

# Fluxes and balance of mercury in the inner Bay of Puck, southern Baltic, Poland: an overview

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## Abstract

The aim of the study was to analyse the balance of mercury (Hg), i.e. the content of this metal, its inflow and outflow, in the ecosystem of the Bay of Puck. Based on literature data and the results of the author's own study, this analysis has shown that the main source of Hg pollution is the atmosphere. An estimated 1.1–3.8 kg of Hg enters annually from the atmosphere, whereas the mass of Hg carried there by river waters per annum is about 7 times lower (0.13–0.44 kg year<sup>-1</sup>). The 0.9–2.7 kg year<sup>-1</sup> of Hg released from Bay of Puck waters to the atmosphere is of the same order as the quantity deposited from the atmosphere. The total amount of Hg deposited in the upper (0–5 cm deep) layer of the sediments has been estimated at 240–320 kg, its rate of entry being c. 2.25–2.81 kg year<sup>-1</sup>. 0.25–1.25 kg year<sup>-1</sup> of Hg are released from the bottom sediments to bulk water, while 0.61–0.97 kg remains confined in aquatic organisms, including 133 g in the phytobenthos, 2.6 g in the zooplankton, 420–781 g in the macrozoobenthos and 34 g in fish.

## 1. Introduction

The considerable attention being focused on environmental pollution by mercury (Hg) is a consequence of the high toxicity and stability of its chemical species (mainly methylmercury and the vapours of elemental

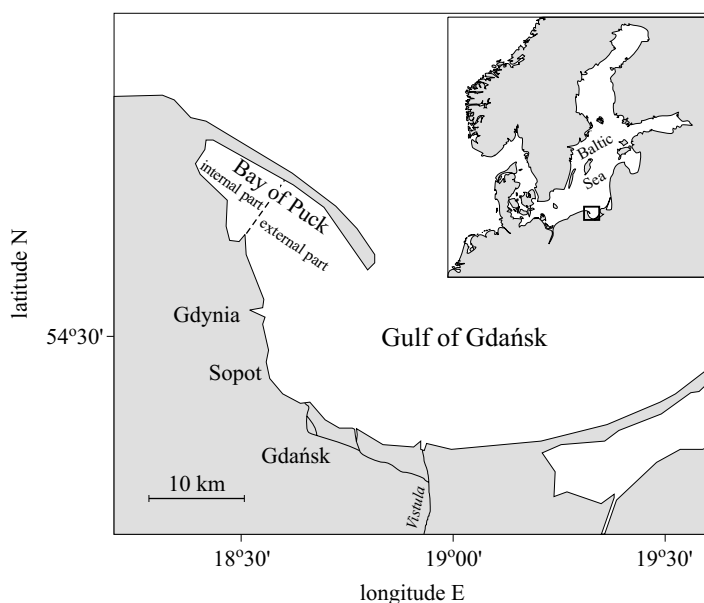
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Hg), its prevalence resulting from the large number of its sources and the ease of its dispersal (Lindqvist & Rodhe 1985, Stein et al. 1996, Morel et al. 1998). Moreover, in view of the fact that bioaccumulation of Hg occurs, especially in fish (there is evidence for the lethal poisoning of fish by Hg), the migration of this element in the natural environment has been studied for many years. Areas most at risk to Hg pollution include bays, especially those in the vicinity of large conurbations, and the estuaries of large rivers flowing through heavily industrialised areas (Nakanishi et al. 1989, Bryan & Langston 1992, Ojaveer 1995, Locarnini & Presley 1996). The problem of the Hg pollution of the several biotic and abiotic components of the Bay of Puck ecosystem has been relatively well researched (Falandysz et al. 1993, Falandysz 1994, Boszke & Falandysz 1999, Falandysz & Boszke 1999, Boszke et al. 2003a). Nevertheless, the problem of the mass balance of Hg in this ecosystem, including the identification of the sources of this pollutant, the modes of its transportation, and hence, the estimated risk to the Bay from such pollution, are still very poorly understood.

A detailed mass balance of Hg in any aquatic ecosystem requires a large database containing the results of determinations of this metal in practically all the abiotic and biotic matrices both in the past and at present. For lack of data on inflow from the atmosphere, the exchange of Hg with natural watercourses and waste-water outfalls, surface inflow, and exchange with the Gulf of Gdańsk, it is not yet possible to draw up such a detailed mass balance with respect to the Bay of Puck. Nevertheless, on the basis of such information as is available, it has been possible to draw up an approximate balance and arrive at some conclusions regarding the mass balance of Hg in the Bay of Puck.

## 2. Characterisation of the study area

The Bay of Puck is a sub-region of the Gulf of Gdańsk (Fig. 1) and can be divided into two parts: the eastern, outer part and the western, inner one. They are separated by the Rybitwia Mielizna, the submerged part of an 8.6 km-long sandbank originating at the Rewski Cypel, a sand spit whose length depends on the water level (Cyberski & Nowacki 1993). The inner part of the Bay is known as the Zalew Pucki – it is 103 km<sup>2</sup> in area and contains the small volume of 0.32 km<sup>3</sup> of water. Shallow, with a mean depth of 3.13 m, the Zalew Pucki has three ‘deeps’: Jama Kuźnicka (9.40 m), Jama Chałupska (4.0 m) and Jama Rzucewska (5.7 m) (Nowacki 1993a, Andrulowicz & Janta 1997). Water exchange between the two parts of the Bay of Puck is impeded, taking place only through the artificial underwater channels known as Głębinka and Przejście Kuźnickie, and, when water levels are sufficiently high, across the Rybitwia Mielizna (Nowacki



**Fig. 1.** A map of the study area

1993b). The hydrological situation in the Bay of Puck is also affected by the inflow of water from the surrounding catchment area (908.8 km<sup>2</sup>), the most important watercourses being the rivers Reda, Gizdepka, Płutnica, Zagórska Struga and Chylonka. Locally, even small watercourses can play a very important role, especially in the coastal zone (Cyberski & Nowacki 1993). Very strong winds are also important in shaping the hydrological conditions and atmospheric transport in the inner Bay of Puck, since the main sources of Hg pollution – the local industrial complexes (Gdańsk–Sopot–Gdynia) and the estuary of the river Vistula (Wisła) – lie on the eastern side of the Bay (Nowacki 1993b).

The conditions in the inner part of the Bay of Puck are very favourable to the development of life on the bottom and in the water itself as this is fairly shallow and has relatively little contact with the open sea waters. The biodiversity of the Bay of Puck is thus the greatest of all the waters in the Polish coastal zone. In the mid-1970s, the uncontrolled discharge of untreated sewage into the Bay from the surrounding towns and villages led to a breakdown in the environmental equilibrium. Large amounts of nutrients stimulated the growth of *Ectocarpaceae* algae, which are capable of producing toxins. As a result, the underwater meadows that used to cover almost the entire bottom became degraded, and now only a few remnants of these meadows, with a much poorer and altered species composition are left. The phytophilous fauna has been similarly impoverished. Despite

these changes, the species diversity is still relatively high, with about 25 species of macroalgae and 8 species of vascular plants. The Bay of Puck thus remains the ecosystem with the greatest biodiversity on the Polish Baltic coast (Andrulewicz & Janta 1997).

In 1978 the inner Bay of Puck became part of the Coastal Landscape Park, and as such, the first marine area in Poland subject to legal protection. In 1992 it was added to the list of Baltic Sea Protected Areas (BSPA) established under the auspices of the Helsinki Committee, and subsequently placed on the list of the World-Wide Fund for Nature (WWF).

### 3. Abiotic components

#### 3.1. Atmosphere

Different species of mercury possess different physico-chemical properties, and thus behave differently in biogeochemical cycles. For instance, divalent mercury Hg(II) is relatively soluble in water and can be removed from the atmosphere by wet and dry deposition near its source; elemental Hg, on the other hand, can be transported over large distances because of its high vapour pressure and poor solubility in water. It is possible to eliminate elemental Hg from the atmosphere by means of wet or dry deposition, but only after it has been oxidised to the readily water-soluble Hg(II) (Lindqvist & Rodhe 1985, Stein et al. 1996, Morel et al. 1998, Boszke & Siepak 2002).

Air pollution with Hg in the Bay of Puck area has not been comprehensively studied: the only data available is in Marks & Beldowska (2001). According to this work, the Hg concentration in the air above the Bay of Puck in spring varied from 0.4 to 2.6 ng m<sup>-3</sup>, and in summer from 1.1 to 4.8 ng m<sup>-3</sup>. Moreover, the same authors found the Hg concentration in the air in Gdańsk, Sopot and Gdynia in spring to be 3–4 ng m<sup>-3</sup>, and in the vicinity of the ‘Dębogórze’ power station in Gdynia to be 5 ng m<sup>-3</sup>. According to Wängberg et al. (2001), the mean Hg concentration in the air over the southern part of the Baltic Sea was 1.70 ng m<sup>-3</sup> in summer and 1.39 ng m<sup>-3</sup> in winter. These authors also calculated the mean concentration of dissolved gaseous mercury (DGM) in water samples from the southern part of the Baltic Sea to be 17.6 ng m<sup>-3</sup> in summer and 17.4 ng m<sup>-3</sup> in winter.

In order to estimate the Hg influx into the inner part of the Bay of Puck ecosystem, it was assumed that the Hg inflow to the Bay with wet precipitation (rain, snow) was the same as in the southern Baltic, i.e. 26–72 ng m<sup>-2</sup> day<sup>-1</sup> (Schmolke et al. 1997). A further assumption was that Hg deposition with dry precipitation was the same for the Bay as for the Baltic Sea, hence on average, 10% of the total deposition (Petersen

et al. 1995). Thus, the Hg inflow with wet and dry precipitation to the inner Bay of Puck was estimated at  $29\text{--}80\text{ ng m}^{-2}\text{ day}^{-1}$ , which gives us from  $1.1$  to  $3.0\text{ kg year}^{-1}$ . According to Ebinghaus et al. (1995), the mean Hg concentration in the wet precipitation over the southern Baltic was  $58\text{ ng dm}^{-3}$  ( $20\text{--}110\text{ ng dm}^{-3}$ ). Assuming the annual wet precipitation to be  $59.2$  million  $\text{m}^3$  (Cyberski & Nowacki 1993) and that dry deposition makes up 10% of the total deposition, the mean Hg inflow from the atmosphere with wet and dry precipitation is  $100\text{ ng m}^{-2}\text{ day}^{-1}$  ( $35\text{--}190\text{ ng m}^{-2}\text{ day}^{-1}$ ), i.e.  $3.8\text{ kg year}^{-1}$  ( $1.3\text{--}7.2\text{ kg year}^{-1}$ ). For southern Scandinavia, Schmolke et al. (1997) estimated the inflow of Hg with wet precipitation at  $13\text{--}41\text{ ng m}^{-2}\text{ day}^{-1}$  (wet and dry –  $15\text{--}46\text{ ng m}^{-2}\text{ day}^{-1}$ ).

Table 1 presents the deposition of mercury from the atmosphere in different parts of the world: the values vary from  $2.7$  to  $1300 \pm 200\text{ ng m}^{-2}$  per day. In Scandinavia, the geographical distribution of the Hg inflow is a derivative of the annual fluctuations in the atmospheric precipitation and the regional differences in Hg concentrations in the local precipitation (Iverfeldt 1991). In northern Scandinavia Hg deposition from the atmosphere was estimated at  $8.2\text{ ng m}^{-2}\text{ day}^{-1}$ , in the south at up to  $96\text{ ng m}^{-2}\text{ day}^{-1}$ . The explanation for the high atmospheric deposition of Hg over southern Scandinavia was that the metal had been transported there from over eastern Europe (Iverfeldt 1991). Regional differences in Hg inflow from the atmosphere have been reported not only from Europe: in northern North America – Canada and the northern states of the USA – the inflow of mercury from the atmosphere was much lower than in the southern states of the USA (Table 1). In general, Hg inflow from the atmosphere can be 2–3 times greater over an industrialised area than over a rural area (Mason et al. 2000). Moreover, the contribution of dry deposition from the atmosphere can be greater in deposition over land (Schroeder & Munthe 1998). For example, over the Lake Gårdsjön catchment area in Sweden, dry deposition made up as much as 50% of the total atmospheric deposition of mercury, a total of  $16.4\text{ ng m}^{-2}\text{ day}^{-1}$  (Driscoll et al. 1994).

Apart from being deposited on land and water bodies from the atmosphere, mercury is also released from the land (vegetation, soil) and from water bodies (seas, lakes, rivers) into the atmosphere; this is a natural consequence of the volatility of elemental Hg and some of its species (Mason et al. 1994, Lindberg et al. 1998, Lindberg & Zhang 2000, Gårdfeldt et al. 2001). It is suspected that Hg(II) can be reduced by humus matter in the presence of solar radiation (or perhaps in the absence of such irradiation) but accurate mechanisms of the conversions have not yet been established

**Table 1.** Atmospheric deposition of mercury in various parts of the world

	Deposition [ng m <sup>-2</sup> day <sup>-1</sup> ]	Reference
<b>Europe</b>		
Scandinavia	8.2–96	Iverfeldt 1991
Scandinavia	27	Iverfeldt et al. 1995
Southern Sweden	336	Xiao et al. 1998
Baltic Sea	13–41	Schmolke et al. 1997
Baltic Sea	38–85	Petersen 1992
Baltic Sea	2.7–164	Petersen et al. 1995
Finland (polluted region)	1300 ± 200	Lodenius 1998
Svartberget Catchment (Sweden)	19	Lee et al. 2000
Coniferous catchment (Germany)	148	Schwesig & Matzner 2000
Deciduous catchment (Germany)	167	Schwesig & Matzner 2000
Lake Gårdsjön (Sweden)	33	Driscoll et al. 1994
Small boreal catchments (Finland)	12.9–17.0	Porvari 2003
<b>North America</b>		
Chesapeake Bay (USA)	41–77	Mason et al. 1997
Michigan (USA)	32–65	Hoyer et al. 1995
Florida (USA)	71	Guentzel et al. 1998
Florida (USA)	33–85	Guentzel et al. 1995
Lake Superior (USA)	27	Hoff et al. 1996
Wisconsin (USA)	19	Lamborg et al. 1995
St-Aicet, Quebec (Canada)	21	Poissant & Pilote 1998
Lake Onondaga (USA)	55–82	Bigham & Vandal 1996
Forested catchment in Vermont (USA)	116–127	Scherbatoskoy et al. 1998
Lake Champlain Watershed (USA)	21.6	Rea et al. 1996
Lake Champlain Watershed (USA)	24.6 ± 1.6	Rea et al. 2002
Lake Huron Watershed (USA)	23.8 ± 1.4	Rea et al. 2002
Blacklick Run Watershed – Maryland (USA)	40 ± 34	Lawson & Mason 2001

(Lidqvist & Rodhe 1985, Xiao et al. 1991, Weber 1993, Lindberg et al. 1995, Amyot et al. 1997, Costa & Liss 1999, 2000, Beauchamp et al. 2000, Lindberg & Zhang 2000, Liu et al. 2000).

The estimated outflow of Hg from the water to the atmosphere in the southern Baltic is 26–72 ng m<sup>-2</sup> day<sup>-1</sup>, a value comparable to the wet deposition of Hg from the atmosphere (Schmolke et al. 1997, Wängberg et al. 2001). Assuming the same values for the Bay of Puck, 1.8 kg year<sup>-1</sup> (0.9–2.7 kg year<sup>-1</sup>) of Hg are released from the water into the atmosphere from the inner part of the Bay. If c. 96 ng Hg<sup>0</sup> per 1 gram of primary production is released from the water into the atmosphere (Mason et al. 1994), then the average amount of Hg released from the Bay of Puck at a primary production rate of 0.54 g C m<sup>-2</sup> day<sup>-1</sup> (Renk 1993) is 2.0 kg year<sup>-1</sup>, which is consistent with the value calculated earlier. For comparison, an estimated 23–45 ng m<sup>-2</sup> day<sup>-1</sup> of Hg are released from the water into the atmosphere in the North Sea (Coquery & Cossa 1995); in other parts of the world this value varies from 8 to 140 ng m<sup>-2</sup> day<sup>-1</sup> (Table 2). Mason et al. (1994) estimated that the release of Hg from the water to the atmosphere in the Arctic can be 5 times lower than its inflow from the atmosphere to the water, whereas in the equatorial zone of the Pacific Hg release into the atmosphere can be 6 times greater than its deposition from the atmosphere. According to Gårdfeldt et al. (2001), Hg release from river water into the atmosphere was on average one order of magnitude greater than its release from the sea, which was explained by the higher temperature of the river water, as well as the faster photoreduction processes and the higher organic matter content in this water. The rate of Hg release into the atmosphere also depends on the time of day: in summer it reaches a maximum during day but is much lower at night

**Table 2.** Outflow of mercury from water to the atmosphere in various parts of the world

	Outflow of mercury from water to the atmosphere [ng m <sup>-2</sup> day <sup>-1</sup> ]	Reference
Baltic Sea	26–72	Schmolke et al. 1997
North Sea	40–60	Coquery & Cossa 1995
Scheldt Estuary (Belgium)	140	Leermakers et al. 1995
Mediterranean Sea	28	Cossa et al. 1997
Laptev Sea (Russia)	10–16	Cossa et al. 1993
Lake Superior (USA)	8	Hoff et al. 1996
Narragansett Bay (USA)	20–40	Mason et al. 1993
Gulf of Mexico (USA)	192	Amyot et al. 1997
Various Lakes (Canada)	14.4–120	Beauchamp et al. 2000

(Xiao et al. 1991, Lindberg et al. 1995, Lindberg & Zhang 2000, Gårdfeldt et al. 2001).

### 3.2. Water

A perusal of the literature data on Hg levels in natural water reveals that these concentrations extend over a wide range of values, very probably as a consequence of the natural variation in the composition of Hg species and their concentrations in water, not to mention the discrepancies arising out of the application of different analytical techniques (Stein et al. 1996, Boszke et al. 2002). It has been estimated that the natural total Hg concentration in sea and ocean water varies from 0.5 to 3.0 ng dm<sup>-3</sup>, and in estuaries and in rain water from 2.0 to 15 ng dm<sup>-3</sup> (Stein et al. 1996). The available literature supplies no data on the Hg content in Bay of Puck water. The 1987 report of the Baltic Marine Environment Protection Commission (the Helsinki Committee) states that the Hg concentration in Gulf of Gdańsk water is 227–630 ng dm<sup>-3</sup>, a much higher value than in any other region of the Baltic Sea (BMEPC 1987). The most recent data (Pempkowiak et al. 1998) puts the Hg concentration in filtered Gulf of Gdańsk water at 0.28–0.94 ng dm<sup>-3</sup>, and in unfiltered water taken from between the Pomeranian Bay and Bornholm at 0.44–1.1 ng dm<sup>-3</sup>. According to Wurl et al. (2001), the total Hg concentration in Baltic Sea water ranges from 0.2 to 1.87 ng dm<sup>-3</sup>.

In North Sea water the proportion of Hg bound to suspended matter has been estimated at 13–82% of the total Hg concentration in unfiltered water (Coquery & Cossa 1995), whereas in the early 1980s this proportion in the southern Baltic was calculated at 28.4% (Brzezińska et al. 1984). In Bay of Puck water the total Hg concentration varies from 0.39 to 1.3 ng dm<sup>-3</sup>: if we assume that the concentration of this metal in unfiltered water is roughly the same as in Gulf of Gdańsk water, the proportion of Hg bound to suspended matter is thus 28.4% of the total mercury. Given that the volume of water in the inner part of Bay of Puck is 0.32 km<sup>3</sup> (Nowacki 1993a), we obtain a total amount of Hg in this water of 125–416 g.

The concentration of mercury in Lake Chesapeake catchment water (USA) is 3–4 ng dm<sup>-3</sup>, including 1.6 ng dm<sup>-3</sup> in the dissolved phase (Mason et al. 1997), so 55% of the total Hg concentration is bound to suspended matter. In forests and marshland, the proportion of Hg bound to suspended matter was 20–30%, but in industrialised areas this proportion reached 52–80% (Hurley et al. 1998, Kolka et al. 1999, 2001). This value of 55% of the total Hg bound to suspended matter was used to work out the Hg concentration in the water of the rivers entering the Bay of Puck, with the further assumption that the Hg concentration in the dissolved phase was



**Table 3.** Outflow of mercury from river catchment areas in various parts of the world

	Catchment area (1000x m <sup>2</sup> )	Outflow with river water [ng m <sup>-2</sup> day <sup>-1</sup> ]	Reference
<b>Europe</b>			
Lake Iso catchment (Finland)	39	4.1–4.7	Verta et al. 1994
15 various catchments (Sweden)		2.2–16	Johansson et al. 1991
Lake Gårdsjön catchment (Sweden)	37	6.3–9.6	Hultberg et al. 1994
Lake Gårdsjön catchment (Sweden)	37	8.2	Driscoll et al. 1994
Tiveden National Park (Sweden)	14	9.3	Aastrup et al. 1991
Catchment in South Sweden	50	4.2	Pettersson et al. 1995
Svartberget catchment (Sweden)		2.7–9.3	Lee et al. 2000
Small boreal catchments (Finland)	71	5.5–14.5	Porvari et al. 2003
Boreal forest catchments (Finland)	2–1300	2.5–4.9	Porvari & Verta 2003
– upland		2.5–3.0	Porvari 2003
– upland/peatland		4.4	Porvari 2003
– peatland		2.74–4.93	Porvari 2003
<b>North America</b>			
Chesapeake Bay catchment (USA)	$16 \times 10^7$	2.6	Mason et al. 1997
Rivers in Wisconsin (USA)		5–14	Hurley et al. 1995
Lake Onondaga catchment (USA)	$6.9 \times 10^5$	14–66	Henry et al. 1995
Wetland in New York region (USA)		6.0	Driscoll et al. 1998
Forested catchment in Vermont (USA)	110	6.0–8.8	Scherbatosky et al. 1998
Forested catchments (USA)		1.9–7.7	Kolka et al. 1999
Minnesota (USA)	100	$7.7 \pm 0.8$	Grigal et al. 2000
Watersheds in Wisconsin (USA)		< 107	Babiarz et al. 1998

the same as for the Gulf of Gdańsk. The Hg concentration in unfiltered water from the rivers flowing into the Bay of Puck was estimated at 0.62–2.1 ng dm<sup>-3</sup>. Assuming after Nowacki (1993a) that the amount of river water flowing into the Bay of Puck is 212.1 million m<sup>3</sup> per year, the Hg load carried by these waters can be put at 132–445 g.

The catchment area of the inner Bay of Puck covers 693.5 km<sup>2</sup> (Cyberski & Nowacki 1993), so the amount of mercury brought into the Bay from that area is 0.52–1.8 ng m<sup>-2</sup> day<sup>-1</sup>. Against the background of the world data on Hg carried with catchment water (Table 3), the value estimated for the Bay of Puck lies among the lower group of values for regions not polluted with this element. The estimated outflow of Hg with river water varies from 2.2 to 107 ng m<sup>-2</sup> day<sup>-1</sup> (Table 3). Higher outflows have been obtained for industrialised regions (14–107 ng m<sup>-2</sup> day<sup>-1</sup>) than for rural areas (2.2–6.8 ng m<sup>-2</sup> day<sup>-1</sup>). In industrialised regions, Hg deposition from the atmosphere and the amount of suspended particulate matter in the water are both much greater (Bigham & Vandal 1996, Mason et al. 1997, 2000, Hurley et al. 1998, Mason & Sullivan 1998, Neumann et al. 2000).

Assuming that Hg inflow from the atmosphere over the catchment area is the same as over the Bay of Puck, i.e. 29–80 ng m<sup>-2</sup> day<sup>-1</sup>, it is estimated that only a small quantity (1.8–6.0%) of the Hg deposited on to the catchment area from the atmosphere actually reaches Bay of Puck waters. A value of the same order (< 10%) was given by Mason et al. (1997) for Chesapeake Bay (USA). The high sorption capacity of forest soils means that 92–95% of Hg deposited from the atmosphere on to the catchment area can be retained by the soil. Only during intense rainfall do occasional instances of strong elemental Hg release occur, with the resulting water flows carrying up to half the annual load of elemental Hg derived from the soil (Bonzongo et al. 1996, Scherbatskoy et al. 1998). A model study has shown that an increase in wet precipitation by 35% relative to the normal annual standard can bring about a 2 to 3-fold increase in Hg release from the soil, while a drop in wet precipitation by 18% can reduce Hg release by 5–7 times (Tsiros 2001).

### 3.3. Sediments

A good indicator of the pollution of water by mercury is its content in the sediments, where it can undergo a number of conversions. On the one hand, Hg accumulates there as a result of the deposition of suspended matter, on the other, it is released from the sediments to become available for further biogeochemical transformation. The intensity of these two processes depends on the environmental conditions in the given water system, e.g. pH,

redox conditions, concentrations of sulphides and organic matter, activity of micro-organisms (Stein et al. 1996, Boszke et al. 2003b).

The amount of mercury deposited in the upper sediment layer (0–5 cm deep) over the bottom of the inner Bay of Puck has been estimated. A strong correlation was found between the total Hg concentration and the organic matter content (as loss on ignition at 550°C) in the surface layer of sediments in the Bay of Puck; the correlation coefficient of  $r = 0.93$  was positive for all samples (Boszke & Falandysz 1999). The amount of Hg accumulating in the surface layer of the sediments in the Bay was estimated on the basis of data on the Hg concentration in sediments (Boszke & Falandysz 1999), and data on the organic matter content in sediments (Gołębiowski & Musielak 1978, Jankowska & Łęczyński 1993). To begin with, the areas of the sea bed characterised by the same organic matter content were estimated. The ranges of organic matter content taken into consideration were < 1%, 1–5%, 5–10% and 15–20% (variant I – Jankowska & Łęczyński 1993), and 0–5%, 5–10%, 10–15%, 15–20% and > 20% (variant II – Gołębiowski & Musielak 1978). The total area of the bottom of the inner Bay of Puck was taken to be 103 km<sup>2</sup> (Nowacki 1993a), and the weighted mean values of the Hg concentration in the areas of the same organic matter content were calculated. Then, on the basis of the densities of sediments in the Bay of Puck, which range from 2.45 g cm<sup>-3</sup> for organic-rich sediments to 2.65 g cm<sup>-3</sup> for sandy sediments poor in organic matter (Bolałek 1993), the mass of the deposited Hg was calculated from the equation:

$$M_{\text{Hg}} = \sum C_n V_n \delta_n, \quad (1)$$

where

$M_{\text{Hg}}$  – mass of total Hg in the Bay of Puck sediments [ng],

$C_n$  – mean total Hg concentration in the sediment with a given content of organic matter [ng g<sup>-1</sup>],

$V_n$  – volume filled by the surface (0–5 cm deep) layer of the sediment with a given content of organic matter [cm<sup>3</sup>],

$\delta_n$  – density of the sediment with a given content of organic matter [g cm<sup>-3</sup>],

$n$  – number of areas with the same organic matter content in the surface layer of the sediments.

Depending on the organic matter content, the mass of Hg deposited in the surface layer of the sediments in the inner Bay of Puck was estimated at 240 kg for variant I and 320 kg for variant II (Table 4).

On the basis of the spatial distribution of Hg concentration in the Bay of Puck sediments and the available data on the rate of sedimentation in the Bay, an attempt was made to evaluate the annual influx of mercury

**Table 4.** Mass of mercury deposited in the bottom sediments of the Bay of Puck

	Layer [cm]	Mass of mercury [kg] estimated from the data on the organic matter content in the sediments	
		Jankowska & Łęczyński 1993	Gołębiewski & Musielak 1978
Mean mass of Hg in the bottom sediments of the inner Bay of Puck	0–5	240	320
Mean mass of Hg in bottom sediments with an organic matter content < 5%	0–5	68	95
Percentage of the mass of Hg deposited in the bottom sediments with an organic matter content < 5% in the total mass of Hg contained in the bottom sediments of the entire Bay of Puck	0–5	28%	30%
Mean mass of Hg in the core layers of bottom sediments from the regions characterised by an organic matter content in the surface layer of < 5%	0–5 5–10 10–15	86 103 88	45 49 50

into the Bay. According to Pempkowiak & Ciszewski (1990), the rate of sedimentation was assumed to be  $0.45 \pm 0.07$  mm year<sup>-1</sup>; the minimum rate of 0.38 mm year<sup>-1</sup> was recorded in sandy deposits with an organic matter content < 5%, the maximum of 0.52 mm year<sup>-1</sup> in areas with an organic matter content > 10%. Based on the amounts of Hg accumulating in the upper layer of the sediments, the influx of this metal to the Bay of Puck in the last 132, 111 and 96 years was estimated at 2.25 kg year<sup>-1</sup> (60 ng m<sup>-2</sup> day<sup>-1</sup>) and 2.81 kg year<sup>-1</sup> (75 ng m<sup>-2</sup> day<sup>-1</sup>), assuming the first and the second rates of sedimentation, respectively. The annual influx of Hg depends on the type of sediment. For sediments with an organic matter content < 5%, 5–10% and > 10%, it reaches 44, 117 and 323 ng m<sup>-2</sup> day<sup>-1</sup> for the first rate of sedimentation or 36, 123 and 244 ng m<sup>-2</sup> day<sup>-1</sup> for the second one.

Leivuori & Niemistö (1995) estimated the Hg influx to the sediments in the Gulf of Bothnia and Bothnian Sea at 90 and 88 ng m<sup>-2</sup> day<sup>-1</sup>, whereas according to Borg & Jonsson (1996) the respective figures were 112 and 16 ng m<sup>-2</sup> day<sup>-1</sup>. The latter authors also estimated the Hg influx to the sediments of the Gulf of Finland, Gulf of Riga and the central Baltic Sea at

112, 47 and 41  $\text{ng m}^{-2} \text{day}^{-1}$ , and the average value for the whole Baltic at 44  $\text{ng m}^{-2} \text{day}^{-1}$ . This value is in agreement with the estimated amount of Hg deposited in the Baltic from the atmosphere – 6–13 tonnes year<sup>-1</sup> (38–85  $\text{ng m}^{-2} \text{day}^{-1}$ ) (Petersen 1992). The Hg influx to the sediments in the seas of the world is compared in Table 5.

**Table 5.** Influx of mercury into bottom sediments in various parts of the world

	Influx of mercury to bottom sediments [ $\text{ng m}^{-2} \text{day}^{-1}$ ]	Reference
<b>Europe</b>		
Baltic Sea	44	Borg & Jonsson 1996
Baltic Proper (central part of the Baltic Sea)	41	Borg & Jonsson 1996
Bothnian Bay	112	Borg & Jonsson 1996
Bothnian Bay	90	Leivuori & Niemistö 1995
Bothnian Sea	16	Borg & Jonsson 1996
Bothnian Sea	88	Leivuori & Niemistö 1995
Gulf of Finland	112	Borg & Jonsson 1996
Gulf of Riga	47	Borg & Jonsson 1996
Central Norway	28	Hermanson 1998
<b>North America</b>		
Lakes in Wisconsin (USA)	126–305	Hurley et al. 1994
Sequenay Fjord (Canada)	537	Gagnon et al. 1997
Lake Imitavik (Canada)		Hermanson 1998
– 1980s	34	
– 1990s	49	

Along with the deposition of Hg in the sediments, the reverse process of its release from the sediments to the bulk water takes place (Luoma & Davis 1983, Rutgers van der Loeff et al. 1984, Gobeil & Cossa 1993, Gagnon et al. 1997, Boszke et al. 2003b). For instance, mercury sulphide is strongly bound to sediments, but some of it can dissolve as a result of processes involving bacteria or as a result of oxidising conditions appearing as a result of e.g. bioturbation. For the above reasons, many authors question the usefulness of investigating sediment cores for the estimation of past mercury influxes. Nevertheless, the Hg content in dated sediments do show good agreement with historical data on Hg released from point anthropogenic sources (Lockhart et al. 1998).

According to Gobeil & Cossa (1993) the amount of mercury released from the deeper layers of the sediment to its surface layer by means of molecular diffusion makes up 3% of the amount of Hg deposited from the bulk water to the sediment; according to Gagnon et al. (1997) this value is about 7%. Rutgers van der Loeff et al. (1984) have shown that the amount of Hg redeposited from the sediment to the bulk water as a result of the activity of benthic organisms was 2 to 10 times greater than the amount redeposited from the sediment to the bulk water as a result of molecular diffusion. Assuming that in the Bay of Puck 5% of the Hg deposited in the bottom sediment is released during molecular diffusion and the magnitude of bioturbation is as indicated by Rutgers van der Loeff et al. (1984), the amount of Hg released from the sediment varies from 0.25 to 1.25 kg year<sup>-1</sup>, i.e. from 6.6 to 33 ng m<sup>-2</sup> day<sup>-1</sup>.

## 4. Biotic components

### 4.1. Zooplankton

As there are no data on the magnitude of the zooplankton biomass in the Bay of Puck ecosystem it was assumed to be the same as the Baltic Sea average, that is 34 mg dry weight per m<sup>3</sup> of bulk water (Brügmann & Hennings 1994). The volume of the water in the inner Bay of Puck was assumed to be 0.32 km<sup>3</sup> (Nowacki 1993a), while the mean concentration of mercury in zooplankton was 240 ng g<sup>-1</sup> dry weight (Boszke et al. 2003a). The amount of mercury bound to zooplankton in the Bay of Puck thus came to 2.6 g.

### 4.2. Macrophytobenthos

The phytobenthos concentration in the inner Bay of Puck in the summer of 1987 varied from 1.6 to 53.9 g m<sup>-2</sup>, on average 27.3 g m<sup>-2</sup> (Kruk-Dowgiałło 1994). The respective proportions of *Pilayella littoralis*, *Zanichella palustris* and the other species to the total phytobenthos biomass were ~75%, ~19% and ~4%, and the respective mean Hg concentrations in these species were 51, 41 and 36 ng g<sup>-1</sup> dry weight (Boszke et al. 2003a). Therefore, the estimated amount of Hg bound to the phytobenthos was 133 g.

### 4.3. Macrozoobenthos

Osowiecki (1994) put the biomass of the macrozoobenthos in the Bay of Puck at 396 g m<sup>-2</sup>, the respective proportions of bivalves, polychaetes, marine gastropods, crustaceans and other species being 82%, 10%, 7%, 1% and 0.23%. According to Wiktor (1993), the mean biomass of the

macrozoobenthos was  $215 \text{ g m}^{-2}$ , and the proportions of bivalves, polychaetes, marine gastropods and crustaceans were 90.1%, 4.5%, 5.2% and 0.1% respectively. In the calculations of the quantity of Hg bound to the zoobenthos it was assumed that the mass of the shell of bivalves and molluscs was 52% and 66% respectively of their body masses (soft tissues and shell) (Wiktor 1993). For the zoobenthos from the inner Bay of Puck, the Hg concentrations in the soft tissues was calculated at 40, 22, 28 and  $35 \text{ ng g}^{-1}$  for bivalves, polychaetes, marine gastropods and crustaceans, respectively (Boszke et al. 2003a). The data for the bivalves from the outer part of the Bay and for snails from the rivers Płutnica and Reda, and for *Lymnea stagnalis* were excluded from the calculations. The amount of Hg bound to the macrozoobenthos in the Bay of Puck was thus estimated at 781 g (for a biomass of  $396 \text{ g m}^{-2}$ ) or 420 g (for a biomass of  $215 \text{ g m}^{-2}$ ). The respective amounts of Hg bound to particular components of macrozoobenthos, i.e. to bivalves, polychaetes, marine gastropods and crustaceans was put at 382–642 g, 22–90 g, 14–34 g and 1–14 g.

#### 4.4. Fish

The fish resources of the Bay of Puck have not yet been reliably estimated. This work makes use of the fragmentary data on the numbers of fish in the Bay of Puck reported by Skóra (1993, 1997). It follows from his study that the three-spined stickleback *Gasterosteus aculeatus* and the nine-spined stickleback *Pungitius pungitius* make up over 95% of the total biomass of fish in the shallow waters of the inner part of the Bay, that is from 7.8 to 18.8 tonnes. However, these data are very inaccurate. According to the same author (Skóra 1993), the catches of commercial fish in the inner Bay of Puck in 1978 and 1988 were officially estimated at 570 and 130 tonnes respectively. Therefore, if we take into account the fact that the eel is the main fish species of commercial importance in the area, that the real size of the catch (of eel and other species) is unknown, and that the species *Neogobius melanostromus* expanded dynamically in the 1990s, the biomass of fish in Bay of Puck can be estimated at around 600 tonnes. The proportions of the three-spined and nine-spined sticklebacks were assumed to be the same in the whole Bay, i.e. 50%, while the mean concentrations of Hg in these fish were 23 and  $44 \text{ ng g}^{-1}$  of wet mass (Falandysz & Boszke 1999, Boszke et al. 2003a). The estimated amount of Hg in three-spined and nine-spined sticklebacks in the Bay of Puck was thus 0.52–1.3 g. The respective biomasses of eel, *N. melanostromus*, and the other species were estimated at 200, 300 and 100 tonnes, and the mean Hg concentration in them at 75, 45 and  $40 \text{ ng g}^{-1}$  wet mass (Falandysz & Boszke 1999,

Boszke et al. 2003a). The total amount of Hg bound up in the biomass of fish was therefore 34 grams.

## 5. A scheme of the mercury balance

As I have already mentioned, there are at present insufficient data to make an accurate and complete balance of mercury in the Bay of Puck ecosystem. Nevertheless, despite the lack of exact data, an attempt can be made at estimating this balance on the basis of the information available for analogous matrices and parameters. On the basis of our own results and the literature data a simple model is proposed to describe the fate of Hg in the inner Bay of Puck. The main assumption of the model is that the data on the influx, deposition and redeposition of Hg established for the Baltic Sea and Central Europe are also valid for the Bay of Puck. The balance of Hg in the Bay can be approximated by the equation:

$$a + b + c + d + e = f + g + h + i, \quad (2)$$

where:

the import of Hg is described by  $a + b + c + d + e$ ,

the export of Hg is described by  $f + g + h + i$ ,

a – inflow from natural watercourses (0.132–0.445 kg),

b – deposition from the atmosphere (1.1–3.8 kg),

c – inflow from waste waters (? kg),

d – inflow from Gulf of Gdańsk water (? kg),

e – redeposition from sediments (0.25–1.25 kg),

f – evaporation (0.9–2.7 kg),

g – deposition to sediments (2.25–2.81 kg),

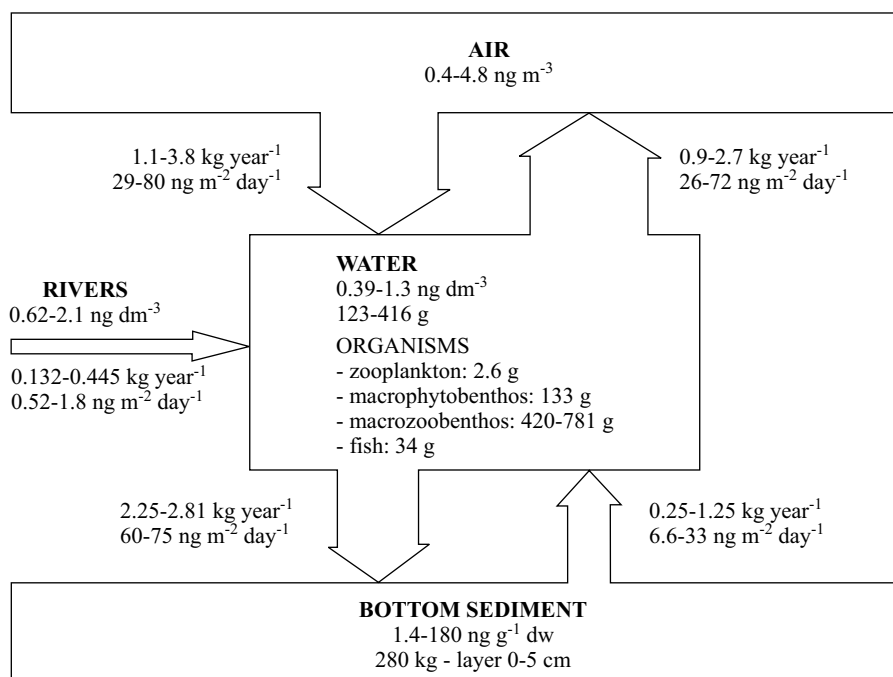
h – outflow from Bay of Puck water (? kg),

i – fishing (? kg).

Assuming that the inflow from Gulf of Gdańsk water is unchanged and that the amount of Hg in the catch of fish and the quantity flowing in from industrial and municipal sewage are relatively small, as indicated by the homogenous distribution of Hg in the sediments after normalisation with respect to the organic matter content, we can state that the main source of Hg in the Bay of Puck water is the atmosphere (1.1–3.8 kg year<sup>-1</sup>). This conclusion is indirectly supported by the spatial distribution of the metal in the sediments (Boszke & Falandysz 1999).

The amount of Hg flowing in from natural watercourses – mainly the rivers Reda, Płutnica and Gizdepka, is about 7 times smaller (0.132–0.445 kg year<sup>-1</sup>) than that entering via the atmosphere (Fig. 2). The fact that the Hg influx from the atmosphere is more important than that from





**Fig. 2.** A scheme of the mercury balance in the Bay of Puck ecosystem

natural watercourses has been generally observed (St. Louis et al. 1996). For Chesapeake Bay (Mason et al. 1997) and Lake Michigan (Rossmann 2000) the atmospheric Hg deposition was estimated at 50% of the total Hg influx from all sources. The proportion of Hg flowing into Lake Onondaga from the catchment area was calculated at 25% of the total influx of this element (Biggam & Vandal 1996). The proportion of Hg flowing into Florida Bay (USA) from the catchment area was put at less than 20% of the total influx of this metal from all sources in areas distant from watercourses, but it increased to 60–80% in the vicinity of estuaries (Kang et al. 2000).

The amount of Hg entering the Bay of Puck water from the atmosphere (1.1–3.8 kg year<sup>-1</sup>) and watercourses (0.132–0.445 kg year<sup>-1</sup>), a total of 1.232–4.245 kg year<sup>-1</sup>, is only 1.8–6.0% of the amount of Hg entering the Bay of Puck's catchment area from the atmosphere. This is mainly because the local soils have a high Hg sorption capacity, an excess of which may have unpredictable consequences. As is the case in the Bay of Puck catchment area, Hg was strongly bound up in the soils in other regions too: about 90% of the total Hg entering from the atmosphere was actually retained in the catchment area (Mason et al. 1997, Allan & Heyes 1998).

An important aspect of the mercury balance in the Bay of Puck ecosystem is the process by which the metal is released from the water to the

atmosphere. The amount of Hg released from the Bay's waters per year is of the same order of magnitude as that deposited from the atmosphere, which indicates that it is an important secondary source of Hg in the atmosphere above the Bay and confirms the significance of the atmosphere's role in the migration and transformation of mercury in the natural environment.

The sediments are the main reservoir of mercury in the water. The estimated mass of Hg deposited in the upper layer of the sediments (0–5 cm) in the Bay is 280 kg, the Hg accumulating at a rate of 2.5 kg per a year. This value is of the same order as that deposited into the Bay from the atmosphere (1.1–3.8 kg year<sup>-1</sup>). The redeposition of Hg from the sediments to water is 0.24–1.24 kg year<sup>-1</sup>; it is this value that determines the bioavailability of the metal. The net amount of accumulated Hg varies from 1.0 to 1.55 kg year<sup>-1</sup> and is of the same order of magnitude as that calculated for aquatic organisms, i.e. 0.613–0.974 kg (excluding the mercury contained in the phytoplankton). The proportions of Hg contained in the macrophytobenthos, zooplankton, macrozoobenthos and fish in the total mass of Hg contained in aquatic organisms were 14–22%, 3–4%, 69–80% and 4–5%, respectively.

The amounts of Hg entering the Baltic Sea from the atmosphere and with river waters were estimated at 20 and 50 tonnes respectively. As much as 93% of the Hg load introduced into the Baltic comes from anthropogenic sources: the most important contributions deriving from natural sources come from the atmosphere – c. 0.1 tonne a year, and from the rivers – c. 4.9 tonnes a year (Matschullat 1997).

## 6. Summary

The waters of the Bay of Puck are moderately polluted with mercury. The fish species tested for their Hg content have proved to be only slightly contaminated with this element: no amount of Hg exceeding the level of tolerance indicated in the Polish Standards for edible parts of fresh fish (300 ng g<sup>-1</sup>) has been reported (Falandysz & Boszke 1999, Boszke et al. 2003a). Estimates of the in- and outflows of Hg in the Bay of Puck ecosystem have shown that the main source of pollution with this metal is the atmosphere. The mass of Hg entering by way of rivers is about 7 times lower than that coming from the atmosphere. Roughly 240–320 kg of Hg is deposited in the upper (0–5 cm deep) layer of the sediments, around 0.13–0.42 kg is confined to the water body, and 0.61–0.97 kg is present in aquatic organisms. Although the current state of mercury pollution in the Bay of Puck is not giving any cause for alarm, monitoring of this metal in the Bay of Puck ecosystem should be continued in view of the extreme toxicity of

some of its species and the fact that the buffer properties of the Bay's water are unknown.

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