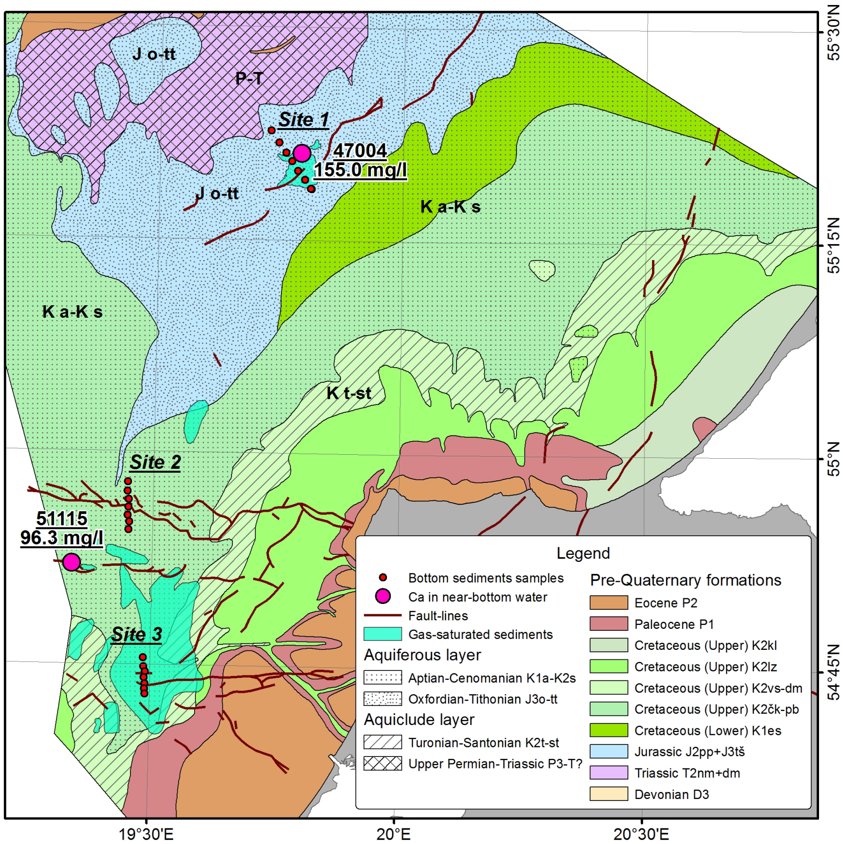

Figure 6. Scheme of a potential aquifer discharge layers (location of Pre-Quaternary formation is given after [*Petrov,* 2010]).

Hydrophysical conditions and Ca content of seawater. The presence of the permanent halocline makes vertical water exchange between the upper and the bottom layers of the Gdansk Deep difficult. Therefore, the processes occurring beneath the permanent halocline are mainly influenced by inflows of transformed North Sea waters and advective water exchange between the deep basins of the Baltic Sea [*Bulczak et al.,* 2016; *Elken and Matthäus,* 2008; *Mohrholz et al.,* 2015].

The concentration of particular chemical elements in the upper layers of the sea tends to depend considerably on season. Thus, the minimum Ca concentration in the UML (see Figure 5) seems to be related to the spring-summer development of planktonic communities, which occurs mainly in the surface layer. Ca consumption in the CIL is minimal and it's concentration is a consequence of accumulation during winter due to dissolution of biogenic carbonates.

The bottom layer is not dominated by seasons and near-bottom water exchange between sub-basins of the Baltic Sea comes first. The last significant renewals of near-bottom water in the Gdansk Deep were observed in 2014 and 2016 and their effects became undistinguished in 2018 [*Krek et al.,* 2019]. The inflows of lower intensity have been observed much more frequently, but their main path of propagation is through the Gdansk-Gotland Sill, bypassing the Gdansk Deep [*Krek et al.,* 2021]. Therefore, stagnant conditions have developed since 2018.

The normal Ca concentration in near-bottom water of the Gdansk Deep varies between 80 and 130 mg/l [*VNIIMORGEO,* 1978], and between 120 and 140 mg/l in pore water [*Carman and Rahm,* 1997]. The 60% increase in Ca concentration in near-bottom water at site 1 in comparison with the background station is evidence of Ca supply. Vertical sedimentation does not explain these differences because all sampling points are located beneath the halocline and input of additional Ca with suspended particulate matter and detritus along with its dissolution in the pockmark area seems unlikely.

Table 4. Concentration of Major Ions in Seawater and Groundwater

Ions	Seawater ion concentrations, mg/l [*Mikhnevich et al.,* 2019]	Groundwater ion concentrations in Upper Jurassic sediments, mg/l [*Grigyalis and Kondratas,* 1983]
Na^+	2261.25	4492.9
K^+	81.75	
Ca^{2+}	108.00	569.2
Mg^{2+}	284.25	266.1

No sources of additional Ca at the sediments surface are detectable because the sampling points are located within the benthic "desert", where shells are absent. Earlier, in 2001, the benthic "desert" area in the Gdansk Deep was located at depths greater than 80 m. The maximum depth of the bivalve *Limecola balthica* (Linnaeus, 1758) on the Gdansk Deep slope was 81 m [*Gusev and Jurgen-Markina,* 2012]. Lately bivalves on the slope have been found only up to 70 m depth [*Aleksandrov et al.,* 2021]. Horizontal transport and redeposition of shells by contour currents and their further dissolution require a source of such material on the slope at identical depths. No such source is known in the south-eastern part of the Baltic Sea. The main hypothesis of Ca increase in seawater is its endogenous input with the Jurassic waters enriched with it. The similar overall salinity and subsurface mixing zone [*Krek et al.,* 2020] does not allow such waters to be identified in the near-bottom layer by CTD.

K, Na, Ca, Mg in bottom sediments. Ca is most commonly present in bottom sediments as a carbonate of terrigenous or biogenic origin. The calcium carbonate concentration in bottom sediments in the southern and south-eastern parts of the Baltic Sea is generally less than 1% [*Gudelis and Emelyanov,* 1976; *Uścinowicz et al.,* 2011]. Ca concentration in bottom sediments (both average and maximum values) was significantly higher at site 1 (see Table 3). Moreover, the maximum Ca values at the study sites were noted precisely in the field of gas-saturated sediments (Figure 7). Added with fluid coming from deep aquifers through fractures Ca, when passing through the upper sediment layers (50 cm) is bound in formation of calcium carbonate, which is deposited into sediments, as well as Na [*Emelyanov,* 1998]. Sulphate reduction in pockmarks is more intensive than in gas-saturated or ordinary silt, which is related to the presence of methane [*Pimenov et al.,* 2010]. The increased Ca concentrations at site 1 in pockmark area are likely to appear due to this mechanism.

It is well known, that the distribution of Na in sediment cores is related to chlorination and total pore water salinity, but it can be assumed that conditions are similar at all studied sites, so the increased Na concentration at site 1 probably indicates the additional Na input. No source of additional Na other than the groundwater input is known (see Table 4). The high Na concentrations in the pockmark area has been noted earlier [*Emelyanov,* 2002]

The elevated Ca and Na concentrations, as well as the absence of anomalous Mg values, provide further evidence of groundwater deposition. The mean trace elements concentrations for the upper bottom sediment layer for cores 35084, 35097, 37056 and 37057 are shown in Table 5.

Table 5. Background Concentrations of Trace Elements (mg/l) in the Upper (0–5 cm) Bottom Sediment Layer [*Krek et al.,* 2020]

K	Na	Ca	Mg
1.86	1.65	1.24	0.97

When comparing the average values of trace elements in the upper layer of bottom sediments, the higher values of K, Na and Mg are noted at all study sites, while the high Ca concentration is observed only at site 1. This appears to be related to the chemical composition of discharge water, where

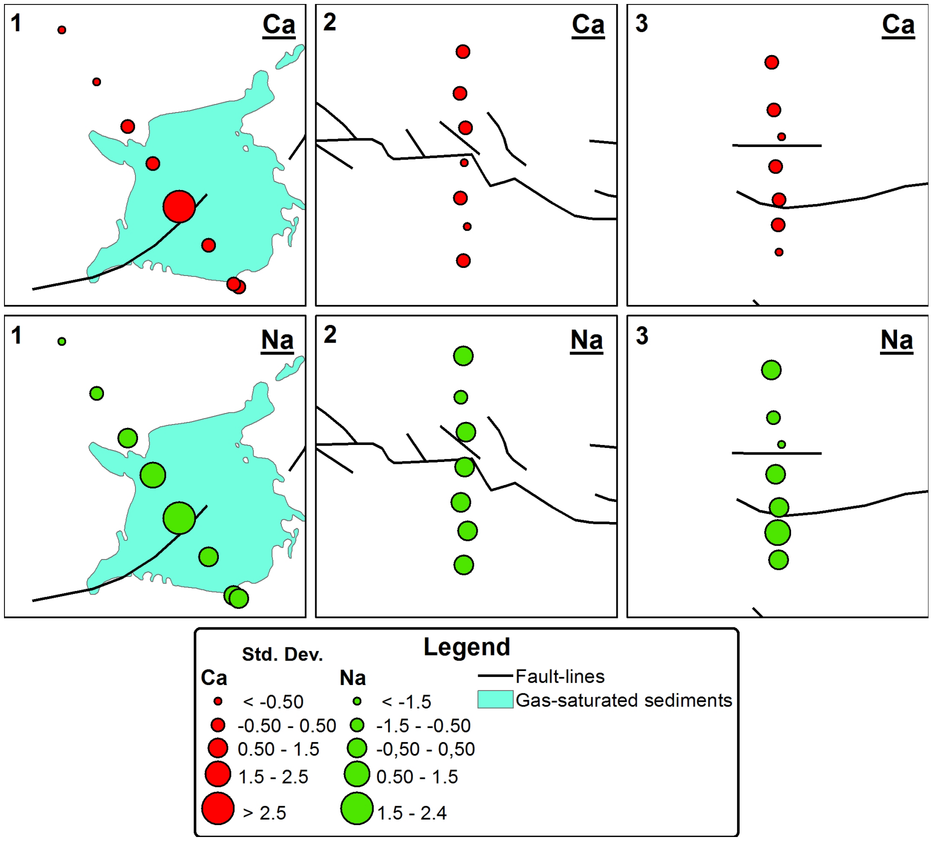

Figure 7. Ca and Na distribution in the bottom sediment study areas (number of sites are written in the left corners).

a significant increase in Ca concentration is noted in the Oxford-Tithonian aquifer. The potential discharge of the Aptian-Cenomanian aquifer, on the contrary, has the low Ca concentrations [*Grigyalis and Kondratas,* 1983].

Thus, geochemical evidence of endogenous elemental input was noted at all study sites. Differences in K, Na, Ca and Mg content between the study sites allowed us to suggest that discharge occurred from different horizons. The main marker of the Oxford-Tithonian horizon discharge is the maximum Ca concentration anomaly.

6. Conclusions

The aquifers discharge occurs along the tectonic faults that conduct fluids into the Holocene sediments. The groundwater mixes with seawater in the sediment strata, the exchange of macroelements occurs here too. This process may be detected by the trace elements anomalies in the composition of surface sediments. Zones of groundwater discharge often coincide with zones of gas-saturated sediments, which seems to be related to the fact that gas and water fluids uplift through the same pathways. Thus, pockmarks are not only the markers of sediment gas release to the water, but also the markers of groundwater discharge. The anomaly of Ca concentration in near-bottom water layer and in bottom sediments at site with the methane gassy sediments testifies the discharge of the Oxford-Tithonian aquifer.

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