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The Relationship between Total Mercury, Its Fractions and Species Diversity of Diatom Taphocoenoses Deposited in Surface Sediments (Southern Baltic Sea)

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Abstract: Mercury is a toxic metal that in excessive amounts negatively affects the human nervous system. It can biomagnify to enrich successive levels of the trophic web with higher and higher concentrations of Hg. Diatoms are cosmopolitan microalgae that represent the base of the trophic web. The objective of this study was to determine the importance of diatoms in the accumulation of Hg in surface sediments in the example of Puck Lagoon (southern Baltic Sea). The study verified the relationship of mercury forms with diatoms' characteristic salinity groups and individual diatom species. The identified diatom flora permitted the distinguishment of three regions with different salinity preferences. The obtained results suggest that, together with organic matter and fine sediment fraction, diatoms are an important element of the marine environment in accumulating Hg. Area I, dominated by euhalobous and mesohalobous diatom species, was characterized by a high statistically significant correlation of mesohalobous with $HgSO_4$, Hg in complexes with organic matter and HgS. In Area II, associated with the highest Hg concentrations (ca. 100 ng g^{-1}), euhalobous showed a statistically significant linear correlation with $HgSO_4$. In contrast, in Area III, the freshwater diatom community was associated with stable Hg forms. The identified species showed a connection with labile forms of Hg that can undergo biomagnification.

Keywords: Hg_{tot}; diatoms; microalgae; Puck Bay

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1. Introduction

Mercury is one of the most toxic metals negatively affecting the human nervous system. It may even contribute to Parkinson's disease and Alzheimer's disease [1]. Human activity has been recently showing the greatest impact on the increase in the concentration of mercury in the environment. In Europe and Asia, anthropogenic sources of Hg are dominated by fossil fuel [2]. Hg, along with sewage, rivers and terrestrial runoff, enters the coastal zone of the seas. There, it is incorporated into the trophic web where it undergoes biomagnification in subsequent levels, reaching concentrations up to a million times higher in marine mammals compared to the surrounding water [3–5]. The main source of Hg into the human body is the consumption of fish and seafood. An important vector incorporating Hg into the trophic web is phytoplankton. Due to their high abundance and frequency of occurrence, diatoms play a major role. Mercury is easily accumulated by diatoms, which create the entry point for the food web [6–8]. Diatoms are one of the most numerous groups of microorganisms dominant in terms of biomass abundance [9]. It is estimated that diatoms produce about 25% of the total carbon bound on Earth [10]. Data presented by Legrand et al. [11] and Staniszewska et al. [12] show that in the Baltic Sea, planktonic diatoms can account for up to >90% of the biomass during the bloom period (spring, autumn). Previous studies show that diatoms accumulate metals intensively, including Hg [13,14]. Therefore, diatom flora may be an important determinant of the mercury concentration in estuarine sediments and at the same time, a major vector allowing mercury into the food web.

Research on mercury (particularly forms of mercury) accumulated by diatoms in the sediments is relatively new. Previous studies have observed the relationship between changes in total mercury (Hg_{tot}) concentration in sediments and the number of diatom valves [15]. In addition, the possible role of diatoms in Hg concentration in sediments has so far been postulated based on biogenic Si content [16]. Research conducted in Antarctica showed that mercury can be stored in diatom flora, becoming the main sink for Hg deposited in sediments [16]. Moreover, previous studies on individual diatom species have confirmed the persistence of diatoms under high concentrations of heavy metals (including mercury) under laboratory conditions and their ability to convert mercury to forms harmless to diatoms [17–20]. Hg accumulation by diatoms can be controlled by other environmental factors [13,21]. Therefore, it is important to investigate the accumulation of Hg by diatoms in diverse marine environments.

A valuable test site for identifying environmental processes is Puck Lagoon, which is a basin with different hydrological conditions [22]. It is an area exposed to high concentrations of organic substances (e.g., pesticides) and metals, including mercury [23]. Characteristic of Puck Bay is the occurrence of the beach wracks, which provide storage for, among other things, Hg [24]. This creates a secondary source of Hg to the coastal water. Since 1990, a gradual decrease in the emission and deposition of Hg has been observed [25]. Consequently, a decrease in mercury concentrations in Baltic Sea surface sediments has been recorded over the years [26]. Nevertheless, the Puck Lagoon area is still exposed to Hg supply through river inflow, atmospheric deposition and cliff erosion [27,28]. Considering the fact that diatoms constitute one of the most abundant groups of microorganisms in Puck Lagoon and the fact that microorganisms efficiently accumulate Hg, it is possible that they may accumulate a considerable mass of mercury, potentially undergoing biomagnification. Conducted research indicates that Hg concentrations of epilithic diatoms reached lower values compared to the epiphytic and planktic assemblage but reached much higher values than mercury contained in sediments and macroalgae [6].

The concentration of mercury (Hg) as well as other metals in sediments is related to the presence of organic matter (OM) and the fine sediment fraction (FSF) [29]. The shallow water part of Puck Bay on the Polish coast of the Baltic Sea was characterized by benthic diatom biomass ranging from 50% (spring) to 59% (autumn) [30]. Therefore, the objective of this study was to determine the role of diatoms next to OM and FSF in Hg accumulation in Baltic sediments at the base of a lagoon in the southern Baltic Sea (Puck Lagoon), as is the case in the Arctic region. The scientific novelty will be the estimation of the impact of discrete salinity groups (euhalobous, mesohalobous, oligohalobous halophilous and oligohalobous indifferent) and individual diatom species on the accumulation of Hg and its forms in the sediments. The analyses were extended to include studies on five fractions of different forms of mercury. The studies were carried out on the sediments of Puck Lagoon, however, the processes discussed could take place in other analogous embayments of the world. Due to the risk of contamination, mercury was analysed from bulk sediments, and not in the diatom. Diatoms are microalgae ranging in the size of 1.5–5000 µm [31]. In contrast, in the present study, diatoms ranged in size from 4 to 150 µm. Therefore, it is extremely difficult to isolate them from among FSF and OM under the conditions and quantities required for trace analyses. Statistical analyses were used to verify the hypothesis of the study.

2. Materials and Methods

2.1. Study Area

Puck Lagoon is the north-western part of Puck Bay, belonging to the Gulf of Gdańsk. It covers an area of 104.8 km^2 (approximately 40% of Puck Bay) [32]. It is confined to the Hel Peninsula (from Kuźnica to Władysławowo) and by land from Władysławowo to Rewa (Figure 1). Puck Lagoon is a shallow water body with an average depth of 3 m. Greater depths are found in the Kuźnica Deep (max. depth 9.7 m), the Rzucewo Deep (max. depth 5.7 m) and the Chałupy Deep (max. depth 4 m) [32]. The hydrological conditions of Puck

Lagoon are influenced by rivers (Reda, Płutnica and Gizdepka) flowing into it [33]. The most important one is the Reda River, introducing approximately 0.187 km^3 of water into Puck Lagoon per year [34] and accounting for approximately 66% of river inflow to the lagoon [35]. Furthermore, the Seagull Sandbar between Puck Lagoon and the outer Puck Bay limits the water exchange occurring in the northern part through the Kuźnica Passage and in the southern part through Głębinka Passage [36].

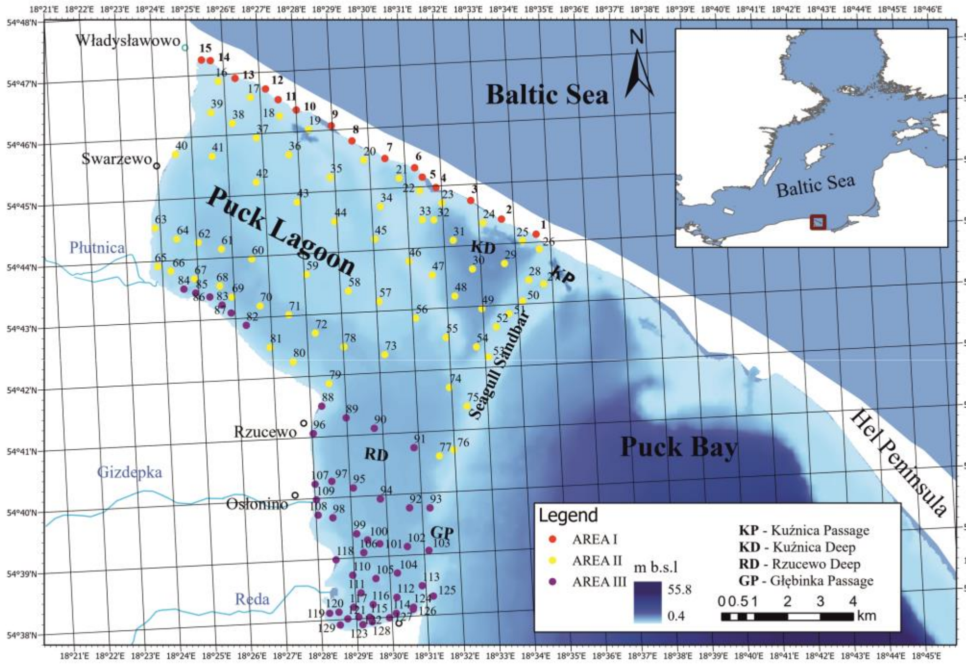

Figure 1. Location of the analysed samples.

Hydrodynamic conditions in the inner Puck Bay (Puck Lagoon) are shaped by water inflow from the outer Puck Bay and wave action [37]. At the bottom of Puck Lagoon, weak bottom currents (10–20 cm s^{-1}) are observed [38]. The main factors generating currents in Puck Bay are the wind and sea level changes associated with the wind [39]. The direction of currents in the inner Puck Bay is parallel to the shoreline, shoals and basins present in the area [39]. Two clockwise circulation systems can be distinguished in Puck Lagoon. The first one in the area of Puck Basin (from Głębinka Passage through Puck to Swarzewo City), while the second one in the area of Kuźnica Basin [39].

The water temperature in Puck Lagoon from June to October in the shallow water zone is 17.1–17.5 °C and drops to 15.1–15.5 °C in the area of Jama Kuźnicka and near Głębinka [32]. However, from December to April the temperature ranges from ca. 3.12 °C in the area of the coastal zone of Władysławowo and Hel Peninsula to 2.88 °C in the area of Głębinka and Kuźnickie Passage [40]. The salinity in Puck Lagoon (annual mean 7.31) is influenced by limited water exchange with the outer Puck Bay (annual mean 7.65) and the inflow of fresh river water [32].

The distribution of sediments in Puck Bay is related to the depth and changes in bottom relief [41]. For further detail on the sediment grain size in the inner Puck Bay, see the Supplementary Material Figure S1. In the whole area of Puck Lagoon, sediment mixing occurs. According to the study by Graca et al. [42], intensive mixing in Puck Bay is likely to occur down to a depth of approximately 80 m. The majority of the bottom of Puck Lagoon is covered by fine-grained sands [41]. However, the coastal zone of Puck Lagoon is associated with the occurrence of coarse-grained sands admixed with gravels. Medium-grained sands are observed up to the 1 m isobath [41]. Deeper areas (Rzucewo Deep, Chałupy Deep and Kuźnica Deep) and near the mouth of the Reda River are covered by silty sand [41]. In addition, sandy silt is present in the deepest part of the Kuźnica

Deep [41]. Sources of the material deposited in Puck Bay include fluvial inflow, storm overflows or eolian processes [41].

The rate of deposition in Puck Lagoon is variable. In Kuznica and Rzucewo Deep, changes in the rate of deposition are observed seasonally (autumn–winter about 2.5 mm year^{-1}; spring–autumn about 5 mm year^{-1}) [43]. In addition, there are spatial variations in the rate of accumulation. Near the mouth of the Reda River, the accumulation rate can be as high as 4.5 mm year^{-1} [44]. A similar accumulation bluntness is observed in Władysławowo pit. In Głębinka Passage, on the other hand, it is approximately 10 mm year^{-1} [43].

Puck Lagoon is a relatively shallow body of water (mean 3 m of depth) with a strongly restricted exchange with the open waters from the Gulf of Gdansk. This makes the dynamics of the processes like resuspension, remobilisation and transport limited. Allochthonous as well as autochthonous diatom species can occur throughout the study area. They may originate from rivers or the outer part of the Puck Bay, and epiphyton, in addition to plankton, may be found in a shallower area and benthos in a deeper area. The water exchange between Puck Lagoon and the outer Puck Bay is poor. Furthermore, material was not collected from the estuary itself. Therefore, the authors felt that they could exclude the influence of these factors on the diatom flora present and its origin.

2.2. Sample Collection

Samples for analysis were collected in August and September 2019 using a pontoon on Puck Lagoon (inner Puck Bay), the shallowest part of the Gulf of Gdańsk, southern Baltic Sea. During sediment collection, good atmospheric conditions prevailed. The salinity of the water was 7.1–7.2 and the water temperature ranged from 14 °C to 21.3 °C. Further details on the atmospheric conditions prevailing at the time of sampling and sampling depth are presented in Supplementary Material Table S1.

In the AREA I and AREA III shoreline area, sampling was dependent on access to the sea. Therefore, the distribution of points in this area is irregular. However, areas influenced by human activity, undisturbed areas and areas close to river mouths were selected. The distance of the measurement points in the shoreline zone is 0.3–4 km. In deeper areas of AREA III, which were easily accessible for sampling by pontoon, the distance between locations ranged from 0.5–2 km. Such a dense grid of measurements in AREA III was planned due to a large number of potential factors influencing this area (river water inflow, human activities and the influence of water inflow from the outer Puck Bay through Głębinka Passage). In AREA II, a less frequent measurement grid was planned, as this area, in contrast to AREA III, is influenced by a smaller number of factors (mainly, the influence of the outer Puck Bay through the Kuźnica Passage). In AREA II, the distance between points was 1–3 km. A total of 129 sandy and muddy surface sediment samples were collected from the bottom of the entire lagoon (depth $\leq$ 9 m) by means of a Van Veen Grab sampler, with a surface area of 250 cm^2 and a depth no more than 10 cm (Figure 1).

2.3. Diatom Analysis

The diatom analyses were prepared according to the standard Battarbee [45] procedure. To remove calcium carbonate and organic matter, all samples for diatom analysis (approximately 1 g of dry sediment) were treated with 10% HCl and 30% H_2O_2, respectively. Quantitative and qualitative diatom analyses were carried out on all samples. Permanent diatom slides were made using Naprax, UK (refractive index nD—1.73). The concentration of diatom valves per unit weight of dry sediment was estimated according to the Bodén [46] method. The analysis was performed under a light microscope Nikon Eclipse Ci, Japan, under a 100× oil immersion objective. The counting method of Schrader and Gersonde [47] was used, and in each sample, approximately 300–500 valves were counted in order to estimate the percentage abundance of particular taxa. Taxonomic identification was carried out based on Hustedt [48], Krammer and Lange-Bertalot [49–52], Pankow [53], Lange-Bertalot [54], Bąk et al. [31], Snoeijs [55], Snoeijs and Balashova [56], Snoeijs and Kasperovičiene [57], Snoeijs and Potapova [58], Snoeijs and Vilbaste [59], and Witkowski et al. [60]. The ecological preferences, including habitat [9], salinity [61], pH [48] and trophic [62] and saprobic status [63], were determined based on OMNIDIA 6.08 software. For further detail on the division of diatoms according to the analysed ecological preferences, see Supplementary Material Table S2.

2.4. Hg Analysis

The collected study material was preserved at −20 °C until mercury analysis. Before analysing mercury contained in the sediment, the samples were freeze-dried and homogenized using a ball mill. The analysis of total mercury (Hg_{tot}) and mercury groups was carried out by the thermodesorption method using a DMA-80 analyser by Milestone, Italy. The method presented earlier by Reis et al. [64] has been improved and can be used to analyse sediment with high OM and even biological matter content [65]. The analysis was performed on material representing 0.1–0.4 g of dry sediment. The research material was placed in nickel boats, previously roasted for 1 min at 800 °C. The resulting research material was subject to total mercury analysis and 5-step mercury analysis in accordance with the procedure presented by Bełdowska et al. [66]. Total mercury analysis involved measuring the concentration of the sample burned at 750 °C. Mercury fraction analysis involved measuring the concentration of mercury groups released when the material was burned at specific temperatures. The first analysed fraction of Hg related to halides (Hg_{F1}) and can be adsorbed on sediment and diatom shells and is released at 175 °C. The second analysed fraction absorbed inside the cells (embedded in diatom frustules) (Hg_{F2}) was Hg forming complexes with organic matter released at 225 °C. The last labile form adsorbed on diatom frustules and released at 475 °C was associated with mercuric oxide (HgO) and mercuric sulphate ($HgSO_4$) (Hg_{F4}). In contrast, the analysed stable forms included mercury sulphide (Hg_{F3}) released at 325 °C and mercury associated with the matrix (Hg_{F5}) released at 750 °C. The determination of Hgtot was verified by the analysis of certified reference materials: soil NCS DC 87103 (HgTOT: 17 ng g^{-1}) and marine sediment GBW 07314 (HgTOT 43 ng g^{-1}) at mean recovery > 95% and standard deviation (SD) > 85%. The limit of detection (LOD) was calculated based on the SD of the Hg concentration in the blank samples, measured in 10 repetitions (LOD = 3 × SD). In the case of analysis of HgTOT, LOD was 1 pg Hg. More details are described by Wilman et al. [67].

There are no reference materials for mercury speciation (except for MeHg). Therefore, the method was tested on certified reference materials (tea leaves—INCT-TL-1; soil—NCS DC 87103), as well as on 3 synthetic materials ($HgCl_2$, HgS and HgO) (Sigma Aldrich Co., St. Louis, MO, USA, purity ≥ 97%) and two natural materials (seal muscle and seal faeces). More details are disrobed by Wilman et al. [67]. The accuracy of the method was verified by comparing the sum of the Hg concentrations measured by the 5-step fractionation method with the result of the HgTOT analysis. The accuracy of the Hg forms analysis averaged 94%, with a standard deviation not exceeding 5%. The accuracy and precision of the method were verified by the analysis of certified reference materials (soil NCS DC 87103—HgTOT 17 ng g^{-1} and marine sediment GBW 07314—HgTOT 43 ng g^{-1}). The analysis of reference materials was carried out in three replicates, for which the recovery was found to be at levels of 96 and 98%, respectively [68].

2.5. Loss on Ignition, Wetness and Granulometry Analysis

The percentage of organic matter (OM) in the analysed sediment was measured by means of loss on ignition (LOI). For this purpose, the sediment was burned at 550 °C for 6 h. Moreover, sediment wetness was determined by the weight ratio of wet sediment to dried sediment at 105 °C [69]. The sediment was then subject to granulometric analysis by means of the sieve method [70]. The sediment was separated on a column of sieves with a mesh diameter ranging from −3 φ (8 mm) to 4 φ (0.063 mm). Sediment with grain diameter > 4 φ (<0.063 mm) was classified as the fine sediment fraction (FSF) based on the classification by Gradziński et al. [71].

2.6. Data Processing

The results of the concentrations of Hg_{tot} (ng g^{-1}) and its fractions (Hg_{F1}, Hg_{F2}, Hg_{F3}, Hg_{F4} and Hg_{F5}) were presented as concentrations in dry sediment. Statistical analyses were performed to determine the association of Hg to the identified diatom species. The percentage of each diatom group was used to characterize diatom ecological groups in relation to the Hg concentration (similar to the content of organic matter and fine sediment fractions). For the characterization of diatom species in relation to Hg concentration, the percentage of individual species was taken into account.

Statistical analysis (PCA, corelation analysis) and graphic representation of the results of mercury analysis were carried out using STATISTICA 13 (StatSoft, Kraków, Poland). The results of the diatom analysis were presented using TILIA 2.0.37 [72]. The diatom diagram shows salinity preference of the species that occurred with ≥5% frequency in at least one sample. Cluster analysis was performed by means of CONISS [72] and presented as percentage diagrams. The map of the study area and maps showing spatial distribution of Hg_{tot} concentrations, contents of diatom valves, %OM and %FSF were created using ArcGIS Pro 2.7.0 (ESRI, New York, NY, USA) with the WGS1984 geographic coordinate system.

3. Results and Discussion

3.1. Diatom Analysis

The analysed diatom flora preserved in the sandy and silty sediments of Puck Lagoon were abundant and well preserved. A total of 159 species belonging to 57 genera were identified (Figure S2). CONISS cluster analysis showed three prominent areas differing in salinity preferences of the dominant diatom species. For further detail on the characteristic of diatom taxa, see Supplementary Material Figure S2.

AREA I (PL1–15) along the Hel Peninsula, due to its shallow depth (<1 m), was characterised by the dominance of benthic species (approximately 100%) (Figure S3). For further detail on the percentage content of the diatom ecological groups, see Supplementary Material Figure S3. Euhalobous species were represented by *Halamphora coffeaeformis* (C. Agardh) Mereschkowsky and *Nanofrustulum krumbeinii* (Witkowski, Witak and Stachura) Morales (Figure 2). Mesohalobous species preferring waters less polluted with organic substances (β-mesosaprobionts) include *Gedaniella flavovirens* (Takano) Li, Witkowski and Ashworth, *G. mutabilis* (Grunow) Li and Witkowski, *G. guenter-grassii* (Witkowski and Lange-Bertalot) Chunlian Li, Sato and Witkowski, and *Planothidium delicatulum* (Kützing) Round and Bukhtiyarova. Furthermore, oligohalobous halophilous species, including *Nitzschia frustulum* (Kützing) Grunow and *Planothidium lemmermannii* (Hustedt) Morales, were observed abundantly.

All species except *P. lemmermannii* were observed in surface sediments along the Hel Peninsula by Witak et al. [73]. Like the other species, *P. lemmermannii* was commonly observed in different parts of the Gulf of Gdansk [74,75]. The concentration of diatom valves in sediments along the Hel Peninsula ranged from 9×10^5 valves g^{-1} (PL5, Pl8) to 71×10^5 valves g^{-1} (PL2) (Figure S3). The estimated concentration in the analysed material was lower in comparison to the results presented by Witak et al. [73], where the concentrations ranged from 42×10^6 valves g^{-1} to 60×10^6 valves g^{-1}.

AREA II (PL16–81), in the central part of Puck Lagoon, like AREA I, showed the dominance of benthic species (84–100%). Planktic taxa were also observed, however, they were particularly abundant in the area of the Kuźnica Deep (PL29–30), with a frequency of 5–12% (Figure S3). Planktic species were mostly represented by *Cyclotella chactawhatcheeana* Prasad and *Chaetoceros* spp. RS. The diatom flora of AREA II are dominated by oligohalobous indifferent taxa, including *Amphora pediculus* (Kützing) Grunow, accompanied by species of genera *Cocconeis* and *Staurosira* (Figure 2). Their occurrence is related to the inflow of the waters of the Płutnica River. Relatively high frequency was recorded for brackish species *Gedaniella mutabilis* and *Planothidium delicatulum*, constituting permanent components of the diatom community of the shallow zones of Puck Bay and the Gulf of Gdansk [74,76,77]. Among marine species, *Cocconeis scutellum* Ehrenberg, *Martyana schulzii*

(Brockmann) Snoeijs, *Opephora horstiana* Witkowski and *Gedaniella guenter-grassii* were observed more frequently in the northern part of AREA II. In the southern part of AREA II, however, *Catenula adhaerens* Mereschkowsky and *Navicula germanopolonica* Witkowski and Lange-Bertalot were observed in the community. The frequency of these taxa suggests that the hydrology of this area is shaped by higher salinity waters flowing from the outer Puck Bay through the Głębinka Passage to the south and through the Kuźnica Passage to the north. Furthermore, the oligohalobous halophilous *Planothidium lemmermannii* is also highly frequent in this area, potentially due to the inflow of the Płutnica River. The species has been observed in the shallow water zone of Puck Lagoon [76,78] and the Gulf of Gdańsk [74]. AREA II is characterised by valve concentration ranging from 5×10^5 valves g^{-1} (PL17) to 127×10^5 valves g^{-1} near the Kuźnica Deep (station PL30). A high concentration was also observed near the Swarzewo coastal area (PL63), where it increased to 81.67×10^5 valves g^{-1} (Figure S3). AREA II features the highest concentration of valves of the entire study area.

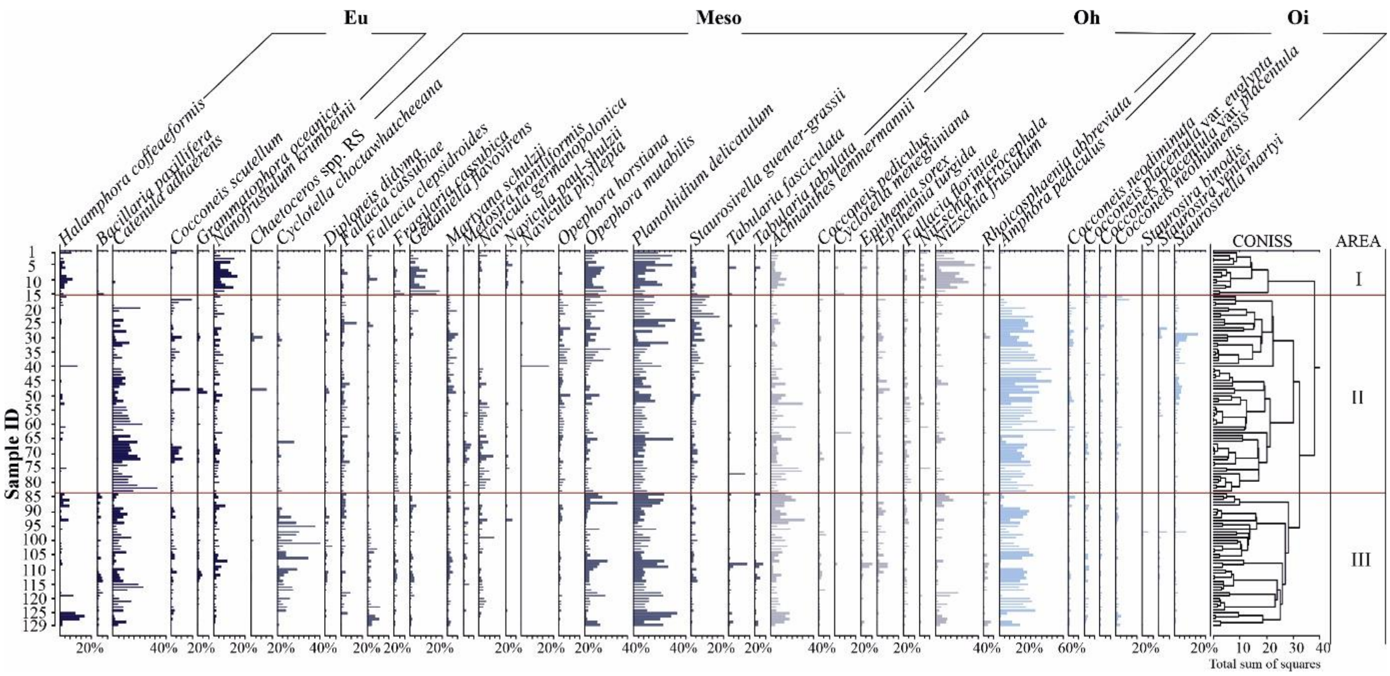

Figure 2. Frequency of the main diatom species: Eu—euhalobous, Meso—mesohalobous, Oh—oligohalobous halophilous, Oi—oligohalobous indifferent.

AREA III (PL82–129), covering the southern part of Puck Lagoon, showed an increased frequency of planktic species (≤37%), particularly in the vicinity of the Rzucewo Deep (Figure S3). Their primary representative was the anthropogenic species *Cyclotella choctawhatcheeana*, often observed in the sediments of the nearby Port of Gdynia [79]. AREA III was characterised by an increased frequency of mesohalobous taxa (19–64%) and a decreased percentage content of oligohalobous indifferent taxa (3–60%), compared to AREA II. *Gedaniella mutabilis* and *Planothidium delicatulum* represented the first group, while the second group was represented by *Amphora pediculus*. This distribution points to a greater role of brackish waters in shaping the hydrology of waters in the southern part of the lagoon. In the vicinity of the Reda River, a higher percentage of eutraphentic species tolerating high concentrations of nutrients in water was recorded, namely, *Halamphora coffeaeformis* and *Nitzschia frustulum* (Figure 2). They could be associated with the presence of recreational centres in this part of the lagoon's coastal zone. The species *Catenula adhaerens*, preferring less polluted waters, was observed more frequently at the other stations. In comparison to AREA I, lower concentrations of diatom valves in the sediment were observed in AREA III. In AREA III, their concentration ranged from 4×10^5 valves g^{-1} to 102×10^5 valves g^{-1}

in the Rzucewo Deep (PL115). Moreover, near the mouth of the Reda River (PL120), the concentration of diatom valves in the sediment increased to 53 $\times$ 10^5 valves g^{-1} (Figure S3).

3.2. Hg in Surface Sediments

The average concentration of total mercury (Hg_{tot}) in surface sediments of Puck Lagoon was low, reaching 4.6 ng g^{-1} (median 1.1 ng g^{-1}). The highest percentage of Hg_{tot} was represented by mercury in complexes with organic matter Hg_{F2} (37%), Hg_{F3} (36%) and mercury adsorbed in the form of $HgSO_4$ and HgO (Hg_{F4}) (21%). The lowest percentage of Hg_{tot} was matrix-bound mercury Hg_{F5} (5%) and mercury related with halides Hg_{F1} (1%) (Figure 3). According to the study by Kwasigroch et al. [68], such a distribution is characteristic of this region (Gdańsk Basin).

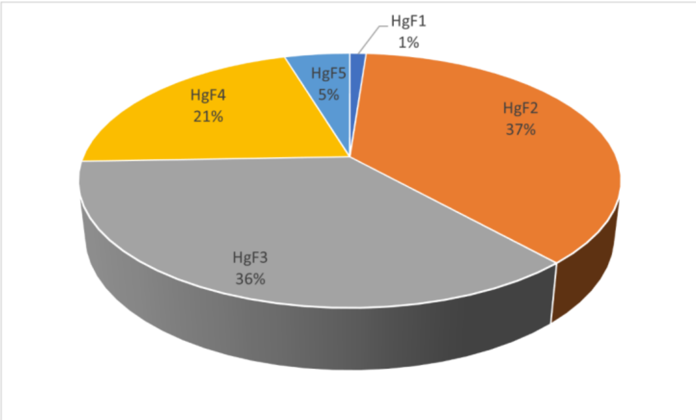

Figure 3. Average percentage of labile and stable forms of mercury in the surface sediment of Puck Lagoon.

AREA I (PL1–15) was the region with the lowest concentrations of Hg_{tot} ($\leq$2.3 ng g^{-1}). Also, some of the lowest values over the entire Puck Lagoon were measured for the analysed forms of mercury. In this area, the highest concentration in sediment was measured for Hg_{F2} (1.2 ng g^{-1}). Such low Hg concentrations in this area were associated with low contents of diatom valves ($\leq$71.2 $\times$ 10^5 valves g^{-1}), OM ($\leq$1.3%) and FSF ($\leq$1.7%) in the surface sediment (Figure S4). In Puck Lagoon, the average organic matter content ranges from 1% up to 5% [80–82]. The low content of Hg, OM, FSF and diatom valves may result from the lack of supply sources for those in close vicinity to the area. In addition, high water dynamics in this location make it difficult for diatom flora to attach to the substrate. Further details on spatial variability of the concentrations of Hg_{tot}, contents of diatom valves, %OM and %FSF, are presented in the Supplementary Material Figure S4.

The northern deepest part of the basin (stations PL29–31, AREA II) (Figure 1) was characterized by the highest concentrations of Hg_{tot}, reaching almost 100 ng g^{-1} (Table 1) and exceeding the value accepted as good environmental status (70 ng g^{-1}) as defined by the Directive 2008/56/EC of the European Parliament and of the Council. High Hg concentrations in this area resulted from its location; it is a deposition zone that contributes to the accumulation of pollutants, including mercury [83]. The increase in bottom depth is associated with an increase in sediment dispersion [41]. Moreover, fine sediment fraction and organic matter delivered by currents in Puck Lagoon are deposited in the Kuźnica Deep depression. Therefore, this area also had the highest diatom valve concentration (127 $\times$ 10^5 valves g^{-1}), content of organic matter (22.7%) and fine fraction (21.69%) in the analysed sediment (Figure S4). The increase in the content of silt minerals in sediments associated with the deepwater area is accompanied by an increase in the concentration of OM [80]. Moreover, the high share of OM in bottom sediments of deep water areas is associated with more intense phytoplankton blooms in these areas [80,84]. This is also indicated by the results of the conducted study. Moreover, vegetation overgrowing the bottom of the shoreline zone contributed to stable conditions, allowing for the retention of organic matter which consequently may have contributed to increased Hg concentrations. On the other hand, mercury in the deeper regions of Puck Lagoon (Kuźnica and Rzucewo Deep) could probably have been transported from the outer Puck Bay with the inflow of more brackish waters through the Kuźnica and Głębinka Passage [85] (Figure 1).

Table 1. Statistical characterization of the Hg_{tot} and its fractions (ng g^{-1}), concentrations of diatoms valves (valve × 10^5 g^{-1}), percentage of organic matter (OM) and fine fraction (FSF) in the surface sediments: * <LD—below the level of detection.

		Hg_{tot}	Hg_{F1}	Hg_{F2}	Hg_{F3}	Hg_{F4}	Hg_{F5}	Concentration of Diatom Valves	OM	FSF
All	Min.	0.1	0.0	0.0	<LD *	<LD *	0.0	5.0	0.1	0.0
	Max.	99.9	0.5	93.6	13.2	5.8	8.2	127.0	22.7	23.7
	Mean	4.6	0.0	2.8	1.0	0.5	0.1	31.0	1.9	2.9
	Median	1.1	0.0	0.3	0.3	0.2	0.0	24.0	1.0	0.7
AREA I	Min.	0.2	0.0	0.1	0.0	0.0	0.0	8.8	0.2	0.0
	Max.	2.3	0.0	1.2	1.3	0.6	0.1	71.2	1.3	1.7
	Mean	0.8	0.0	0.4	0.2	0.1	0.0	30.4	0.6	0.5
	Median	0.6	0.0	0.3	0.1	0.1	0.0	24.5	0.6	0.2
AREA II	Min.	0.1	0.0	0.0	<LD *	<LD *	0.0	5.4	0.1	0.0
	Max.	99.9	0.5	93.6	13.2	2.2	8.2	127.0	22.7	21.7
	Mean	6.6	0.0	4.8	1.0	0.4	0.2	31.7	2.4	2.1
	Median	1.0	0.0	0.2	0.3	0.1	0.0	24.7	1.1	0.9
AREA III	Min.	0.2	0.0	0.0	0.0	0.0	0.0	3.8	0.2	0.0
	Max.	14.7	0.1	2.4	8.3	5.8	0.4	101.7	5.2	23.7
	Mean	3.0	0.0	0.6	1.3	0.8	0.1	25.0	1.5	4.7
	Median	1.9	0.0	0.5	0.6	0.4	0.1	19.3	1.1	1.1

In the southern part of the lagoon, near the shore zone of Swarzewo (station PL40–AREA II), in the vincity of the Płutnica (station PL67–AREA II), Gizdepka (station PL97–AREA III) and Reda rivers (stations PL111, PL118–AREA III), and in the area of the Rrzucewo Deep (station PL101–AREA III), an increase in Hg_{tot} concentrations of up to approximately 20 ng g^{-1} was also observed. In these areas, the concentration of diatom valves (up to 127 × 10^5 valves g^{-1}) and the percentage of organic matter (up to 22.7%) and fine fraction (up to 23.7%) in the sediment increased simultaneously. River water was a substantial source of Hg in the study area. The Reda River is therefore of particular importance, with an average water flow of 5.9 m^3 s^{-1}, contributing metal from a catchment area of 485.2 km^2 [22,84]. Rivers introduce >12.5 tons of sedimentary material into Puck Lagoon, which is deposited near the river mouths [86]. This contributes to the increase in silty and clayey fractions in the surface sediment in this region [81,86]. Cliff erosion is also a prominent source of Hg in this part of Puck Lagoon [87]. The coastal zone of Puck Lagoon is fed by sediment derived from the cliffs along the reservoir [41]. This results in a lower content of fine sediment fraction ($\leq$1.72%) and organic matter (0.4–2.6%) in this area, which translates into lower Hg concentrations (0.3–10.85 ng g^{-1}) in the sediment of the AREA III coastal zone of Puck Lagoon. At the same time, this area is characterized also by low content of diatom valves (4.9–31.3 × 10^5 valves g^{-1}) (Figure S4).

Factors Influencing Hg Concentration in the Surface Sediment

The accumulation of metals (including Hg) in sediments is primarily related to the presence of organic matter (OM) and fine sediment fraction [29]. The statistical analyses in the study area confirmed a high statistically significant correlation of Hg_{tot} concentration with organic matter contained in the sediment ($r = 0.9$, $p < 0.05$) (Table 2) and with the fine sediment fraction ($r = 0.5$, $p < 0.05$). Studies of cores conducted in Arctic lakes [15,16] and the north-west Pacific [88] have revealed another factor influencing Hg_{tot} accumulation in sediments: diatom valves. The diatom flora is an important source of organic matter in Puck Lagoon. Diatoms are one of the most abundant and diverse microorganisms in this area [89]. Diatom valves preserved in the sediment showed a statistically significant relationship with the total mercury deposited in the sediment (r = 0.6, $p < 0.05$), which is also supported by the PCA analysis (Figure 4). This shows that in the study area, mercury is more easily accumulated in the diatom flora than in the fine fraction.

Table 2. Correlation coefficients determined for Hg_{tot}, labile and stable fractions of mercury with OM%, and the concentration of diatoms valves and FSF% in the surface sediments. Statistically significant correlations are marked in bold.

	Hg_{tot}	Hg_{F1}	Hg_{F2}	Hg_{F3}	Hg_{F4}	Hg_{F5}	OM%	FSF%	Concentration of Diatoms Valves	Wetness
Hg_{tot}	1.00	**0.73**	**0.99**	0.02	**0.21**	0.05	**0.88**	**0.54**	**0.59**	**0.54**
Hg_{F1}	**0.73**	1.00	**0.64**	**0.31**	**0.28**	**0.19**	**0.72**	**0.38**	**0.33**	**0.54**
Hg_{F2}	**0.99**	**0.64**	1.00	−0.10	0.11	0.00	**0.85**	**0.42**	**0.57**	**0.52**
Hg_{F3}	0.02	**0.31**	−0.10	1.00	**0.87**	−0.03	0.12	**0.42**	0.08	**0.30**
Hg_{F4}	**0.21**	**0.28**	0.11	**0.87**	1.00	−0.02	**0.23**	**0.48**	0.14	**0.28**
Hg_{F5}	0.05	**0.19**	0.00	−0.03	−0.02	1.00	**0.37**	0.07	0.07	0.10
OM%	**0.88**	**0.72**	**0.85**	0.12	**0.23**	**0.37**	1.00	**0.52**	**0.51**	**0.64**
FSF%	**0.54**	**0.38**	**0.42**	**0.42**	**0.48**	0.07	**0.52**	1.00	**0.30**	**0.59**
Concentration of diatoms valves	**0.59**	**0.33**	**0.57**	0.08	0.14	0.07	**0.51**	**0.30**	1.00	**0.54**
Wetness	**0.54**	**0.54**	**0.52**	**0.30**	**0.28**	0.10	**0.64**	**0.59**	**0.54**	1.00

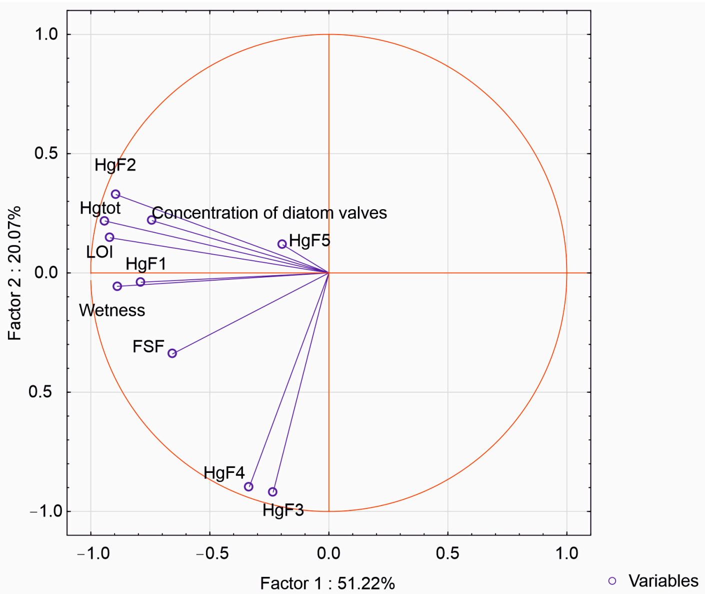

Figure 4. Principal component analysis (PCA) for concentration of diatom valves, Hg_{tot}, fractions of mercury (Hg_{F1}, Hg_{F2}, Hg_{F3}, Hg_{F4} and Hg_{F5}), LOI and wetness.

Among the analysed mercury fractions, Hg_{F2} and Hg_{F1} showed a linear correlation ($p < 0.05$) with the concentration of diatom valves deposited in the sediment of Puck Lagoon (Table 2). Mercury in complexes with organic matter (Hg_{F2}) was absorbed inside the diatom valves (r = 0.6, $p < 0.05$). Mercury in this form can biomagnify, which is of importance for the marine ecosystem. Hg_{F2} accounted for the largest percentage of Hg_{tot} (4–99%, mean 34%). Linear correlations showed that mercury in halide bonds (Hg_{F1}) was associated with diatoms to a lesser extent (although the correlation was statistically significant) (r = 0.3, $p < 0.05$). This is probably related to the fact that these forms of Hg adsorb to the surface of the diatoms, and because they are labile, they can be easily leached back into the surrounding supra-bottom or pore water.

PCA analysis showed that two factors explained 71% of the cases (Figure 4). The first factor was the accumulation of Hg in surface sediments. It confirmed the role of OM and FSF, as well as diatom valves, in the accumulation of Hg_{tot}, Hg_{F1}, Hg_{F2} and Hg_{F5} in sediments.

It shows that with the hydration of sediments and increasing content of organic matter, as well as diatom valves deposited in the sediment and the fine sediment fraction, the concentration of total mercury, primarily associated with halides and organic matter, increased. The second factor was the processes of the formation of stable HgS (Hg_{F3}) from $HgSO_4$ (Hg_{F4}). The resulting mercury sulphide was affected by the reducing conditions and appropriate concentration of sulphate ions and the presence of sulphur bacteria [90]. This suggests that the conditions for the formation of HgS from $HgSO_4$ were important throughout the study area.

3.3. Hg and Diatoms Salinity Regions

The retention of mercury in sediments in AREA I depended largely on the concentration of total mercury (Hg_{tot}) and brackish diatom species (r = 0.5, $p < 0.05$) (Table 3). Considering the analysed Hg fractions, diatoms preferring salinities of 5–20 PSU (mesohalobous) absorbed Hg_{F2} inside the cells (r = 0.4, $p < 0.05$). However, this community showed a better correlation with sulphide (Hg_{F3}) and Hg_{F4} (r = 0.5, $p < 0.05$) adsorbed on the surface of diatom valves. The high affinity of Hg_{F3} with the brackish diatom community was probably a result of the transformation of $HgSO_4$ adsorbed on the valves to HgS, as supported by the correlation of Hg_{F4} and Hg_{F3} (r = 0.9, $p < 0.05$).

A probable source of mercury in the northern Puck Lagoon (AREA I) could be the transport of Hg from remote, more saline parts of the bay. This is evidenced by the intercorrelation of all Hg fractions and the correlation of Hg forms with the mesohalobous group, except for mercury associated with halides (Hg_{F1}). This was further confirmed by carbon isotope studies by Jędruch et al. [87], where in the N part of Puck Lagoon, δ13C values in organic matter were typical of OM of marine origin. This is due to the occurring currents along the Hel Peninsula. These currents transport matter to Puck Lagoon from deeper areas of the Gulf of Gdańsk [39]. The lack of connection of salinity groups with Hg_{F1} may have been related to its dissolution and leaching back into the water. The fact that mercury is more readily accumulated by mesohalobes is further confirmed by an inverse correlation of total mercury (Hg_{tot}) which was observed with oligohalobous halophilous taxa (r = −0.5, $p < 0.05$), with an optimum at a salinity of 5 PSU [61] along the Hel Peninsula. Among the analysed mercury forms, it showed a negative correlation with Hg_{F3} (r = −0.5, $p < 0.05$) and Hg_{F4} (r = −0.5, $p < 0.05$) (Table 3). This may indicate a more persistent accumulation of mercury by marine diatom species, probably related to the strong affinity of Hg for sulphur. The occurrence of this group of diatoms in AREA I indicates substantially higher salinity in this region. On the other hand, sulphate concentrations in the water increase with increasing salinity. Previous studies indicate the transport of OM and FSF from more saline regions [87]. The studies presented here also indicate an important role for marine diatoms (from deeper regions) in the storage of Hg in this area of Puck Lagoon.

In the sediments collected from AREA II, an evident relationship between Hg_{tot}, particularly Hg_{F4} (mainly $HgSO_4$), and the euhalobous group was observed (r = 0.3, $p < 0.05$). Like in AREA I, mercury was more easily accumulated by species preferring saltier waters. The higher frequency of euhalobous taxa was mainly associated with higher Hg concentrations in the Kuźnica Deep and near the Rzucewo Deep. These regions were the deepest where the analysed material was deposited.

Table 3. Correlation coefficients determined for Hg_{tot}, labile and stable fractions of mercury with diatoms salinity groups in distinguished areas. Statistically significant correlations are marked in bold: Eu—euhalobous. Meso—mesohalobous. Oh—oligohalobous halophilous. Oi—oligohalobous indifferent.

		Hg_{tot}	Hg_{F1}	Hg_{F2}	Hg_{F3}	Hg_{F4}	Hg_{F5}
AREA I	Hg_{tot}	1.00	0.01	**0.68**	**0.65**	0.45	0.21
	Hg_{F1}	0.01	1.00	−0.30	0.55	**0.73**	**0.62**
	Hg_{F2}	**0.68**	−0.30	1.00	0.08	−0.01	−0.24
	Hg_{F3}	**0.65**	0.55	0.08	1.00	**0.91**	0.12
	Hg_{F4}	0.45	**0.73**	−0.01	**0.91**	1.00	0.48
	Hg_{F5}	0.21	**0.62**	−0.24	0.12	0.48	1.00
	Eu	0.01	0.07	0.01	−0.19	−0.07	0.16
	Meso	**0.52**	0.10	**0.46**	**0.52**	**0.54**	0.26
	Oh	**−0.54**	−0.14	−0.40	**−0.51**	**−0.58**	−0.37
	Oi	−0.09	−0.06	−0.29	0.19	0.14	−0.17
AREA II	Hg_{tot}	1.00	**0.69**	**0.77**	**0.81**	**0.78**	**0.48**
	Hg_{F1}	**0.69**	1.00	**0.70**	**0.56**	**0.52**	0.05
	Hg_{F2}	**0.77**	**0.70**	1.00	−0.03	0.16	−0.01
	Hg_{F3}	**0.81**	**0.56**	−0.03	1.00	**0.69**	−0.04
	Hg_{F4}	**0.78**	**0.52**	0.16	**0.69**	1.00	−0.07
	Hg_{F5}	**0.48**	0.05	−0.01	−0.04	−0.07	1.00
	Eu	**0.33**	0.03	−0.06	0.20	**0.36**	0.02
	Meso	−0.13	0.23	0.05	0.16	−0.04	0.01
	Oh	0.00	−0.09	−0.10	−0.06	−0.04	−0.02
	Oi	−0.13	−0.14	0.10	−0.23	−0.18	0.00
AREA III	Hg_{tot}	1.00	**0.52**	**0.44**	**0.93**	**0.91**	**0.50**
	Hg_{F1}	**0.52**	1.00	**0.54**	**0.66**	**0.70**	**0.46**
	Hg_{F2}	**0.44**	**0.54**	1.00	**0.57**	**0.56**	0.28
	Hg_{F3}	**0.93**	**0.66**	**0.57**	1.00	**0.80**	**0.60**
	Hg_{F4}	**0.91**	**0.70**	**0.56**	**0.80**	1.00	**0.52**
	Hg_{F5}	**0.50**	**0.46**	0.28	**0.60**	**0.52**	1.00
	Eu	−0.01	−0.22	**−0.35**	−0.11	−0.19	−0.01
	Meso	0.03	−0.03	0.18	−0.07	−0.01	−0.13
	Oh	**−0.40**	−0.07	0.17	**−0.32**	−0.17	**−0.36**
	Oi	**0.31**	0.19	−0.12	**0.39**	0.22	**0.44**

The analysis of sediments collected from AREA III (Figure 1) showed a relationship between total mercury as well as its individual fractions with freshwater diatom flora. In this part of Puck Lagoon, the inflow of river water has a significant influence on the occurrence of diatom flora, as well as on the level of chemical substance concentrations. Rivers are an important source of terrestrial Hg inputs to the bay [91]. They probably influenced the potential accumulation of Hg_{tot} with oligohalobous indifferent taxa preferring low salinity (r = 0.3, $p < 0.05$) in this area. The dominant role of river transport and freshwater diatom communities is also supported by the negligible relationship between mercury and other diatom groups. This may have influenced a decrease in the correlation with marine diatom flora. Among the analysed mercury fractions, the freshwater community showed a significant correlation with stable forms of mercury: Hg_{F3} (r = 0.4, $p < 0.05$) and Hg_{F5} (r = 0.4, $p < 0.05$) (Figure 3). According to the study by Saniewska et al. [83], labile forms of mercury were primarily present in the sediments of rivers flowing into Puck Lagoon (Płutnica, Reda). The HgS form was present to a smaller extent. In comparison with Reda and Płutnica, however, higher concentrations of stable forms were measured in the sediments of the Gizdepka River. This suggests that the diatom flora occurring in AREA III are associated with freshwater and prefers terrestrial mercury. As with AREA I, an inverse correlation was also found between total mercury (Hg_{tot}) and oligohalobous halophilous taxa (r = −0.4, $p < 0.05$). Moreover, this diatom group showed an inverse correlation with Hg_{F3} (r = −0.3, $p < 0.05$). There was also an inverse correlation with euhalobous taxa in this region (r = −0.3, $p < 0.05$). The negative correlation with groups of oligohalobous halophilous and euhalobous taxa suggests that the increased salinity does not favour Hg binding to diatom flora in the southern part of Puck Lagoon. A similar relationship was observed in AREA I.

3.4. Mercury Versus Diatoms

Previous studies regarding Hg and the benthic and planktic diatom flora have been focused on the effects of mercury in breeding species under laboratory conditions [20,21]. The present study observed the association of mercury with benthic species and planktic *Chaetoceros* resting spores that were a minority in each region (AREA II; AREA III). An exception is AREA I, located along the Hel Peninsula, where the mercury concentration was seen not to deposit on particular diatom species, as indicated by the lack of correlation with particular diatom species.

AREA II, covering the central part of Puck Lagoon, was characterised by a correlation of mercury with planktic *Chaetoceros* resting spores and benthic taxa. The correlating salinity groups included the following taxa: euhalobous (*Chaetoceros diadema* RS (Ehrenberg) Gran, *Rhabdonema adriaticum* (C.A. Agardh) Kützing, *R. minutum* Kützing), mesohalobous (*Chaetoceros* spp. RS, *Navicula phyllepta* Kützing), oligohalobous halophilous (*Pseudostaurosira perminuta* (Grunow) Sabbe and Wyverman), oligohalobous indifferent (*Epithemia adnata* (Kützing) Brébisson, *E. frickei* Krammer, *E. goeppertina* Hilse, *Staurosirella martyi* (Héribaud) Lange-Bertalot, *F. inflata* var. *istvantfyi* (Pantoscek) Hustedt, *Navicula cryptocephala* Kützing and *Surirella brebissonii* Krammer and Lange-Bertalot). All of these species showed a moderate to high but statistically significant correlation ($p < 0.05$) with Hg_{F2} and with Hg_{F1} (Table 4).

In AREA II, most species reached the highest frequency in the vicinity of the Kuźnica Deep, ranging from 0.36% to 20.5%. The highest concentrations of Hg_{tot}, Hg_{F1} and Hg_{F2} were also measured in this part of Puck Lagoon. A prominent taxon was planktic *Chaetoceros seiracantus* RS. This taxon is potentially associated with Hg_{F5} (r = 0.43, $p < 0.05$). This suggests that it incorporated toxic Hg into the valves.

A significant correlation (r = 0.52, $p < 0.05$) existed between Hg_{F1} and benthic taxa *Epithemia goeppertina*, *Fragilaria inflata* var. *istvantfyi*, *Pseudostaurosira perminuta* and *Rhabdonema adriaticum*. Moreover, an evident relationship was observed between this form of mercury and benthic *Epithemia adnata* (r = 0.32, $p < 0.05$), *Chaetoceros* spp. RS (r = 0.31, $p < 0.05$) and *Staurosirella martyi* (r = 0.33, $p < 0.05$). The benthic brackish species *Navicula phyllepta*, most abundantly observed near Swarzewo, is also worth attention (PL40, 24%). The species absorbed Hg_{F1} onto its surface (r = 0.6, $p < 0.05$). This suggests that all species correlating with Hg_{F1} could be live diatoms. Hg_{F1} is a labile fraction that is most likely leached from dead diatoms into the surrounding water.

Moreover, in contrast to other species, *N. phyllepta* showed a significant correlation with adsorbed Hg_{F4} (r = 0.4, $p < 0.05$). In the waters of Puck Lagoon, there are regions with reducing conditions, i.e., conditions favourable for the formation of mercury sulphide, (Hg_{F3}) with which *N. phyllepta* showed a high correlation (r = 0.74, $p < 0.05$). Previous studies on the effects of mercury on diatoms have shown that mercury reaching diatom frustules is transformed to less toxic forms through reacting with proteins [20,92,93]. This may indicate that *N. phyllepta* probably protects itself from Hg toxicity in this way.

The mentioned taxa (*Chaetoceros* spp. RS, *Epithemia adnata*, *Epithemia goeppertina*, *Fragilaria inflata* var. *istvantfyi*, *Fragilaria maryti* and *Rhabdonema adriaticum*) are species that are relatively larger in terms of the size of their valve and therefore can accumulate more Hg. Only *Pseudostaurosira perminuta* and *Navicula phyllepta* are smaller taxa, and therefore, they have a smaller adsorption area. *Chaetoceros* in the study material are associated with marine and brackish waters [94]. Resting spores are planktic taxa. Their connection with Hg, however, may result from their long persistence in the sediment with very high mercury concentrations.

Table 4. Correlation coefficients determined for Hg_{tot}, labile and stable fractions of mercury with diatoms species in distinguished areas. Statistically significant correlations are marked in bold.

		$\mathbf{Hg_{tot}}$	$\mathbf{Hg_{F1}}$	$\mathbf{Hg_{F2}}$	$\mathbf{Hg_{F3}}$	$\mathbf{Hg_{F4}}$	$\mathbf{Hg_{F5}}$
AREA II	Hg_{tot}	1.00	**0.77**	**0.99**	0.11	**0.27**	0.04
	Hg_{F1}	**0.77**	1.00	**0.70**	**0.56**	**0.52**	0.05
	Hg_{F2}	**0.99**	**0.70**	1.00	−0.03	0.16	−0.01
	Hg_{F3}	0.11	**0.56**	−0.03	1.00	**0.69**	−0.04
	Hg_{F4}	**0.27**	**0.52**	0.16	**0.69**	1.00	−0.07
	Hg_{F5}	0.04	0.05	−0.01	−0.04	−0.07	1.00
	Chaetoceros diadema RS	**0.52**	**0.37**	**0.52**	−0.08	0.09	−0.02
	Chaetoceros seiracanthus RS	0.03	−0.01	0.01	−0.01	0.03	**0.44**
	Chaetoceros spp. RS	**0.54**	**0.31**	**0.56**	−0.09	0.03	0.03
	Epithemia adnata	**0.48**	**0.32**	**0.49**	−0.10	0.04	0.00
	Epithemia frickei	**0.47**	0.20	**0.50**	−0.07	−0.12	−0.03
	Epithemia goeppertiana	**0.62**	**0.53**	**0.61**	−0.06	0.21	−0.01
	Staurosirella martyi	**0.70**	**0.34**	**0.73**	−0.16	−0.01	−0.03
	Fragilaria inflata var. *instvantfyi*	**0.62**	**0.53**	**0.61**	−0.06	0.21	−0.01
	Navicula cryptocephala	**0.54**	0.24	**0.57**	−0.06	−0.09	−0.02
	Navicula phyllepta	0.12	**0.64**	0.03	**0.75**	**0.35**	0.00
	Pseudostaurosira perminuta	**0.62**	**0.53**	**0.61**	−0.06	0.21	−0.01
	Rhabdonema adriaticum	**0.62**	**0.53**	**0.61**	−0.06	0.21	−0.01
	Rhabdonema minutum	**0.49**	**0.34**	**0.49**	−0.05	0.11	0.01
	Surirella brebissonii	**0.49**	**0.34**	**0.49**	−0.05	0.11	0.01
AREA III	Hg_{tot}	1.00	**0.75**	**0.70**	**0.95**	**0.92**	**0.58**
	Hg_{F1}	**0.75**	1.00	**0.54**	**0.66**	**0.70**	**0.46**
	Hg_{F2}	**0.70**	**0.54**	1.00	**0.57**	**0.56**	0.28
	Hg_{F3}	**0.95**	**0.66**	**0.57**	1.00	**0.80**	**0.60**
	Hg_{F4}	**0.92**	**0.70**	**0.56**	**0.80**	1.00	**0.52**
	Hg_{F5}	**0.58**	**0.46**	0.28	**0.60**	**0.52**	1.00
	Chaetoceros seiracanthus RS	**0.38**	0.25	0.27	**0.44**	0.25	0.27
	Staurosirella martyi	**0.44**	**0.58**	0.33	0.36	**0.44**	0.19

AREA III, including the southern Puck Lagoon, showed a significant correlation of benthic species *Staurosirella martyi* with Hg_{tot} (r = 0.44, $p < 0.05$), Hg_{F1} (r = 0.57, $p < 0.05$), Hg_{F2} (r = 0.32, $p < 0.05$), Hg_{F3} (r = 0.36, $p < 0.05$) and Hg_{F4} (r = 0.44, $p < 0.05$). This phenomenon was also observed in AREA II. Excessive nutrient supply contributed to the formation of diatom resting spores (in AREA III *Chaetoceros seiracantus* RS), efficiently accumulating Hg. Importantly, in the southern part of Puck Lagoon, they are potentially associated with the stable form of mercury Hg_{F3} (r = 0.43, $p < 0.05$) which does not biomagnify.

The association of individual species in AREA II and AREA III with mercury could have been from relationships with high Hg concentrations in the area where these species occur (Kuźnica Deep; Swarzewo river mouth). This would explain the lack of connections of Hg_{tot} and individual forms of mercury with diatom species found in AREA I.

4. Conclusions

Our results showed that diatom taphocoenoses (r = 0.6, $p < 0.05$), next to organic matter (r = 0.9, $p < 0.05$) and fine sediment fraction (r = 0.5, $p < 0.05$), are an important element of the marine environment in terms of absorbing and adsorbing mercury. It seems that, depending on environmental conditions and diatom species composition, they can interact with mercury in several ways. The linear correlation showed a connection of salinity groups with adsorbed (HgS; $HgSO_4$) and absorbed (built into OM) mercury. In the northern part of Puck Lagoon (AREA I) and in areas distant from the coastal zone (AREA II), the forms of mercury showed a statistically significant correlation with species preferring higher salinity (euhalobous and mesohalobous taxa). In these diatom groups, Hg was retained as mercury sulphide (Hg_{F3}; r = 0.5, $p < 0.05$) and mercury sulphate (Hg_{F4}; r = 0.5, $p < 0.05$). In contrast, a higher affinity of mercury with freshwater diatom flora was observed near river mouths (AREA III; r = 0.3, $p < 0.05$).

This is the first study to demonstrate an association between mercury fractions and individual diatom species living in the marine environment. Correlating species were associated with deposition sites (Kuźnica Deep) and the river inflow of mercury (Płutnica, Reda, Gizdepka), at which the highest Hg concentrations were measured in the entire Puck Lagoon (approximately 20–100 ng g^{-1}). Species showing high statistically significant correlation with mercury forms were a minority in the analysed diatom community of Puck Lagoon. These species, however, primarily correlated with mercury forms (Hg_{F2}; r = 0.5, r = 0.6, $p < 0.05$) potentially undergoing biomagnification, enriching subsequent levels of the food web with mercury.

Taking into account the different degrees of Hg_{tot} and its fractions storage by living and dead diatoms, it seems reasonable to conduct analogous studies seasonally—before diatom bloom, during diatom bloom, immediately after diatom bloom and several months after diatom bloom—to determine the percentage of live and dead of individual species.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15223907/s1, Information about spatial variability of individual diatom ecological groups; Figure S1: Sediment grain size in the inner Puck Bay; Figure S2: Characteristic of diatom taxa; Figure S3: Percentage content of the diatom ecological groups; Figure S4: Spatial variability of the: A—concentrations of Hg_{to}t (ng g^{-1}), B—%FSF, C—%OM and D—contents of diatom valves (valve × 105 g^{-1}); Table S1: Environmental conditions during collection of study material; Table S2: Ecological preferences of diatoms in terms of habitat, salinity, pH, trophic and saprobic status. References [95–97] are citied in the Supplementary Materials.

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References

1. Zahir, F.; Rizwi, S.J.; Haq, S.K.; Khan, R.H. Low dose mercury toxicity and human health. *Environ. Toxicol. Pharmacol.* **2005**, *20*, 351–360. [CrossRef] [PubMed]
2. Mukherjee, A.B.; Zevenhoven, R.; Brodersen, J.; Hylander, L.D.; Bhattacharya, P. Mercury in waste in the European Union: Sources, disposal methods and risks. Resources. *Conserv. Recycl.* **2004**, *42*, 155–182. [CrossRef]
3. Wyn, B.; Kidd, K.A.; Burgess, N.M.; Curry, R.A. Mercury biomagnification in the food webs of acidic lakes in Kejimkujik National Park and National Historic Site, Nova Scotia. *Can. J. Fish. Aquat. Sci.* **2009**, *66*, 1532–1545. [CrossRef]
4. Falandysz, J. Mercury concentrations in benthic animals and plants inhabiting the Gulf of Gdańsk, Baltic Sea. *Sci. Total Environ.* **1994**, *141*, 45–49. [CrossRef]
5. Lavoie, R.A.; Jardine, T.D.; Chumchal, M.M.; Kidd, K.A.; Campbell, L.M. Biomagnification of mercury in aquatic food webs: A worldwide meta-analysis. *Environ. Sci. Technol.* **2013**, *47*, 13385–13394. [CrossRef]
6. Bełdowska, M.; Zgrundo, A.; Kobos, J. Mercury in the Diatoms of Various Ecological Formations. *Water Air Soil Pollut.* **2018**, *229*, 168. [CrossRef]

7. Kang, W.; Chen, G.; Wang, J.; Huang, L.; Li, R.; Hu, Y.; Tao, J.; Blais, J.M.; Smol, J.P. Assessing the impact of long-term changes in climate and atmospheric deposition on a shallow alpine lake from southeast Tibet. *Sci. Total Environ.* **2019**, *650*, 713–724. [CrossRef]
8. Gosnell, K.J.; Dam, H.G.; Mason, R.P. Mercury and methylmercury uptake and trophic transfer from marine diatoms to copepods and field collected zooplankton. *Mar. Environ. Res.* **2021**, *170*, 105446. [CrossRef]
9. Round, F.E. *The Biology of Algae*; Cambridge University Press: Cambridge, UK; London, UK; New York, NY, USA; New Rochelle, NY, USA; Melbourne, Australia; Sydney, Australia, 1981; p. 653.
10. Leblanc, K.; Arístegui, J.; Armand, L.; Assmy, P.; Beker, B.; Bode, A.; Breton, E.; Cornet, V.; Gibson, J.; Gosselin, M.P.; et al. A global diatom database—Abundance, biovolume and biomass in the world ocean. *Earth Syst. Sci. Data Discuss.* **2012**, *5*, 147–185. [CrossRef]
11. Legrand, C.; Fridolfsson, E.; Bertos-Fortis, M.; Lindehoff, E.; Larsson, P.; Pinhassi, J.; Andersson, A. Interannual variability of phyto-bacterioplankton biomass and production in coastal and offshore waters of the Baltic Sea. *AMBIO* **2015**, *44*, 427–438. [CrossRef]
12. Staniszewska, M.; Nehring, I.; Zgrundo, A. The role of phytoplankton composition, biomass and cell volume in accumulation and transfer of endocrine disrupting compounds in the Southern Baltic Sea (The Gulf of Gdansk). *Environ. Pollut.* **2015**, *207*, 319–328. [CrossRef] [PubMed]
13. Lee, C.-S.; Fisher, N.S. Bioaccumulation of methylmercury in a marine diatom and the influence of dissolved organic matter. *Mar. Chem.* **2017**, *197*, 70–79. [CrossRef] [PubMed]
14. Schmitt, D.; Müller, A.; Csögör, Z.; Frimmel, F.H.; Posten, C. The adsorption kinetics of metal ions onto different microalgae and siliceous earth. *Water Res.* **2001**, *35*, 779–785. [CrossRef] [PubMed]
15. Outridge, P.M.; Stern, G.A.; Hamilton, P.B.; Percival, J.B.; McNeely, R.; Lockhart, W.L. Trace metal profiles in the varved sediment of an Arctic lake. *Geochim. Cosmochim. Acta* **2005**, *69*, 4881–4894. [CrossRef]
16. Zaferani, S.; Pérez-Rodríguez, M.; Biester, H. Diatom ooze—A large marine mercury sink. *Science* **2018**, *361*, 797–800. [CrossRef]
17. Bérard, A.; Dorigo, U.; Mercier, I.; Becker-van Slooten, K.; Grandlean, D.; Leboulonger, C. Comparison of the eco toxicological impact of the traizines Irgarol 1051 and atrazine on micro algal cultures and natural micro algal communities in Lake Geneva. *Chemosphere* **2003**, *53*, 935–944. [CrossRef]
18. Anantharaj, K.; Govindasamy, C.; Natanamurugaraj, G.; Jeyachandran, S. Effect of heavy metals on marine diatom Amphora coffeaeformis (Agardh. Kutz). *Glob. J. Environ. Res.* **2011**, *5*, 112–117.
19. Deng, G.; Zhang, T.; Yang, L.; Wang, Q. Studies of bio uptake and transformation of mercury by a typical unicellular diatom Phaeodactylum tricornutum. *Chin. Sci. Bull.* **2013**, *58*, 256–265. [CrossRef]
20. Naseri, A.; Saadatmand, S.; Noroozi, M.; Asri, Y.; Iranbakhsh, A. Study the mercury biosorption by unicellular diatom Nitzschia capitellata Hustedt. *Mod. Phytomorphol.* **2020**, *14*, 70–76. [CrossRef]
21. Zhong, H.; Wang, W.-X. Controls of Dissolved Organic Matter and Chloride on Mercury Uptake by a Marine Diatom. *Environ. Sci. Technol.* **2009**, *43*, 8998–9003. [CrossRef]
22. Cyberski, J. Hydrologia zlewiska. In *Zatoka Pucka*; Korzeniewski, K., Ed.; Fundacja Rozwoju UG: Gdańsk, Poland, 1993; pp. 40–71. (In Polish)
23. Zaborska, A.; Siedlewicz, G.; Szymczycha, B.; Dzierzbicka-Głowacka, L.; Pazdro, K. Legacy and emerging pollutants in the Gulf of Gdańsk (southern Baltic Sea)—Loads and distribution revisited. *Mar. Pollut. Bull.* **2019**, *139*, 238–255. [CrossRef] [PubMed]
24. Chubarenko, B.; Woelfel, J.; Hofmann, J.; Aldag, S.; Beldowski, J.; Burlakovs, J.; Garrels, T.; Gorbunova, J.; Guizani, S.; Kupczyk, A.; et al. Converting beach wrack into a resource as a challenge for the Baltic Sea (an overview). *Ocean Coast. Manag.* **2021**, *200*, 105413. [CrossRef]
25. Pacyna, E.G.; Pacyna, J.M.; Steenhuisen, F.; Wilson, S. Global anthropogenic mercury emission inventory for 2000. *Atmos. Environ.* **2006**, *40*, 4048–4063. [CrossRef]
26. Uścinowicz, S.; Szefer, P.; Sokołowski, K. Trace elements in Baltic Sea sediments. In *Geochemistry of Surface Sediments of the Baltic Sea*; Uścinowicz, S., Ed.; Polish Geological Survey: Warsaw, Poland, 2011; pp. 214–274.
27. Kwasigroch, U.; Bełdowska, M.; Jędruch, A.; Saniewska, D. Coastal erosion—A "new" land-based source of labile mercury to the marine environment. *Environ. Sci. Pollut. Res.* **2018**, *25*, 28682–28694. [CrossRef]
28. Jędruch, A.; Falkowska, L.; Saniewska, D.; Durkalec, M.; Nawrocka, A.; Kalisińska, E.; Pacyna, J.M. Status and trends of mercury pollution of the atmosphere and terrestrial ecosystems in Poland. *Ambio* **2021**, *50*, 1698–1717. [CrossRef]
29. Pempkowiak, J. *Zarys Geochemii Morskiej*; University of Gdansk Press: Gdańsk, Poland, 1997; p. 171. (In Polish)
30. Pniewski, F.F.; Biskup, P.; Bubak, I.; Richard, P.; Latała, A.; Blanchard, G. Photo-regulation in microphytobenthos from intertidal mudflats and non-tidal coastal shallows. Estuarine. *Coast. Shelf Sci.* **2015**, *152*, 153–161. [CrossRef]
31. Bąk, M.; Witkowski, A.; Żelazna-Wieczorek, J.; Wojtal, A.Z.; Szczepocka, E.; Szulc, K.; Szulc, B. *Klucz do Oznaczania Okrzemek w Fitobentosie na Potrzeby Oceny Stanu Ekologicznego wód Powierzchniowych w Polsce*; Główny Inspektorat Ochrony Środowiska: Warszawa, Poland, 2012; pp. 20–25. (In Polish)
32. Urbański, J.; Grusza, G.; Chlebus, N. Fizyczna typologia dna Zatoki Gdańskiej, Atlas Cyfrowy, Sprawozdanie merytoryczne z realizacji projektu badawczego nr 2 PO4E00629. 2007. (In Polish)

33. Szarafin, T.; Karwik, A.; Uścinowicz, S.; Fac-Beneda, J.; Nowacki, J.; Boniecka, H.; Gajda, A.; Gawlik, W.; Meissner, W.; Bzoma, S. *Zbiorcze Sprawozdanie z Analizy Dostępnych Danych i Przeprowadzonych Inwentaryzacji Przyrodniczych (Zebranie i Analiza Wyników Inwentaryzacji, Materiałów Niepublikowanych i Opracowań Publikowanych, Przydatnych do Sporządzenia Projektów Planów)*; Zatoka Pucka (PLB 220005); Wydawnictwa Wewnętrzne Instytutu Morskiego w Gdańsku Nr 6823. 2014, pp. 190–247. Available online: https://www.researchgate.net/publication/318502945_Fizyczna_typologia_dna_Zatoki_Gdanskiej (accessed on 11 October 2023). (In Polish)
34. Szymczak, E. The Role of River Sediment Inflow in Present Sedimentation Proceses in the Puck Lagoon. Ph.D. Thesis, Univ. Gdańsk, Gdańsk, Poland, 2006. (In Polish)
35. Majewski, A. *Charakterystyka Hydrologiczna Estuariowych wód u Polskiego Wybrzeża*; Prace PIHM: Warszawa, Poland, 1972; Volume 105, pp. 3–40. (In Polish)
36. Nowacki, J. Morfometria zatoki. In *Zatoka Pucka*; Korzeniewski, K., Ed.; Fundacja Rozwoju UG: Gdańsk, Poland, 1993; pp. 71–78. (In Polish)
37. Cieślikiewicz, W.; Jędrasik, J. Falowanie wiatrowe w Zatoce Puckiej. In *Zatoka Pucka*; Bolałek, J., Burska, D., Eds.; Wydawnictwo Uniwersytetu Gdańskiego: Gdańsk, Poland, 2022; Tom I; pp. 213–243. (In Polish)
38. Jędrasik, J.; Cieślikiewicz, W. Cyrkulacja wód w Zatoce Puckiej. In *Zatoka Pucka*; Bolałek, J., Burska, D., Eds.; Wydawnictwo Uniwersytetu Gdańskiego: Gdańsk, Poland, 2022; Tom I; pp. 183–212. (In Polish)
39. Nowacki, J. Cyrkulacja i wymiana wód. In *Zatoka Pucka*; Korzeniewski, K., Ed.; Fundacja Rozwoju UG: Gdańsk, Poland, 1993; pp. 181–205. (In Polish)
40. Nowacki, J. Termika, zasolenie i gęstość wody. In *Zatoka Pucka*; Korzeniewski, K., Ed.; Fundacja Rozwoju UG: Gdańsk, Poland, 1993; pp. 79–111. (In Polish)
41. Szymczak, E.; Rucińska, M.; Szmytkiewicz, A. Osady powierzchniowe Zatoki Puckiej. In *Zatoka Pucka*; Bolałek, J., Burska, D., Eds.; Wydawnictwo Uniwersytetu Gdańskiego: Gdańsk, Poland, 2022; Tom I; pp. 265–279. (In Polish)
42. Graca, B.; Witek, Z.; Burska, D.; Białkowska, I.; Pawelec, A.; Bolałek, J. Porewater nutrients (phosphate, ammonia and silicate) in the eastern part of the southern Baltic Sea. *Oceanol. Hydrobiol. Stud.* **2006**, *35*, 237–256.
43. Szmytkiewicz, A. Zróżnicowanie Tempa Sedymentacji Współczesnych Osadów Dennych Zatoki Puckiej. Ph.D. Thesis, Univ. Gdańsk, Gdańsk, Poland, 2016. (In Polish).
44. Szmytkiewicz, A.; Zalewska, T. Sediment deposition and accumulation rates determined by sediment trap and 210Pb isotope methods in the Outer Puck Bay (Baltic Sea). *Oceanologia* **2014**, *56*, 85–106. [CrossRef]
45. Battarbee, R.W. Diatom analysis. In *Handbook of Holocene Palaeoecology and Palaeohydrology*; Berglund, B.E., Ed.; John Wiley and Sons Ltd.: London, UK, 1986; pp. 527–570.
46. Bodén, P. Reproducibility in the Random Settling Method for Quantitative Diatom Analysis. *Micropaleontology* **1991**, *37*, 313–319. [CrossRef]
47. Schrader, H.; Gersonde, R. Diatoms and silico agellates in the eight meters sections of the lower Pleistocene at Capo Rossello. *Utrecht Micropaleontol. Bull.* **1978**, *17*, 129–176.
48. Hustedt, F. Die Kieselalgen Deutschlands, Österreichs und der Schweiz 1–3. In *Kryptogamen ora von Deutschland, Österreich und der Schweiz 7*; Rabenhorsts, L., Ed.; Akademische Verlerlagsbuchhandlung: Leipzig, Germany, 1927–1966.
49. Krammer, K.; Lange-Bertalot, H. Bacillariophyceae. 1. Teil: Naviculaceae. In *Süßwasserflora von Mitteleuropa 2/1*; Ettl, H., Gerloff, J., Heynig, H., Mollenhauer, D., Eds.; G. Fischer: Stuttgart, Germany; New York, NY, USA, 1986; p. 876.
50. Krammer, K.; Lange-Bertalot, H. Bacillariophyceae. 2. Teil: Bacillariaceae, Epithemiaceae, Surirellaceae. In *Süßwasserflora von Mitteleuropa 2/2*; Ettl, H., Gerloff, J., Heynig, H., Mollenhauer, D., Eds.; G. Fischer: Stuttgart, Germany; New York, NY, USA, 1988; p. 596.
51. Krammer, K.; Lange-Bertalot, H. Bacillariophyceae. 3. Teil: Centrales, Fragilariaceae, Eunotiaceae. In *Süßwasserflora von Mitteleuropa 2/3*; Ettl, H., Gerloff, J., Heynig, H., Mollenhauer, D., Eds.; G. Fischer: Stuttgart, Germany; Jena, Germany, 1991; p. 576.
52. Krammer, K.; Lange-Bertalot, H. Bacillariophyceae. 4. Teil: Achnanthaceae. Kritische Ergänzungen zu Navicula (Lineolatae) und Gomphonema. Gesamtliteraturverzeichnis. Teil 1–4. In *Süßwasserflora von Mitteleuropa 2/4*; Ettl, H., Gerloff, J., Heynig, H., Mollenhauer, D., Eds.; Fischer: Stuttgart, Germany; Jena, Germany, 1991; p. 437.
53. Pankow, H. *Ostsee-Algenflora*; Fischer: Jena, Germany, 1990.
54. Lange-Bertalot, H. Navicula sensu stricto. 10 Genera Separated from Navicula sensu lato, Frustulia. In *Diatoms of Europe 2*; Lange-Bertalot, H., Ed.; A.R.G. Gantner Verlag K.G.: Ruggell, Liechtenstein, 2001; p. 526.
55. Snoeijs, P. *Intercalibration and Distribution of Diatom Species in the Baltic Sea*; Opulus Press: Uppsala, Sweden, 1993; Volume 1.
56. Snoeijs, P.; Balashova, N. *Intercalibration and Distribution of Diatom Species in the Baltic Sea*; Opulus Press: Uppsala, Sweden, 1998; Volume 5.
57. Snoeijs, P.; Kasperoviciene, J. *Intercalibration and Distribution of Diatom Species in the Baltic Sea*; Opulus Press: Uppsala, Sweden, 1996; Volume 4.
58. Snoeijs, P.; Potapova, M. *Intercalibration and Distribution of Diatom Species in the Baltic Sea*; Opulus Press: Uppsala, Sweden, 1995; Volume 3.
59. Snoeijs, P.; Vilbaste, S. *Intercalibration and Distribution of Diatom Species in the Baltic Sea*; Opulus Press: Uppsala, Sweden, 1994; Volume 2.

60. Witkowski, A.; Lange-Bertalot, H.; Metzeltin, D. *Diatom Flora of Marine Coasts I*; A.R.G. Gantner Verlag K.G.: Ruggell, Liechtenstein, 2000.
61. Kolbe, R.W. *Zur Ökologie, Morphologie und Systematik der Brachwasser-Diatomeen*; Pflanzenforschung: Berlin, Germany, 1927; 146p.
62. Vos, P.C.; De Wolf, H. Diatoms as a tool for reconstructing sedimentary environments in coastal wetlands; methodological aspects. *Hydrobiologia* **1993**, *269*, 285–296. [CrossRef]
63. Kolkwitz, R.; Marsson, M. Őkologie der pflanzlichen Saprobien. *Ber. Dtsch. Bot. Ges.* **1908**, *26*, 505–519.
64. Reis, A.T.; Coelho, J.P.; Rodrigues, S.M.; Rocha, R.; Davidson, C.M.; Duarte, A.C.; Pereira, E. Development and validation of a simple thermo-desorption technique for mercury speciation in soils and sediments. *Talanta* **2012**, *99*, 363–368. [CrossRef] [PubMed]
65. Jędruch, A.; Bełdowska, M.; Kwasigroch, U.; Normant-Saremba, M.; Saniewska, D. Mercury fractionation in marine macrofauna using thermodesorption technique: Method and its application. *Talanta* **2018**, *189*, 534–542. [CrossRef]
66. Bełdowska, M.; Saniewska, D.; Gębka, K.; Kwasigroch, U.; Korejwo, E.; Kobos, J. Simple screening technique for determination of adsorbed and absorbed mercury in particulate matter in atmospheric and aquatic environment. *Talanta* **2018**, *182*, 340–347. [CrossRef]
67. Wilman, B.; Saniewska, D.; Pyta, H.; Wysiecki, D.; Bełdowska, M. Mercury fractionation—Problems in method application. *Mar. Pollut. Bull.* **2023**, *187*, 114560. [CrossRef]
68. Kwasigroch, U.; Bełdowska, M.; Jędruch, A.; Łukawska-Matuszewska, K. Distribution and bioavailability of mercury in the surface sediments of the Baltic Sea. *Environ. Sci. Pollut. Res.* **2021**, *28*, 35690–35708. [CrossRef]
69. Łęczyński, L.; Szymczak, E. *Fizyczne, Biologiczne i Chemiczne Badania Morskich Osadów Dennych*; Jerzy, B., Ed.; Wydawnictwo Uniwersytetu Gdańskiego: Gdańsk, Poland, 2010; pp. 69–118, ISBN 978-83-7326-789-3. (In Polish)
70. Myślińska, E. *Laboratoryjne Badania Gruntów*; Wydawnictwo Naukowe PWN: Warszawa, Poland, 2001; ISBN 8301134305. (In Polish)
71. Gradziński, R.; Kostecka, A.; Radomski, A.; Unrug, R. *Zarys Sedymentologii*; Wydawnictwa Geologiczne: Warszawa, Poland, 1986.
72. Grimm, E.C. *Tilia*, version 2.0.37; Software; Illinois State Museum: Springfield, IL, USA, 2011.
73. Witak, M.; Pędziński, J.; Oliwa, S.; Hetko, D. Biodiversity of benthic diatom flora in the coastal zone of the Puck Bay (southern Baltic Sea): A case study of Hel Peninsula. *Oceanol. Hydrobiol. Stud.* **2020**, *49*, 304–318. [CrossRef]
74. Zgrundo, A.; Dziengo-Czaja, M.; Bubak, I.; Bogaczewicz-Adamczak, B. Studies of the biodiversity of contemporary diatom assemblages in the Gulf of Gdańsk. *Oceanol. Hydrobiol. Stud.* **2008**, *38* (Suppl. 2), 1–15.
75. Witak, M. *Application of Diatom Biofacies in Reconstructing the Evolution of Sedimentary Basins. Records from the Southern Baltic Sea Differentiated by the Extent of the Holocene Marine Transgressions and Human Impact*; Diatom Monographs 12; Witkowski, A., Ed.; A.R.G. Gantner Verlag, K.G.: Ruggell, Liechtenstein, 2010; p. 295.
76. Bogaczewicz-Adamczak, B.; Kłosińska, D.; Zgrundo, A. Diatoms as indicators of water pollution in the coastal zone of the Gulf of Gdansk (Southern Baltic Sea). *Oceanol. Stud.* **2001**, *30*, 59–75.
77. Witkowski, A. Diatoms of the Puck bay coastal shallows (Poland, Southern Baltic). *Nord. J. Bot.* **1991**, *11*, 689–701. [CrossRef]
78. Dziengo-Czaja, M.; Koss, J.; Matuszak, A. Teratological forms of diatoms (Bacillariophyceae) as indicators of water pollution in the western part of Puck Bay (southern Baltic Sea). *Oceanol. Hydrobiol. Stud.* **2008**, *37*, 119–132. [CrossRef]
79. Hetko, D.; Witak, M.; Oliwa, S. Biodiversity of benthic diatom flora in the coastal zone of the Gulf of Gdańsk: A case study of transect Gdynia-Sopot. *Oceanol. Hydrobiol. Stud.* **2022**. [CrossRef]
80. Jankowska, H.; Łęczyński, L. *Zatoka Pucka*; Korzeniewski, K., Ed.; Fundacja Rozwoju UG: Gdańsk, Poland, 1993; pp. 320–327. (In Polish)
81. Damaszke, M.; Szymczak, E. Transport of the river load from the Błądzikowski stream to the puck lagoon (southern Baltic Sea, Poland). *Oceanol. Hydrobiol. Stud.* **2013**, *42*, 216–224. [CrossRef]
82. Huzarska, K. Spatial distribution of biological and physical sediment parameters in the western Gulf of Gdańsk. *Oceanologia* **2013**, *55*, 453–470. [CrossRef]
83. Saniewska, D.; Gębka, K.; Bełdowska, M.; Siedlewicz, G.; Bełdowski, J. Impact of hydrotechnical works on outflow of mercury from the riparian zone to a river and input to the sea. *Mar. Pollut. Bull.* **2019**, *142*, 361–376. [CrossRef]
84. Latała, A. The chlorophyll ''a" contents in the surface waters of Gdańsk Bay. Studia I Mater. *Oceanol. Stud. KBM PAN* **1993**, *64*, 187–195.
85. Szarafin, T.; Karwik, A.; Uścinowicz, S.; Fac-Beneda, J.; Nowacki, J.; Boniecka, H.; Gajda, A.; Gawlik, W.; Meissner, W.; Bzoma, S. *Zbiorcze Sprawozdanie z Analizy Dostępnych Danych i Przeprowadzonych Inwentaryzacji Przyrodniczych (Zebranie i Analiza Wyników Inwentaryzacji, Materiałów Niepublikowanych i Opracowań Publikowanych, Przydatnych do Sporządzenia Projektów Planów)*; Zatoka Pucka (PLB 220005); Wydawnictwa Wewnętrzne Instytutu Morskiego w Gdańsku Nr 6757. 2013; pp. 188–244. (In Polish)
86. Szymczak, E.; Szmytkiewicz, A. Współczesne procesy sedymentacyjne w Zatoce Puckiej. In *Zatoka Pucka*; Bolałek, J., Burska, D., Eds.; Wydawnictwo Uniwersytetu Gdańskiego: Gdańsk, Poland, 2022; Tom I; pp. 255–264. (In Polish)
87. Jędruch, A.; Kwasigroch, U.; Bełdowska, M.; Kuliński, K. Mercury in suspended matter of the Gulf of Gdańsk: Origin, distribution and transport at the land–sea interface. *Mar. Pollut. Bull.* **2017**, *118*, 354–367. [CrossRef]
88. Aksentov, K.I.; Sattarova, V.V. Mercury geochemistry of deep-sea sediment cores from the Kuril area, northwest Pacific. *Prog. Oceanogr.* **2019**, *180*, 102235. [CrossRef]

89. Witkowski, A. Mikrofitobentos. In *Zatoka Pucka*; Korzeniewski, K., Ed.; Fundacja Rozwoju Uniwersytetu Gdańskiego: Gdańsk, Poland, 1993; pp. 395–415. (In Polish)
90. Prokopowicz, A. Badania nad Metodą Oznaczania rtęci w Wybranych Elementach Środowiska. Ph.D. Thesis, Uniwersytet Śląski, Katowice, Poland, 2007.
91. Saniewska, D.; Bełdowska, M.; Szymczak, E.; Kuliński, K.; Bełdowski, J.; Voss, M.; Pryputniewicz-Flis, D.; Burska, D. Processes affecting the transformation of mercury in the coastal zone in the vicinity of two river mouths in the southern Baltic Sea. *Mar. Chem.* **2022**, *238*, 104065. [CrossRef]
92. Mason, R.P.; Reinfelder, J.R.; Morel, F.M.M. Bioaccumulation of mercury and methylmercury. *Water Air Soil Pollut.* **1995**, *80*, 915–921. [CrossRef]
93. Mohan, B.S.; Hosetti, B.B. Aquatic plants for toxicity assessment. *Environ. Res.* **1999**, *81*, 259–274. [CrossRef] [PubMed]
94. Smrchek, J.C.; Zeeman, M. Assessing risks to ecological systems from chemicals. In *Handbook for Environmental Risk Assessment and Management*; Calow, P., Ed.; Blackwell Science Ltd.: London, UK, 1998; Chapter 3; pp. 24–90.
95. Graca, B.; Łukawska-Matuszewska, K.; Burska, D.; Łęczyński, L.; Bolałek, J. Geochemical changes in aquatic environment caused by deep dredging—A case study: The Puck Bay (Baltic Sea). In *Oceanography*; Marcelli, M., Ed.; InTechOpen: London, UK, 2012; pp. 259–280. [CrossRef]
96. Szymczycha, B.; Kłostowska, Ż.; Lengier, M.; Dzierzbicka-Głowacka, L. Significance of nutrient fluxes via submarine groundwater discharge in the Bay of Puck, southern Baltic Sea. *Oceanologia* **2020**, *62*, 117–125. [CrossRef]
97. Hetko, D.; Pędziński, J.; Witak, M. Response of the Diatom Flora of the Hel Peninsula Vicinity, Puck Bay, Baltic Sea, to Anthropopressure. *IOP Conf. Ser. Earth Environ. Sci.* **2019**, *362*, 012050. [CrossRef]