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Research Article

# TOTAL GASEOUS MERCURY OVER THE COASTAL ZONE OF THE GULF OF GDAŃSK

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## Abstract

Measurements of total gaseous mercury (TGM) were carried out in the Polish coastal zone of the Gulf of Gdańsk from Hel, situated at the end the tip of the Hel Peninsula, to Piaski in the Vistula Pit. The results were recorded in the atmosphere 150 cm above the ground using an automatic analyzer Gardis 1A in March, April and May 1999. The mercury concentration over the coastal zone of the Gulf of Gdańsk did not exceed maximum permissible levels and were at levels similar to those in other maritime regions of the Baltic Sea and the North Sea. The average concentration of mercury was 3.4 ng $m^{-3}$. Increased TGM concentrations of approximately 8.2 ng $m^{-3}$ occurred locally near the sewage treatment facility in Sztutowo, on the tip of the Hel Peninsula and in the Tri-Cities near the industrial areas of shipyards and the harbor.

No increased concentrations of gaseous mercury were registered in the vicinity of the heat and power plant or the refinery as chimneys release fumes at high altitudes. What is more, the mercury concentrations recorded in the forests of the Vistula Lagoon and the Hel Pit were nine times lower than the average concentration.

Laboratory experiments carried out on samples of sea water from three coastal measurement stations proved that mixing in the sea surf zone influences the emission of mercury from the water into the atmosphere. The emission of gaseous mercury from the surface waters of the Gulf of Gdańsk was higher in March when the life processes in the water were still slow and bio-accumulation was very limited.

## INTRODUCTION

The usage of mercury dates back more than 2000 years, and during this time many tons of this metal have been discharged into the environment. Since 1890, 200 000 tons of Hg have been emitted into the atmosphere, but only after the outbreak of Minamata disease in the 1950s was the dangerous impact of mercury on human health recognized (Wrembel 1997).

Regional differences in the annual world-wide emission of mercury into the atmosphere were revealed by Pierrone *et al.* (1996). In general, the annual increase of the Hg level in the atmosphere of the northern hemisphere was estimated at 1.5% by Slemr and Langer (1992). However, a more recent paper by Slemr (1996) suggested that this tendency may be reversed. Presently, the emission of Hg has been limited considerably, but the concentration of mercury in the atmosphere has not decreased in proportion to the protective measures which have been implemented (Slemr and Langer 1992, Slemr 1996). The concentration of mercury in the atmosphere is influenced not only by emission, but also by re-emission from natural sources (approximately 2000 t $yr^{-1}$, Mason *et al.* 1994) and by limited anthropogenic emission (4000 t $yr^{-1}$ Porcella *et al.* 1997). The many thousands of tons of Hg (10 800 tons in water organisms) which have been discharged into the marine environment can presently be re-emitted to the atmosphere, and together with the currently emitted mercury it increases the Hg budget (Mason *et al.* 1994).

During the last two centuries, factors such as the industrial revolution, and particularly the growing pulp and paper industry (especially in Finland, Jernelöv *et al.* 1975), the chlorine industry (especially in Sweden, Lindqvist *et al.* 1984) and the enhanced use of mercury in electric equipment and in paints, have all contributed to making the Baltic Sea one of the most mercury-contaminated basins in the world (Wrembel 1997). Furthermore, the use of mercurial fungicides and agricultural pesticides has contributed to mercury pollution. Due to these applications alone, total deposits of Hg in the Baltic Sea area over the last century range from 1000 to 2000 tons (Förstner and Wittmann 1981, Gerlach 1981, Wrembel 1984, Brügmann *et al.* 1985, Lindqvist and Rodhe 1985, Nilsson *et al.* 1989, Lindqvist *et al.* 1990, Lindqvist 1991). The electrolytic decomposition of salty brine for the production of chlorine, high-purity sodium hydroxide (i.e. caustic soda) and hydrogen by means of mercury electrodes is one of the most important sources of mercury in the Baltic Sea (Förstner and Wittmann 1981, Gerlach 1981, Wrembel 1984, Brügmann *et al.* 1985, Nilsson *et al.* 1989, Lindqvist *et al.* 1990, Lindqvist 1991). In 1998, the highest emission of mercury to the atmosphere among the Baltic countries was in Germany (31 t $yr^{-1}$) and Poland (29 t $yr^{-1}$). Research carried out in 1997-98 indicated that the Baltic Sea waters were supersaturated with gaseous mercury (Wängberg *et al.* 2001). Consequently, the Baltic Sea, especially the shallow waters of the Gulf of Gdańsk, along with land sources could become a significant source of gaseous mercury during summertime (Marks and Bełdowska 2001). Mercury is easily absorbed and accumulated by plankton, invertebrates, fish, birds and sea mammals, and as a consequence it enters the food web. Due to the fact that mercury is transmitted from the prey to the consumer and because of direct toxicity and the process of bio-accumulation, the concentration of this metal is higher at higher trophic levels, especially in predatory fish and sea mammals.

The three cities of Gdynia, Gdańsk and Sopot, commonly known as the Tri-Cities, are surrounded by forests and the waters of the Gulf of Gdańsk. The population of the Tri-Cities is approximately one million. The main sources of mercury in the city come from combustion (fossil fuels, coal, oil, gas and wood), wastes (municipal, medical, hazardous wastes and sewage sludge), manufacturing (metal processing, the chemical industry, the refinery, paint production) and other sources such as fluorescent lamps, hazardous and municipal waste sites and deforestation.

The objective of this project was to study gaseous mercury concentrations and the variability of them in the Polish coastal zone around the Gulf of Gdańsk. In addition to the atmospheric research, laboratory experiments were carried out on sea water samples collected in March, April and May. The authors wanted to study the influence of physical mixing and biological processes on the level and the rate of air enrichment with mercury re-emitted from the sea water during the period of the winter-spring shift.

## MATERIAL AND METHODS

### *Field measurements*

The measurements of total gaseous mercury (TGM) were carried out using an automated analyzer Gardis 1A. The automated atomic absorption (AAS) analyzer has a detection limit of 0.5 pg Hg which is comparable or even better than that of existing AFS detectors (Urba *et al.* 1995). The Gardis analyzer is portable and sensitive, which allowed profiles to be made along the Polish coastal zone of the Gulf of Gdańsk (from Hel to Piaski) from March to May (1999) (Fig. 1). The samples of air for the analyses were collected 150 cm above the ground. Samples were collected every 3 minutes during a period of 300 seconds (5 minutes of sampling and 3 minutes for the analysis procedure).

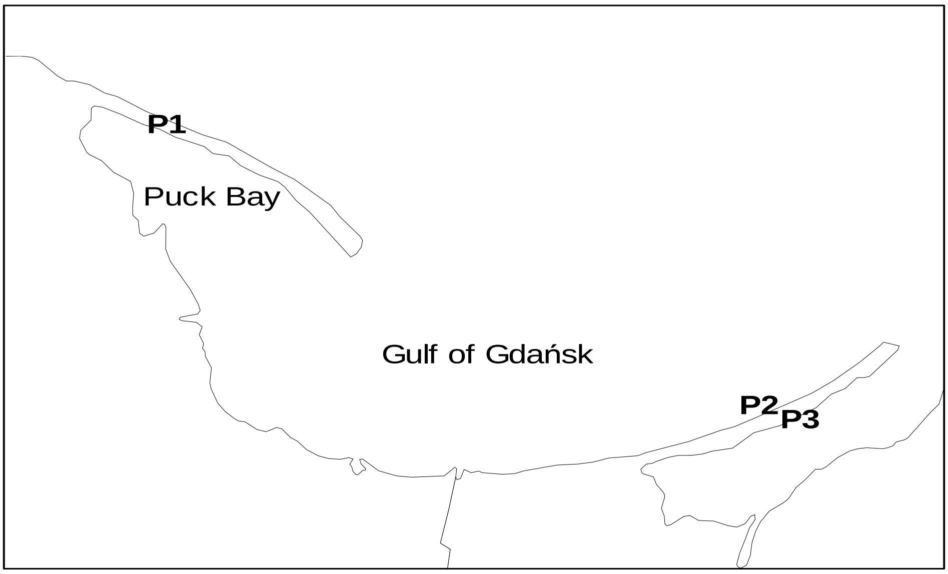


**Fig. 1.** Location of the research stations: P1 Puck Bay – Chałupy, P2 The Gulf of Gdańsk – Krynica Morska, P3 Vistula Lagoon – Krynica Morska.

Prior to the experiments, the device was calibrated on mercury vapor at room temperature. The detection limit was determined as the triple sigma of the blank readings. Mean blank readings did not exceed 0.5 pg. The value of the detection limit for the 300 s sampling period is, by quantity, 0.1 ng $m^{-3}$ and the concentration range is 0.00-500 ng $m^{-3}$. The coefficient of variation calculated for values greater than 20 pg was 3%. The results of TGM were recorded automatically by a computer.

The results of wind direction measurements from four research stations - Gdynia, Sopot, Gdańsk Wrzeszcz and Gdańsk Śródmieście - were obtained from the ARMAG Foundation.

***Laboratory experiments***

Samples of surface water were collected monthly at three research stations - P1 Puck Bay - Chałupy, P2 the Gulf of Gdańsk - Krynica Morska and P3 Vistula Lagoon - Krynica Morska - from March to May 1999 (Fig. 1). The experiments were carried out in March, April and May at the Air-Sea Interaction Laboratory of the Institute of Oceanography, Polish Academy of Sciences in Sopot. Each of the experiments was conducted at a temperature of $10^{o}$ C a few hours after sampling. Filtered clean air was “bubbled” through the water-samples placed in a chamber to simulate the mixing induced by wind or rainfall. After the 'bubbling' process started, the level of TGM concentration above the chamber was recorded with the Gardis 1A analyzer for 30 to 90 minutes. A precise description of the water-air exchange chamber is presented in the paper by Marks and Bełdowska (2001).

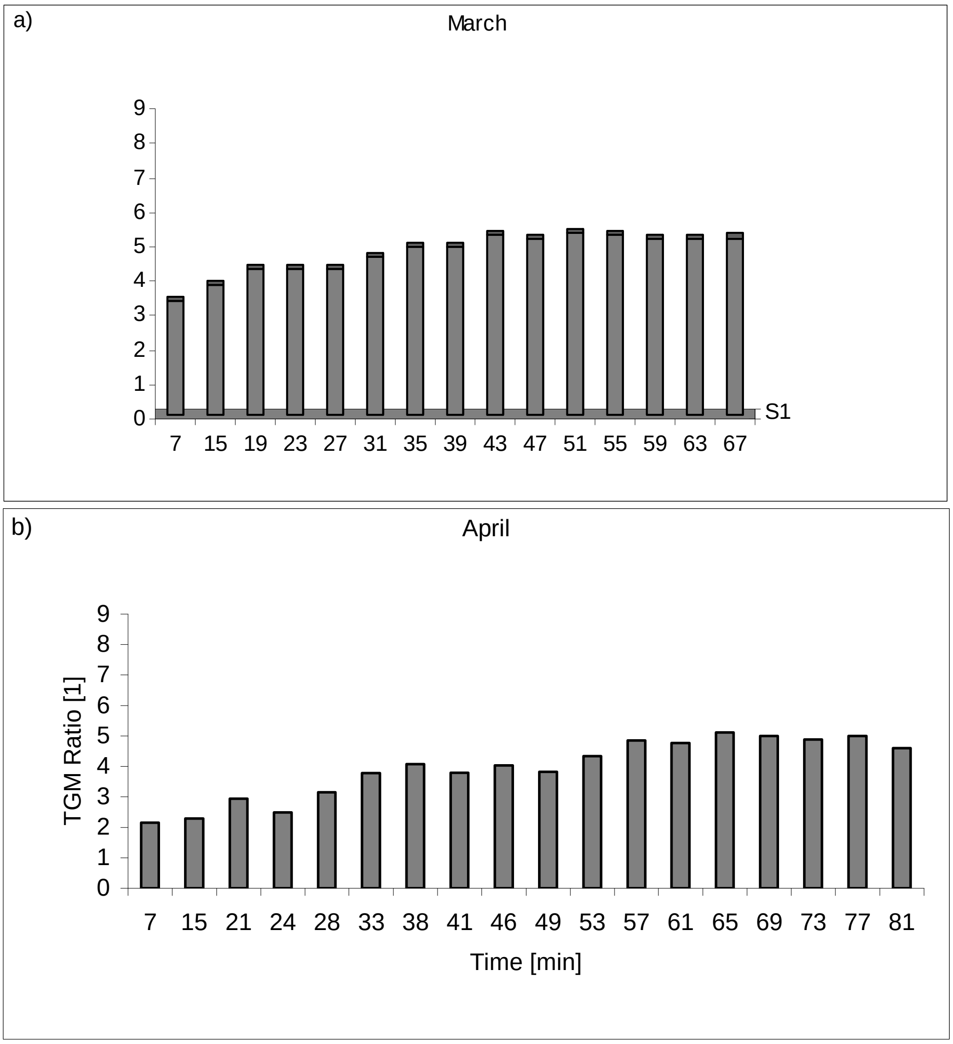


**Fig. 2.** The ratios of TGM measured in the water/air chamber during air bubbling through the water column to that measured without bubbling for water samples drawn from Puck Bay at Chałupy (P1) on a)18 March; b) 21 April.

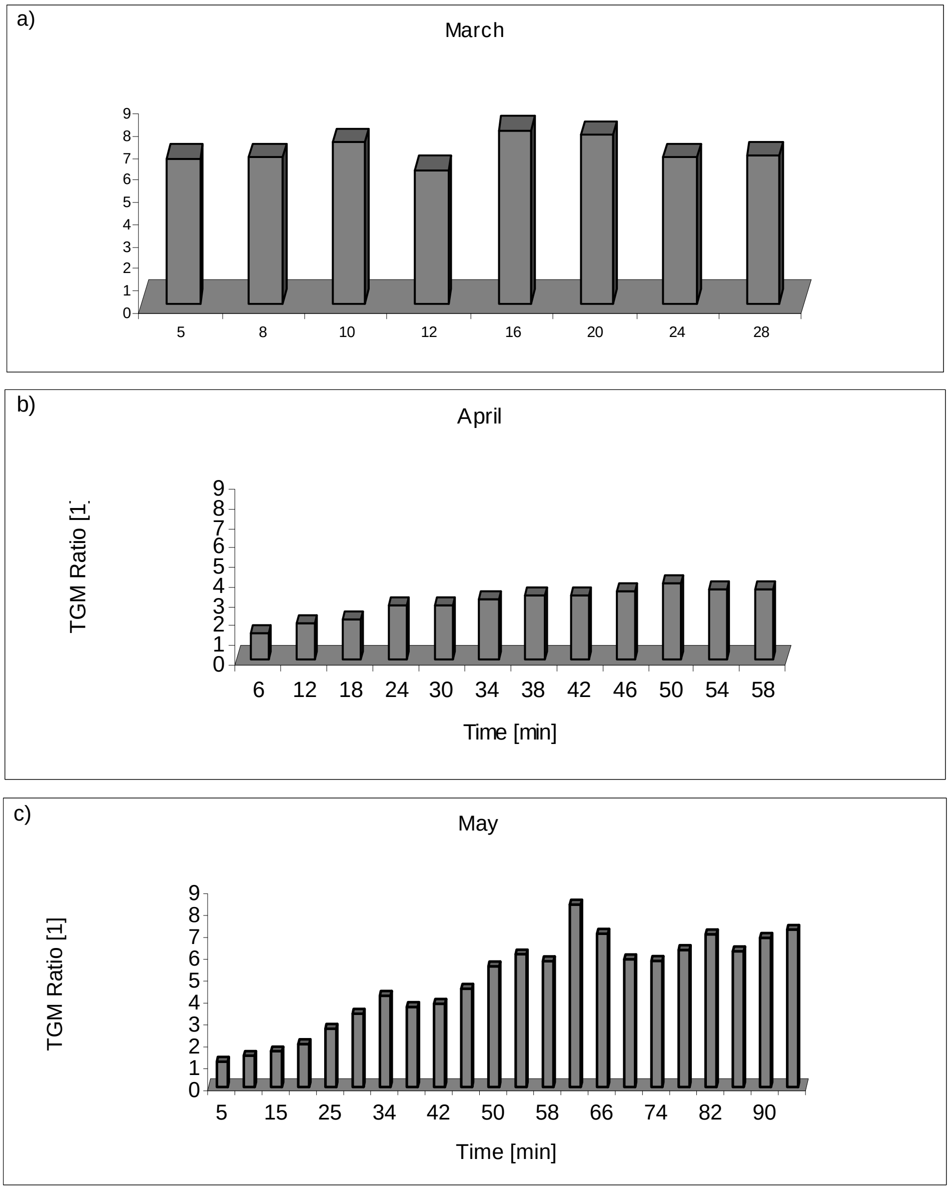


**Fig. 3.** The ratios of TGM measured in water/air chamber during air bubbling through the water column to that measured without bubbling for water samples drawn from Gdańsk Bay at Krynica.

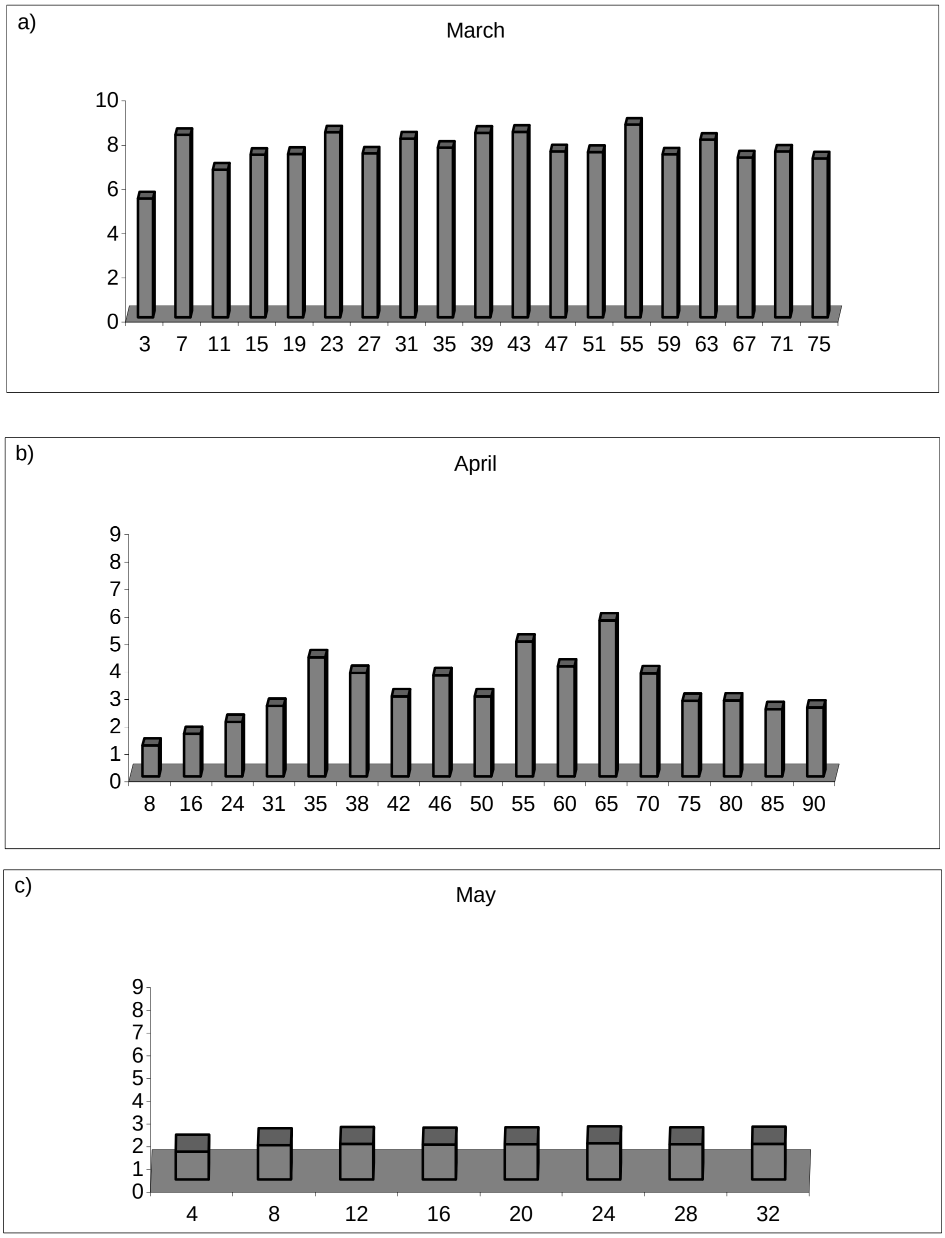


**Fig. 4.** The ratios of TGM measured in water/air chamber during air bubbling through the water column to that measured without bubbling for water samples drawn from Vistula lagoon at Krynica (P3) on a) March b) April c) May.

## RESULTS AND DISCUSSION

### *Laboratory experiments*

The gaseous mercury emission from the water samples into the atmosphere was represented by the coefficient of enrichment R - after 5 minutes ($R_5$), 10 minutes ($R_{10}$) or n minutes ($R_n$). This parameter was calculated as the quotient of the value of the TGM concentration during the process of 'bubbling' after n minutes and the value of TGM concentration at t=0 (the beginning of the experiment).

$$R_n = \frac{TGM_{t=n}}{TGM_{t=0}}, \quad (1.1)$$

where:
$R_n$ – coefficient of enrichment;
$TGM_{t=0}$ – gaseous mercury concentration over the chamber before the 'bubbling' process;
$TGM_{t=n}$ – gaseous mercury concentration after n minutes.

The highest coefficients of enrichment with gaseous mercury were achieved most quickly in March. After several minutes the coefficients of enrichment reached $R_7$=3.5 for station P1, $R_5$=7 for station P2 and $R_7$=8.2 for station P3 (Figs. 2-4). In comparison with the experiments carried out in the following months, in March the surface layer of water in the coastal zone at the three research stations proved to be the most prominent source of mercury which was emitted from water to the atmosphere in the mixing simulation ("bubbling"). In April and May, in the first several minutes of the experiments, the coefficients of enrichment above the samples were comparatively low at 1-2 (Fig. 2-4). After a longer mixing time, the coefficients of enrichment increased ($R_{57}$=4.9 in the station P1; $R_{62}$=8.3 in the station P2). Most probably, during winter storms the shallow waters of the coastal zone are saturated with mercury from both sediments and external sources and thus they become a mercury sink as is suggested by Marks and Bełdowska (2001). Additionally, slower life processes and very limited bio-accumulation in phytoplankton and zooplankton are also conducive to storing mercury in the water at the close of winter.

The above laboratory experiments indicate that the dissolved gaseous mercury (DGM) found in the surface water layer can be emitted to the atmosphere during the mixing process and can be measured as TGM. Due to long residence time, mercury can be transported in the atmosphere to locations which are distant from the source.

### *Potential anthropogenic sources of TGM in the coastal zone of the Gulf of Gdańsk*

The Tri-Cities, similarly to other large industrial centers, is a source of mercury emission into the atmosphere. Depending on the meteorological conditions, the influence of this metal can be either local or regional. The main sources of mercury are fossil fuel combustion, high-temperature processes, paint production, the use of devices containing mercury, municipal waste grounds and their treatment. Not only do these sources increase the concentration of TGM in the air in Gdańsk, Sopot and Gdynia, they also do so in locations situated beyond these municipalities. When meteorological conditions are favorable, the mercury emitted in the Tri-Cities can be transported to neighboring areas.

The concentration values of Hg (0) registered in the air of Gdańsk, Sopot and Gdynia were in most cases higher than the results recorded in the remaining sections of the coastal zone of the Gulf of Gdańsk (Fig. 5). A three-fold increase of TGM was registered in Gdynia when strong winds transported the air from above the shipyard and the harbor (March, April) (Fig. 5). A similar correlation was noted when winds transported air from over the shipyard in Warnemünde, and an increase in the amount of gaseous mercury in the air was observed over the northern part of the Baltic Sea (Marks 2002). The main causes of Hg emission into the atmosphere above shipyards and harbors are high-temperature processes during ship production and painting, ship maintenance using mercurial compounds and the application of fungicides containing mercury.

Even if the shipyards do not use paint or varnish containing mercury compounds (in accordance with current ecological trends), shops and constructions in the shipyards where mercury preparations were previously stored are saturated with Hg vapors. Some of the shipyard wharves were also painted with mercury-containing paints. These constructions emit considerable amounts of TGM (Ebinghaus *et al.* 1999).

An increased concentration of TGM was also observed in the area of the sewage treatment plant in Sztutowo (Fig. 5). There are large amounts of sludge in sewage treatment facilities which can contain mercury compounds. As organic matter in the waste is decomposed (bacterial reduction, abiotic factors), bound mercury can be released and transformed into its gaseous form (Mason *et al.* 1993, Mason *et al.* 1994, Costa and Liss 2000).

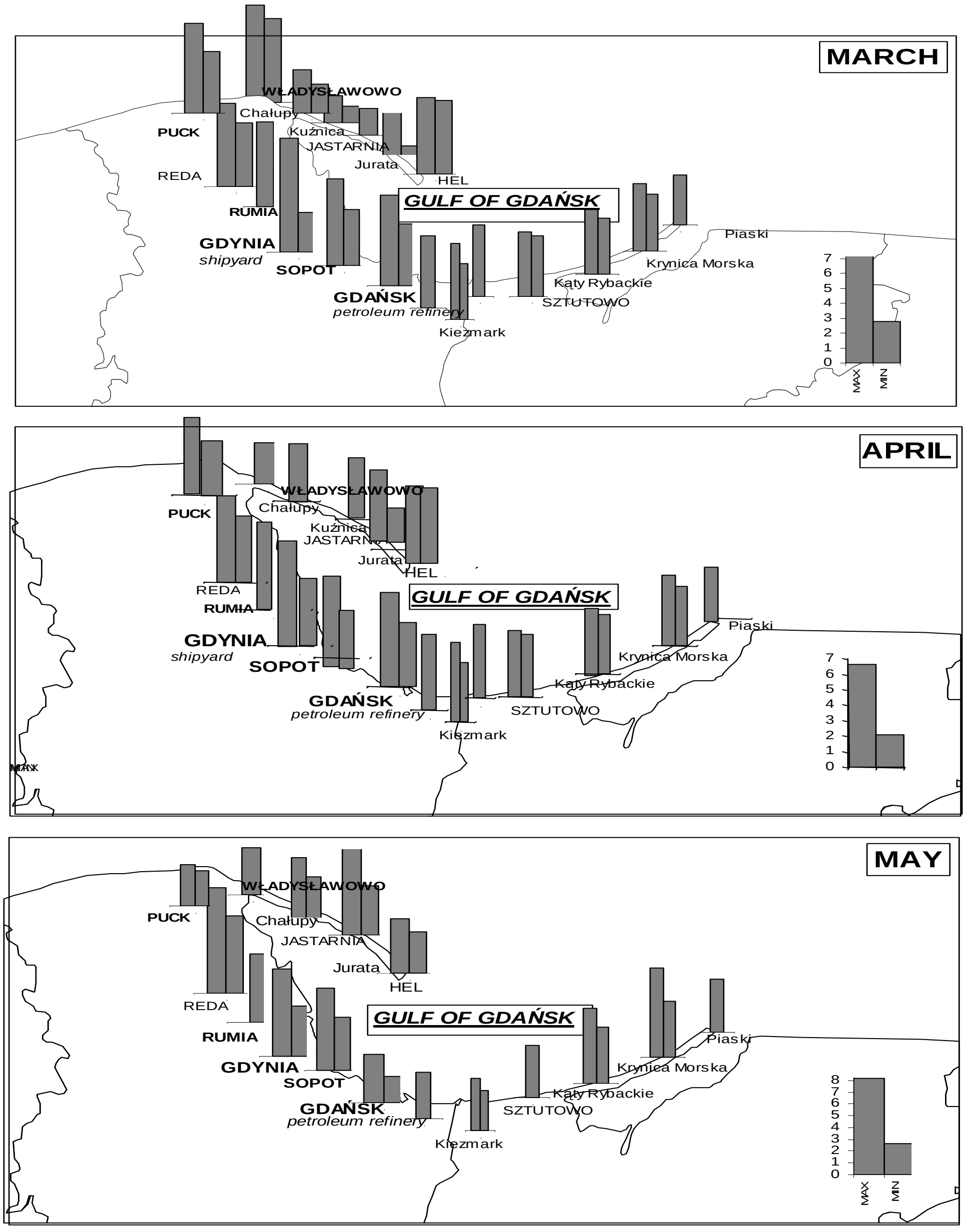


**Fig. 5.** The TGM concentration in the atmosphere over the Gulf of Gdańsk from Hel to Piaski on March (SE-S-SW), on April (SW-W-NW-N) and on May (SE-S-SW). Left bar represents max values on TGM, right bar represents min. values. Single bars represent single measurements.

Measurements taken along the Hel Peninsula indicated a high concentration of gaseous Hg in Hel (Fig. 5). This town is located at the end of the peninsula and is its largest one with port docks, smokehouses, a fishing port and numerous individual household furnaces.

The large amounts of mercury compounds found in fossil fuels can be released into the atmosphere during combustion, i.e. during the production of fuel in refineries and in coal combustion at heat and power generating plants (Wilhelm and Bloom 2000). However, observations do not show a higher concentration of TGM in the vicinity of the heat and power generating plant in Gdynia or at the refinery in Gdańsk (Fig. 5). This is because tall chimneys release combustion products at high altitudes, thus enabling their transport to locations distant from the source of emission.

### *Emission of TGM from the Gulf of Gdańsk and the Vistula Lagoon*

Concentrations of Hg (0) ranged from 0.6 to 8.2 ng $m^{-3}$ in the atmosphere of the coastal zone of the Gulf of Gdańsk. At the close of winter the lowest concentration of gaseous mercury ranged from 0.6 to 3.5 ng $m^{-3}$ in non-urban areas (Hel Peninsula, Vistula Pit) (Fig. 5). With a rise in temperature, the level of mercury vapor concentration was 8.2 ng $m^{-3}$ (Fig. 5). Nearly 50% of the measurements of mercury in the air indicated an average concentration of 3.4 ng $m^{-3}$. The vegetation of forests, meadows and fields in the Polish coastal zone played an important role in maintaining low levels of mercury concentration. In the vicinity of such areas, especially in the Vistula Pit, the registered concentration of TGM was several times lower and was even as much as nine-fold lower in March (Fig. 5).

Mercury contamination in the marine environment reaches maximal levels in the coastal zone and in esturine waters where this element is transported by rivers and the atmosphere. The Helsinki Commission (HELCOM 1993) reported that the atmospheric transport of mercury to the sea is very high and constitutes 50% of the load discharged into the Baltic Sea from all anthropogenic sources. Direct mercury discharge into the Vistula River in the late 1980s reached 39 tons per annum. By the mid 1990s the annual amount of Hg inflow had fallen considerably to only 5 tons per year (Grelowski and Pastuszak 1996). Despite such a substantial reduction in mercury inflow, the amount of this element and its compounds which have managed to penetrate the environment is very hazardous.

Depending on meteorological conditions and the concentration of DGM in the water, the air above the coastal zone of the gulf can be purified by the deposition of mercury from the atmosphere to the sea or, conversely, the level of mercury concentrations in the air can increase. The DGM in water can, through the influence of abiotic and biotic factors, be turned into ionic mercury or undergo methylation. This process occurs in the muddy sediment of the gulf, and the resulting dimethyl mercury (II) $(CH_3)_2Hg$ is toxic. In this form, Hg is accumulated by plankton and benthos and enters the food chain (Keckes and Miettinen 1973, Gerlach 1981, Honda *et al.* 1990). Mercury is cumulated in plant and animal tissues, including those of fish, and its bio-accumulation increases at each trophic level. The circulation of mercury depends on the atmosphere surrounding the water and on meteorological conditions which can allow the ‘escape’ of gaseous mercury from the water into the air (Poissant and Casimir 1996, Ebinghaus *et al.* 1999). The dissolved gaseous mercury (DGM) in the surface layer of the water can be emitted into the atmosphere directly through diffusion, evaporation or during aerosol generation. Increases in the difference between both air (Ta) and water (Tw) temperatures ($\Delta T=Tw-Ta$) and mixing in the surface layer increase the emission of mercury (Marks and Bełdowska 2001).

The intensive mixing of surface waters in the Puck Bay in May which were induced by wind and rain may have caused $Hg_{(g)}$ to be emitted from the water into the air. A high concentration of gaseous mercury was observed at the Hel Peninsula near the coastal line in Chałupy and Jastarnia at this time (Fig. 5). The impact of the re-emission of mercury from the sea to the atmosphere was rather local and was marked in the Polish coastal zone over “clean” areas such as agricultural, tourist and recreational areas where the lowest concentrations of mercury in the air are usually recorded. This was observed several times in Krynica Morska, Kąty Rybackie, Piaski and in the vicinity of the arable land and forests of the Vistula Pit (Fig. 5).

The analysis of wind direction and velocity indicated that at lower wind speeds (approximately 1 m $s^{-1}$) the air from both the Gulf of Gdańsk and the Vistula Lagoon, which may have elevated mercury levels due to re-emission from the water, stayed in the region of the Vistula Pit. Simultaneous laboratory experiments proved that the re-emission of mercury from the sea in each of the measurement periods was probable and that it occurred most rapidly in March ( $R_7$=8, Fig. 2a; $R_5$=7, Fig. 3a).

However, the illustrated process is presumed to be of local importance only. On a global scale, it is estimated that the emission of mercury to the atmosphere from seas and oceans reaches $5x10^5$ g $yr^{-1}$, but this amount is considered to be rather small in comparison with that from combustion which is estimated at $1.6x10^9$ g $yr^{-1}$ (GESAMP 1980).

The content of mercury in the atmosphere over the Gulf of Gdańsk does not differ much from that in other maritime regions of the Baltic Sea and the North

Sea. At the Rorvik coastal station in the Danish Straits the average registered concentration was 2.8 ng Hg $m^{-3}$ and in Overbygd by the North Sea it was 2.6 ng Hg $m^{-3}$ (Lindqvist 1991).

The measured concentration values of gaseous mercury in the atmosphere of the coastal zone and over the open area of the Gulf of Gdańsk are much lower and have never exceeded the values permitted by the regulations of the Ministry of Environmental Protection, Natural Resources and Forestry (MOŚZNiL) issued on 28 April 1998 (Dziennik Ustaw, No 55, art. 355). According to this regulation, the maximum permissible level of mercury is 0.7 $\mu$g $m^{-3}$ in a 30-minute sample, 0.3 $\mu$g $m^{-3}$ in a 24-hour sample and 30 ng $m^{-3}$ in a one-year sample.

The concentrations of gaseous mercury registered over the Gulf of Gdańsk were not in compliance with the strict standards for the air over health resorts.

## CONCLUSIONS

The concentration of mercury in the air over the coastal zone of the Gulf of Gdańsk did not exceed maximum permissible levels and remained at levels similar to those of other maritime regions of the Baltic Sea and the North Sea.

There are sources of gaseous mercury in the coastal zone of the Gulf of Gdańsk. The high concentration of mercury in the air was registered in the Tri-Cities in the shipyards, the harbor in Gdynia and at the sewage treatment facility in Sztutowo.

The Gulf of Gdańsk and the Vistula Lagoon are potential sources of mercury which, through re-emission from the water to the atmosphere, enriches the air of the coastal zone.

Mixing processes increased the re-emission of mercury from the water to the atmosphere. The most rapid enrichment of air with mercury occurred in samples collected at the close of winter when life processes in the water were still in the undeveloped stage.

The heat and power generating plant in Gdynia and the Gdańsk Refinery did not induce a local increase of gaseous mercury concentration in the air 150 cm above the ground.

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