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Bimodal variation in mercury wet deposition

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Bimodal variation in mercury wet deposition to the coastal zone of the southern Baltic

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Abstract

In the following periods: November 2005–June 2006 and October 2007–January 2009, concentrations and deposition rates of total mercury (THg) and Hg(II) were measured in precipitation over the urbanized and industrialized area of the southern Baltic – the city of Gdynia. Rains over the coastal zone had different concentrations of total mercury, they ranged from 8.6 to 118.0 ng L⁻¹, out of which about 32% were labile, inorganic forms, easily reducible in a SnCl₂ solution. Over the southern Baltic two maxima of concentrations were observed: first, in the heating season and second, in the non-heating season. Elevated concentrations of mercury in precipitations during heating seasons were the result of the activity of local emission sources (intensive combustion of fossil fuels in domestic furnaces and individual power and heat generating plants). During the warm season, precipitation over the southern Baltic could clean the air from Hg remitted from sea and land surfaces. Precipitations, which purified marine and continental air masses were responsible for the comparable input of mercury to the coastal zone. The wet deposition value in 2008 was estimated to be 28.9 μg m⁻². In the coastal zone of the southern Baltic, acid precipitations with the elevated Hg concentrations are very frequent.

1 Introduction

Mercury and its derivatives are highly toxic. Contamination of land and marine ecosystems with mercury poses threats to organisms living in them. This heavy metal has the ability to bioaccumulate and biomagnify through the marine aquatic food chain (Chang, 1996; Morel et al., 1998). Against the background of complex pathways for mercury in the environment, the atmospheric processes play a crucial role in the global mercury cycle. Mercury vapors may be carried by air currents to areas remote from natural or anthropogenic emission sources. Atmospheric mercury exists in the form of organic and inorganic compounds, in a gas phase – Total Gaseous Mercury (TGM), partic-

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ulate phase – Total Particulate Mercury (TPM) and dissolved or suspended in cloud drops and hydrometeors. According to published data, global emission of mercury is estimated to be 4500–5000 tons a⁻¹, out of which about 50–70% comes from anthropogenic sources (Pirrone et al., 1996; Lamborg et al., 2002; Mason and Sheu, 2002).

5 Elemental mercury can be transported over long distances in the atmosphere (long-range transport) or undergo transformations mainly by chemical reactions with ozone, hydroxyl radical, hydrogen peroxide, nitrate, halogens containing compounds such as Cl[·], ClO, Br[·], BrO, ClBr (Lin and Pehkonen, 1999; Raofie and Ariya, 2004; Saiz-Lopez et al., 2008) to the forms more effectively deposited, e.g. Reactive Gaseous Mercury (RGM) and/or TPM.

10 Both geographic location of the sources and seasons are of great importance in terms of emission rates of mercury and its spatial distribution (Pirrone et al., 1996; Kim et al., 2005). Urbanized and industrial centers are characterized by concentration levels of RGM, TPM over the long-term periods and by an easily predictable diurnal pattern of TGM concentration associated with the activity of anthropogenic emission sources of mercury (Chen et al., 2004). Numerous independent monitoring and experimental studies on unnatural emission sources confirmed a similar diurnal pattern of reactive gaseous mercury concentration for most of the urban sampling sites: maximum concentrations were observed at about midday and minimum – at night (Nakagawa, 1995; Lindberg and Stratton, 1998; Keeler and Dvonch, 2005; Liu et al., 2007). In contrast, uninhabited regions (e.g. deserts, steppes, open seas and oceans) where there are no anthropogenic sources of Hg, had relatively low reported concentrations of TGM, often at the level of the environmental background, resulting from natural processes and reemission. For European and North American areas TGM concentrations are about 1.5–1.4 ng m⁻³ (Lindberg et al., 2007). In natural environmental conditions, besides areas of the environmental background, there are also places with remarkably high emission of mercury associated with volcanism, geothermal activity, exploitation of ores containing mercury and biomass combustion.

Coastal zones play an exceptional role in the transformation of atmospheric mercury,

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both on a regional and global scale. Some of the zones, e.g. in the polar regions in the Arctic and Antarctica, can be used as an example of the changes to the environment caused by mercury. These coastal areas have no direct contact with anthropogenic sources. Nevertheless, they act as reservoirs for toxic mercury. Due to high mercury deposition on snow and ice surfaces, Hg^o is periodically depleted in the atmosphere over such places. As a result of this phenomenon called AMDE (Atmospheric Mercury Depletion Event), 325 tons of mercury per annum are deposited in the Arctic from the atmosphere, while deposition without AMDE amounts to 225 tons per annum on average (Ariya et al., 2004).

In coastal areas with well developed industrial infrastructure, qualitative and quantitative transformations of atmospheric mercury are conditioned both by anthropogenic and natural processes. Apart from ozone, hydroxyl radicals and hydrogen peroxide, other chemical compounds – sea salts, halogens and marine aerosols - should also be taken into account while considering transport, dispersion and transformation of mercury. In certain conditions the sea surface can become the main source of mercury for the near-water atmospheric layer. Reemission of Hg across the marine surface microlayer to the atmosphere is controlled by various physical, chemical and biological factors. The effectiveness of reemission is strictly connected to the influence of solar radiation and the ability of organic matter to reduce divalent mercury ions (Marks and Bedowska, 2001; Rolfhus and Fitzgerald, 2001; Wängberg et al., 2001; Lanzillotta et al., 2002; Andersson et al., 2007).

Among the atmospheric phenomena which are of crucial importance to mercury transformation in the liquid phase, special attention should be paid to precipitation. Deposition links the atmospheric and land-sea part of the mercury cycle. At moderate latitudes, the most effective way of mercury deposition is by wet precipitation. However, dry precipitation of gases which are subject to phototransformation and sorption on to atmospheric aerosol particles can purify the air from mercury compounds as well (Sakata and Marumoto, 2005). The concentration of mercury in precipitation varies seasonally (Sorensen et al., 1994; Hoyer et al., 1995) and with the place of occurrence

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(Mason et al., 1997; Glass and Sorensen, 1999). A group of meteorological factors that influence chemical reactions of mercury in the liquid phase includes the following parameters: air temperature and relative humidity, solar radiation, velocity and direction of wind, occurrence of thermal inversion layers, tropopause properties (height of the marine and planetary boundary layers). The character and duration of precipitation are also important factors. It is commonly known that long-lasting drizzle purifies the atmosphere from mercury and many other chemical compounds. When considering mercury speciation in precipitation, we should take into account chemical composition and pH of the fall, dissolved and suspended organic matter, compounds which catalyze oxidation/reduction processes (Munthe and McErloy, 1992). Typical for urbanized and industrialized city centers, low pH of the precipitation very often creates conditions in which methylation of Hg^{2+} into its most toxic derivative occurs (Hammerschmidt et al., 2007). Coastal zones, which are the source of sea salts and marine aerosols, are the places where under long-lasting low pH conditions, the problem of acid precipitation can become a problem of toxic precipitation. For this reason, the aim of the present study was to determine conditions for mercury transformation in precipitation occurring over the coastal zone – the place where continental and marine air masses meet.

About a four scale decrease in Hg emission was reported in Europe for the period 1980–2005 as a result of economic changes and efforts directed at air protection (Munthe et al., 2001; Pacyna et al., 2009). On the other hand, elevated concentrations of mercury in precipitation over the Scandinavian part of the Baltic Sea observed in previous years can be associated with the emission in Asia, which indicates the importance of mercury as a worldwide pollutant (Wilson et al., 2006; Pacyna et al., 2009). As a consequence of climate warming, mercury removed from water returns to the atmosphere in a shorter time and is transformed into more mobile forms. The exchange of mercury across boundary layers can be caused by biological factors and water circulation (Strode et al., 2007; Sunderland and Mason, 2007). In the oligotrophic parts of the world ocean, the inflow from the atmosphere is considered to be the main source of Hg to the surface waters, while in the coastal zones, contamination of the environment

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by mercury is proportional to the number of emission sources. It is claimed that the elevated concentration of Hg in precipitation over coastal zones can be caused by two reasons. Firstly, it can be associated with the aerosol activity of the sea (reemission, sea breeze, storms, upwelling, diffusion fluxes through the boundary layers, blue-green algae blooms) and, secondly, it can occur as a result of the additional input from the scattered terrigenous and/or anthropogenic sources on the land.

The data published by HELCOM reveals that the annual input of mercury to the Baltic Sea only from anthropogenic sources is estimated at several dozen tons. The total annual deposition of mercury to water surface decreased from 72 tons in 1990 to 41 tons in 2004. Despite the fact that a significant reduction in mercury emission is observed to the air by the Baltic countries (i.e. Denmark, Estonia, Finland, Germany, Latvia, Lithuania, Norway, Poland, Sweden and Russia), the issue of toxic mercury in precipitation over the Baltic still remains an open problem.

In the coastal zone of the Gulf of Gdansk, anthropogenic sources of mercury contamination can be linked to the processes of burning fossil fuels in different heat and power plants, individual power and heat-generating stations, domestic boiler-rooms, furnaces, hospital incineration units. According to the inventory of anthropogenic heavy metal emissions in Poland, the annual total mercury emission in 2000 was 25.6 tons (Hlawiczka and Fudala, 2003). It was confirmed that combustion gases (untreated or after passing through a desulfurization system, electro-filter and fabric filter) contain the highest concentrations of gaseous elemental mercury – Hg⁰, slightly lower amounts of Hg²⁺ oxidized forms and mercury bound to suspended particles, as well as traces of methylmercury (Hlawiczka et al., 2003). In Poland, 10% of the countrywide Hg emission to the atmosphere is the result of the combustion of power-generating materials in the municipal-domestic sector (domestic furnaces, small and individual heat-generating stations etc.) (Hlawiczka, 1998). The processes of fuel combustion are currently regarded as the largest source of mercury to the atmosphere in the area of the Gulf of Gdansk.

2 Materials and methods

2.1 Description of the measurement site

Mercury concentrations and wet deposition measurements were carried out in the coastal zone of the Gulf of Gdansk, in the city of Gdynia, Pomerania, Poland. The measurement site was located in the center of Gdynia ($\phi=54^{\circ}30' N$, $\lambda=18^{\circ}32' E$), Oceanography Institute, at the University of Gdansk (20 m over ground level, above the tree-tops, about 1 km from the coastline). Gdynia is located in the morphologically and economically diversified area, known as the Tricity Agglomeration, in a temperate climate zone with the features of marine and continental type (Fig. 1), within the range of zonal circulation initiated by cyclonal activity, along the polar and arctic front and under the influence of the permanent Azorean High and the Icelandic Low. Polar marine and tropical marine humid air masses from the Atlantic Ocean, polar continental dry air from east Europe and arctic air from the north flow in to the Gulf of Gdansk. This area is affected by frequent thermal changes due to the flow of air masses having different physical properties. Winter weather alternate between relatively warm and drizzly or snowy, dry and frosty. Summer periods are either cool and rainy or warm and dry. On the basis of data collected over many years from the coastal zone of the southern Baltic, the average month temperature for winter periods is slightly below 0°C and for summer around 23°C . The mean annual sum of precipitation for the Polish coast is 550–600 mm. Precipitation structure is often influenced by the prevailing circulation type.

2.2 Hg measurements

Precipitation samples for total and ionic mercury concentration analyses were collected during the following periods: November 2005–June 2006 and October 2007–February 2009. Samples were collected during precipitation or straight after it with bulk samplers (reception surface of 0.0314 m^2) to the acid-washed borosilicate glass bottles. All the

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elements of the measurement system were made of Teflon and materials which do not absorb mercury. Each time, after exposition finished, the samples were transported to the Analytical Laboratory of Mercury at the Oceanography Institute. Total mercury concentration in precipitation was determined, after its oxidation by BrCl and reduction by SnCl₂ to Hg⁰, by atomic absorption technique. Both forms of Hg were measured by an automatic analyzer of total gaseous mercury Gardis3. The detection limit of the Gardis3 analyzer is 0.5 pg (Urba et al., 1995). The detection limit of the method applied for THg and Hg(II) in precipitation was established on the basis of 3×SD of the blank. Values for the blanks were not higher than 0.5 pg.

Since in the period from spring to late autumn precipitation contained large amounts of suspended atmospheric particles, the dissolved fraction for further analyses was separated from the suspended fraction by filtration using Whatman filters (the effective pore diameter of 0.7 μm). In the laboratory, the samples of precipitation were acidified using 12M HCl in the amount of 0.5% per volume in order to eliminate mercury loss prior to analyses. Samples were protected from light and stored at 4°C in borosilicate glass bottles. Each time, the influence of preservation on analyte concentration in the samples and the storage period were taken into account when calculating the final result.

For each (i)-sample, deposition of mercury in precipitation (μg m⁻²) was calculated from the Eq. (1)

$$\text{THg}_{\text{deposition}} = \text{THg}_i P_i \quad (1)$$

where THg – mercury concentration in rainwater sample (microgram per l) and *P* – precipitation amount in bulk sampler (mm).

The pH values were measured with a glass electrode (type SenTix 41-3). The pH-meter was calibrated prior to measurement with two standard solutions of 7.00 and 4.00 pH values. Snowfall samples were melted thoroughly at room temperature.

Environmental conditions for each rain episode were described on the basis of primary air parameters i.e. temperature, relative humidity, direction and velocity of wind

(10 s record averaged to 30 min). Meteorological data were recorded continuously by stationary instrumentation (Huger Weather Station). Trajectories of air masses moving over the measurement site were retrieved from the HYSPLIT model based on GDAS (global, 2005-present) (Draxler and Rolph, 2003). End points of trajectories were established on the basis of the number of hours from the time of starting the exposition to the time of collecting the precipitation, in relation to the stable point in space, with a standard time step of 96 h.

3 Results and discussion

Many scientists emphasize the importance of mercury and its transformations in the atmosphere for coastal areas of oceans and seas (Marks and Bedowska, 2001; Pirrone et al., 2003; Wängberg et al., 2008; Sprovieri et al., 2009). The results of the measurements of mercury concentration in precipitation over the coastal area of the southern Baltic suggest the complex character of Hg transformations with predominance of land factors and the crucial influence of the sea as a source and reservoir of atmospheric mercury.

In the years 2005–2008, the concentration of total mercury in precipitations over the coastal zone of the southern Baltic ranged between 8.6 and 118.0 ng L⁻¹, 90% of values were within the range of 11.6–82.1 ng L⁻¹. The mean of THg concentrations in precipitations over Gdynia was 42.6 ng L⁻¹ (the median amounted to 39.3 ng L⁻¹) (Table 1). The concentration of reactive mercury forms – Hg(II) (complex compounds of mercury easily reducible in SnCl₂ solution) – oscillated between around 1.0 and 53.9 ng L⁻¹, constituting on average 7–46% of the total mercury.

In the coastal zone of the Baltic, distribution of Hg concentrations in precipitation is diversified. Concentrations of THg measured over the Lithuanian coast (eastern Baltic) were lower than those found at the Polish coast and were between 10 and 30 ng L⁻¹ (Milukaite, 2008).

Ranges of total mercury concentration in rains over the coastal zone of the south-

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ern Baltic were higher than values from Europe and the United States, where bulk collectors were also applied (Landing et al., 1998; Ebinghaus et al., 1999). For example, the average concentration of total mercury measured in precipitation over Mace Head (the Irish coast of the Atlantic) was 17.0 ng L^{-1} (Ebinghaus et al., 1995). In the 1990s, at the stations monitoring the marine atmosphere (EMEP centers) located by the North Sea (Sylt) and on the western coast of the Baltic (Zingst), concentrations of THg in precipitation were reported to be within the following ranges ($6.1\text{--}18.3 \text{ ng L}^{-1}$ and $20.0\text{--}110.6 \text{ ng L}^{-1}$, respectively (Ebinghaus et al., 1995). The values of THg in precipitation over the southern Baltic were higher than those measured in the Upper Midwest, USA: ($4.3\text{--}28.9 \text{ ng L}^{-1}$) (Glass and Sorensen, 1999) and in coastal and inland zones of FAMS, Florida, USA: up to 20.0 ng L^{-1} (volume-weighted average concentrations for bulk deposition samples) (Landing et al., 1998).

3.1 Bimodal character of precipitation over the coastal zone of the southern Baltic

Inorganic and organic, reactive and particulate forms are the main mercury compounds deposited through wet precipitation. Wet deposition values of THg presented in Fig. 2 revealed a bimodal character (in an annual cycle) of precipitation, which purifies the air of mercury. Despite the fact that HELCOM reports (2006) emphasize the lack of seasonal differentials in Hg concentrations, in precipitation over the southern Baltic two maxima (winter and summer) of Hg were observed.

Usually, the highest concentrations of total mercury (over $42.6 \text{ ng L}^{-1} \text{ month}^{-1}$) were measured either in the middle of the heating season or at the end of winter—during large-scale and convectonal precipitation events. At the beginning of spring (April–May), the concentration of mercury in precipitation was low until the warmer days of the vegetation season came (July–August), when the concentration of Hg in precipitation started to become higher. In autumn months (September–October) the concentrations were lower again. The winter maximum of THg concentration in precipitation was strongly correlated to the activity of anthropogenic sources, namely intensive combus-

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tion of power-generating raw materials in the municipal-domestic sector (domestic furnaces, individual heat-generating stations and heat-generating plants). Similar cause-effect connections were observed in other urbanized and industrial areas of coastal zones (Wängberg et al., 2008). Elevated concentrations of Hg in summer precipitations, when some of the point sources of Hg emission were not active (e.g. domestic furnaces), were associated with the reemission of Hg, which took place in the surface waters of the Baltic and the adjacent land during periods of intensive sun radiation and high temperature. The occurrence of that process in the warm season was proved by Ferrara et al. (2000a); Mason et al. (2001); Wängberg et al. (2001).

Deposition of mercury through wet precipitation in the coastal zone of the southern Baltic is dependent on the type, height, intensity and duration of precipitation. In July and August, high precipitations (downpours) connected to atmospheric front passages or storms caused the highest mercury fluxes. For example, in August 2008 when the highest precipitation (120.6 mm) occurred, $5.8 \mu\text{g m}^{-2} \text{ month}^{-1}$ of mercury was deposited, which constituted about 20% of the annual input from the atmosphere to the coastal zone ($28.9 \mu\text{g m}^{-2} \text{ a}^{-1}$). High deposition ($3.7 \mu\text{g Hg m}^{-2} \text{ month}^{-1}$) was also reported at the end of the 2007/2008 heating season (March). However, in this case, heavy snow and sleet falls (on average 71.6 mm) were the cause of this.

On the basis of the following information on the components of the mercury cycle in the atmosphere over the Baltic:

- average concentrations of TGM are: 1.7 ng m^{-3} (heating season) and 3.0 ng m^{-3} (non-heating season) (Beldowska et al., 2008), and
- the average concentration of THg in 1 L of precipitation is 42.6 ng,

it was found that a 1000-fold lower volume contains a 14-fold higher amount of mercury, which proves that wet precipitation over the coastal zone of the southern Baltic purifies the atmosphere effectively.

Despite various amounts of mercury in precipitation from different months, the average concentration values of THg in precipitation over the coastal zone of the southern

Baltic in heating and non-heating seasons were similar (Fig. 3).

During the 2005/2006, 2007/2008 and 2008/2009 heating seasons, medians amounted to: $54.4 \text{ ng Hg L}^{-1}$, $45.0 \text{ ng Hg L}^{-1}$ and $39.7 \text{ ng Hg L}^{-1}$, respectively. The largest dispersion of THg concentration values in precipitation over the measurement site was reported in the winter 2005/2006. The minimum value was 9.8 ng L^{-1} , whereas the maximum was 118.0 ng L^{-1} . Almost 3-fold higher values of Hg(II) concentrations in precipitation during the 2006 heating season in comparison with the non-heating season were the result of intensified emission of that metal into the atmosphere during the long and frosty winter, when air temperatures were about -30°C . In the 2005/2006 heating season, the processes of fossil fuels combustion was intensified over the southern Baltic, especially in the individual power and heat-generating stations. It was reflected by the elevated values of $\text{TPM}_{0.7}$ (up to 2000 pg m^{-3}) recorded from December 2005 to March 2006 at the coastal station in Gdynia by Beldowska et al. (2007). The input of mercury labile forms from precipitations during that period was 3 t higher (17.3 ng L^{-1}) than during the other seasons. The 2008/2009 heating season was warmer than 3 years before and the ranges of $\text{TPM}_{2.2}$ and $\text{TPM}_{0.7}$ concentrations were lower: $3\text{--}151 \text{ pg m}^{-3}$ and $1\text{--}40 \text{ pg m}^{-3}$, respectively, which consequently influenced concentration values of inorganic, ionic forms of mercury in precipitation from 0.6 to 43.7 ng L^{-1} .

The variability in Hg(II) concentrations and pH in precipitation over the southern Baltic in the year 2008 showed that pH could control the forms of $\text{Hg}_{(aq)}$ (Fig. 4). In the heating season, precipitations with Hg(II) concentrations over the average value (12.6 ng L^{-1}) predominated in the acid samples ($\text{pH} < 5.0$) and the highest concentration (53.9 ng L^{-1}) of ionic mercury was reported at $\text{pH} = 4.9$. In the summer, the majority of precipitation samples had a pH over 5.0.

It seems that the dependences presented above (in winter–predominant number of precipitation events with $\text{pH} < 5.0$, and in summer—with $\text{pH} > 5.0$) could be typical for industrialized coastal areas with seasonal distinction and crucial for the bimodal distribution of mercury input with precipitation during a year. The lack of a clear statistical

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relationship between the pH of precipitation and the concentration of mercury confirmed the complexity of mercury transformation processes in the coastal zone, where continental and marine masses of air meet and neutralize (Siudek et al., 2006).

3.2 Spatial diversification of Hg concentrations in precipitation over the southern Baltic

Over the coastal area of the Baltic Sea, precipitation varies in concentration of Hg and the input of mercury through wet deposition (HELCOM, 2006). Rains purifying the air in coastal zones of the northern part of the Baltic usually have lower values of mercury concentration in comparison with rains from the southern or western parts. In the south-western part of the Baltic, the highest concentrations of THg in precipitations were observed out of the heating season (HELCOM, 2006). Maxima of total mercury concentration in precipitations over the Baltic EMEP stations were measured at Zingst in summer and Råö in spring (24.0 and 20 ng L⁻¹ month⁻¹, respectively). These values were lower than concentrations of THg in precipitations over Gdynia. Data on mercury wet deposition collected by us in the years 2005–2008 varied within the wide range: 0.4–5.8 μg m⁻² month⁻¹. The values were almost four times as high as those measured in wet precipitation by Milukaite et al. (2008) in Preila (Lithuanian coast), where deposition maxima (about 1.0 μg m⁻² month⁻¹) were reported in the heating season.

Variabilities of mercury concentrations observed over urbanized and industrialized areas of the southern Baltic could be typical for similar coastal regions in temperate latitudes. Previous studies showed that the total deposition of mercury at the coastal stations situated in the North Sea and in the Danish Straits varied within the range of 6.1–42.0 μg m⁻² a⁻¹ (Jensen and Iverfeldt, 1993). In the Mediterranean Basin, at Cabo de Creus (Spain) the average concentration of THg in wet precipitation was calculated to be 23 ng L⁻¹ and the total deposition amounted to 6–8 μg m⁻² a⁻¹ (Wängberg et al., 2008). Concentration values of Hg measured in rains over Mediterranean were twice lower than those observed over the southern Baltic. However, taking into account the fact that volumes of precipitation from the Mediterranean were 3 times lower, dif-

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ferences in deposition rates between those two areas were significant. In the tropical zone (south-eastern coast of Florida), the highest concentrations of Hg in precipitation—about 82 ng L^{-1} were generally reported in the warmer season (Landing et al., 1998). According to the NADP data, the coastal zone of the Gulf of Mexico (especially the Gulf Coast and Florida) where hurricanes and heavy rains occur very often, are the regions of North America with the highest (over $18 \mu\text{g m}^{-2} \text{ a}^{-1}$) Hg input in precipitation (Prestbo and Gay, 2009).

The emission of Hg in Poland was significantly reduced from 38.8 tons in 1985 to 21.0 tons in 2006 (Hlawiczka et al., 2006). Nevertheless, in the southern region of Poland (Silesia), 500 km from the Baltic coast, deposition of Hg through precipitation still remains high. Zielonka and Nowak (2009) estimated that wet deposition of THg in that highly industrialized and urbanized region of Poland was 28.7 mg m^{-2} in 2008, out of which 20.0 mg m^{-2} in the heating season. That area could be the source of Hg in precipitation for the Baltic, when the southern advection of air masses occurs.

3.3 Distant and nearby sources of mercury deposited through precipitation

Solar radiation as well as the occurrence and concentration levels of compounds having oxidizing properties, i.e. ozone, hydrogen peroxide, nitrogen and hydroxyl radicals, influence transformation of atmospheric mercury. Spatial variability of particulate and reactive forms of Hg is controlled by atmosphere dynamics.

Over the coastal zone of the Baltic when the wind speeds were low (up to 1 m s^{-1}), the precipitation was the main factor responsible for purifying the air from mercury originating from the sources located close to the measurement site (Fig. 5). Elevated concentrations of mercury in precipitation ($>20 \text{ ng L}^{-1}$) were observed when the speed of wind was 3 m s^{-1} and it blew to the coastal area from different directions. When the wind speed was over 3 m s^{-1} and it blew from north, north-east (the marine sector) or eastern and southern directions (the land sector), even higher concentrations of mercury in precipitation ($>30 \text{ ng L}^{-1}$) were reported. That high Hg concentration was probably caused by emission from the nearby industrial sources (i.e. oil refinery,

transshipment harbour, repair shipyard, heat and power plants) and from distant (several hundred kilometers or more) sources located in strongly industrialized regions of Poland, Ukraine, Belarus and Germany.

The analysis of the of air masses flowing over the coastal zone of the southern Baltic enabled to determine directions of the transboundary transport. The assignment of trajectory to one of the separated sectors: marine or continental, was done under the condition that at least 50% of it was included in the area described by the borders of the sector (the division along the 54th parallel). The analysis of back trajectories (24–, 48– and 96–h) over Gdynia led to the conclusions that high concentrations of mercury in precipitation were caused by air masses flowing over the most polluted with Hg regions in Poland (Silesia) and abroad (Ukraine, Germany, Belarus, Czech Republic, Austria). Regardless of the season, THg concentrations over the average value (42.6 ng L^{-1}) were measured in precipitations which purified masses of air flowing from the land (Fig. 6). The additional crucial parameter which also controlled the amount of Hg in precipitation was the height at which the air masses were flowing. The lower they were flowing, the higher were the concentration of Hg in wet precipitation. This dependence was observed during both seasons.

The average values of THg in precipitations over the coastal zone of the southern Baltic measured during the flow of air masses from the north were comparable to the values obtained for continental masses of air (Table 3). Reemission of mercury from the sea surface is a possible reason of the elevated concentrations of this metal in precipitation. The presence of this process was confirmed in the studies on transformations of mercury in the Baltic surface water (Marks and Bedowska, 2001; Wängberg et al., 2001). The authors pointed out that high temperature favors releasing the mercury from water. This process can be intensified by the increase in turbulent mixing in the sea-air boundary layer.

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Concentrations of mercury in a liquid phase in the atmosphere over industrialized and urbanized areas of the southern Baltic were measured from November 2005 to January 2009. Data obtained during the measurements proved the wide variability of the analyzed forms of mercury. The average concentration of the total mercury was $42.6 \pm 23.5 \text{ ng L}^{-1}$, out of which the reactive forms constituted approximately 32%. Data of Hg concentrations collected over long periods reveals their bimodal character with the maximum values in the heating season and during summer. Elevated mercury concentrations and low pH in precipitation were more often reported during the heating season, which indicates the basic problem of coastal air pollution - the combustion of fossil fuels. In Poland, fossil fuels are the basic source of energy. During the very frosty and snowy winter of 2005/2006, when the temperature of air fell below -30°C , the concentrations of Hg(II) in precipitations were 3 times higher than during mild winters. In summer, apart from anthropogenic sources, the additional input of mercury to the coastal zone was through reemission from the coastal waters of the Baltic and the adjacent land. The reemission could occur in the periods of stronger sun radiation and higher temperatures. The input during summer downpours (height of the fall $>20 \text{ mm}$) was the highest in the year. Therefore, frosty winters and high temperatures in the summer together with intensive precipitation will intensify the variation in mercury deposition.

Apart from local emission sources, which supplied the coastal air with mercury, an important influence on the high concentration of this metal in precipitation were air masses flowing over the most polluted regions of Poland (Silesia) and other countries (Ukraine, Belarus, Germany, Czech Republic, Austria). An important parameter which also controlled the amount of Hg in precipitation was the height of the flowing air mass. The lower the air mass was flowing, the higher was the concentration of Hg in wet precipitation. This dependence was observed during every season of the year.

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Acknowledgements. Research on atmospheric mercury changes over the coastal zone of the Gulf of Gdansk was supported by the Polish Ministry of Science and Higher Education (grant 2533/T02/2007/32) and the European Union within the European Social Fund within the framework of the project “Innodoktorant–scholarship for PhD students, I edition”.

The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for providing access to the HYSPLIT transport and dispersion model and the READY website (<http://www.arl.noaa.gov/ready.html>). Special thanks to the ARMAAG Foundation for making meteorological data available.

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Table 1. Statistical characteristics of Hg(II) and THg concentrations (ng L^{-1}) in precipitations over the coastal zone in Gdynia from the end of 2005 until the beginning 2009.

	N	Mean	Median	Min – Max	Lower quartile – Upper quartile	Percentile 5.0 – Percentile 95.0	SD
Hg(II)	143	13.6	10.5	0.6–53.9	5.1–19.2	1.5–34.3	11.1
Thg	145	42.6	39.3	8.6–118.0	24.0–59.1	11.6–82.1	23.5

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Table 2. Average concentrations of Thg (ng L^{-1}) in precipitation over the southern Baltic in the years 2006–2008.

Year (N-number of events)	Marine air masses $X \pm \text{SD}/\text{Min} - \text{Max}$	Continental air masses $X \pm \text{SD}/\text{Min} - \text{Max}$
2006 ($N=37$)	41.4 ± 30.1 (9.6–118.0)	35.2 ± 23.8 (9.4–99.4)
2008 ($N=89$)	47.3 ± 20.7 (11.6–89.1)	45.8 ± 23.0 (11.9–87.8)

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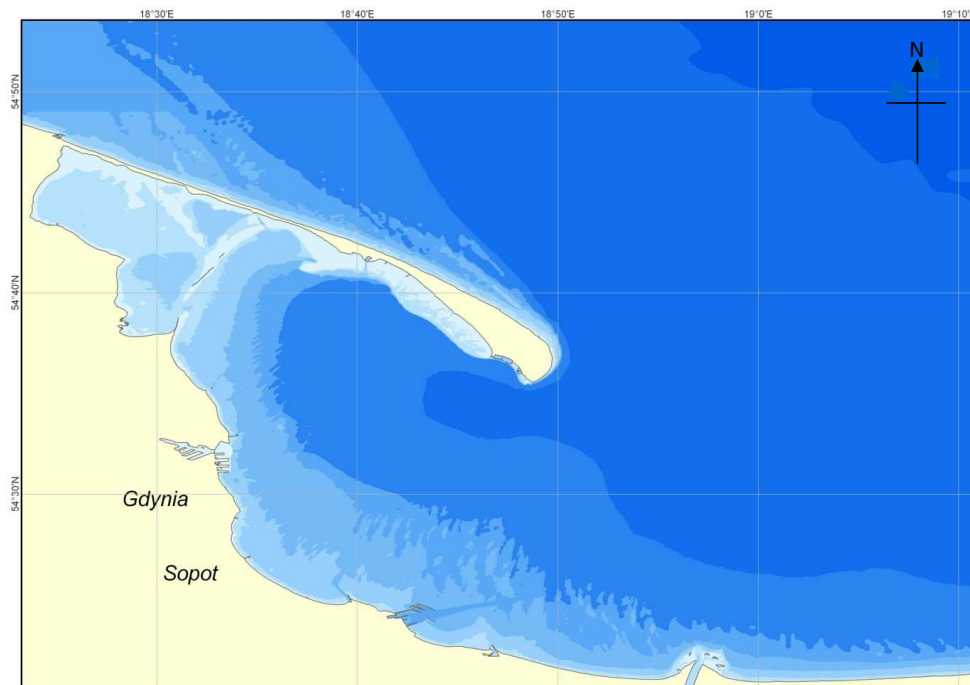


Fig. 1. The measurement site location at Gdynia ($54^{\circ}30' \text{ N}$, $18^{\circ}32' \text{ E}$).

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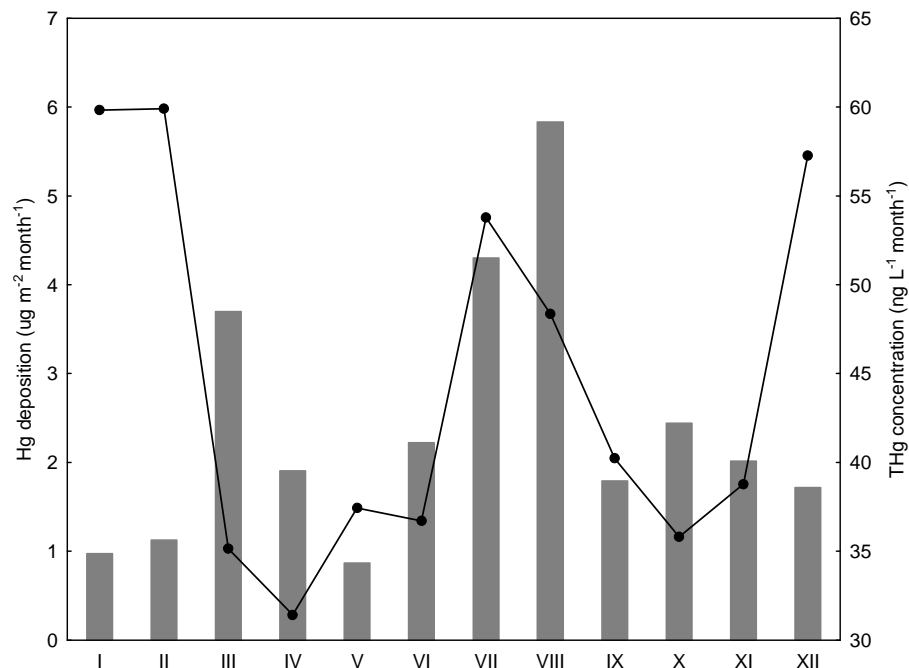


Fig. 2. Average monthly values of wet deposition and concentrations of total mercury in the coastal area of the southern Baltic (Gdynia) in the years 2005–2008 (columns – wet deposition of THg, solid line with solid circles – THg concentration).

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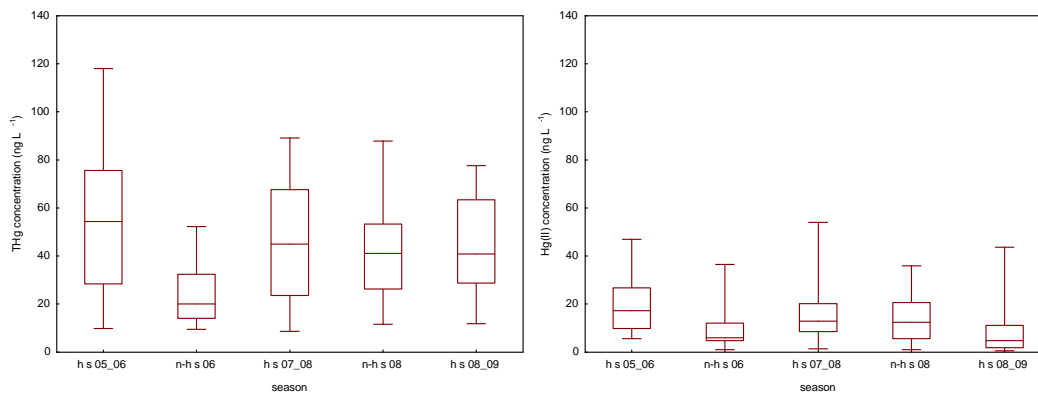


Fig. 3. Total mercury (a) and reactive mercury (b) in precipitation over the coastal zone of the southern Baltic in heating (h s) and non-heating (n-h s) seasons over the period 2005–2008.

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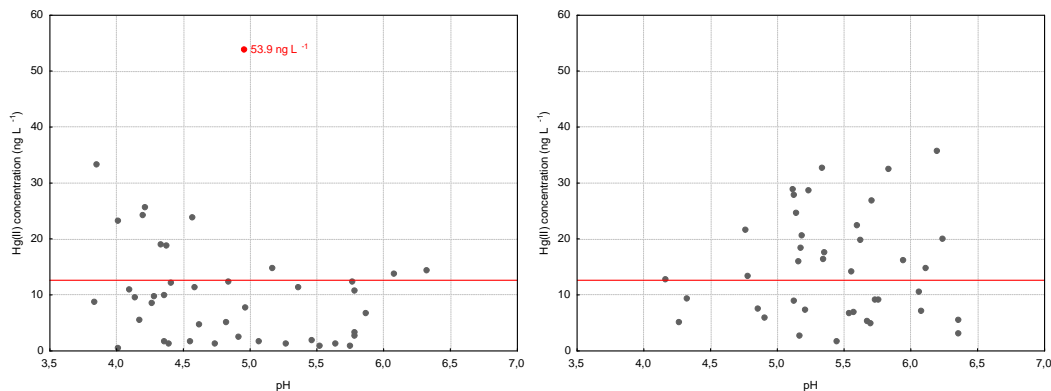


Fig. 4. The dependence of Hg(II) concentration on pH in wet precipitation over the southern Baltic during heating (left) and non-heating (right) seasons in the year 2008. (red solid line - the average concentration of Hg (II) in the year 2008: 12.6 ng L^{-1}).

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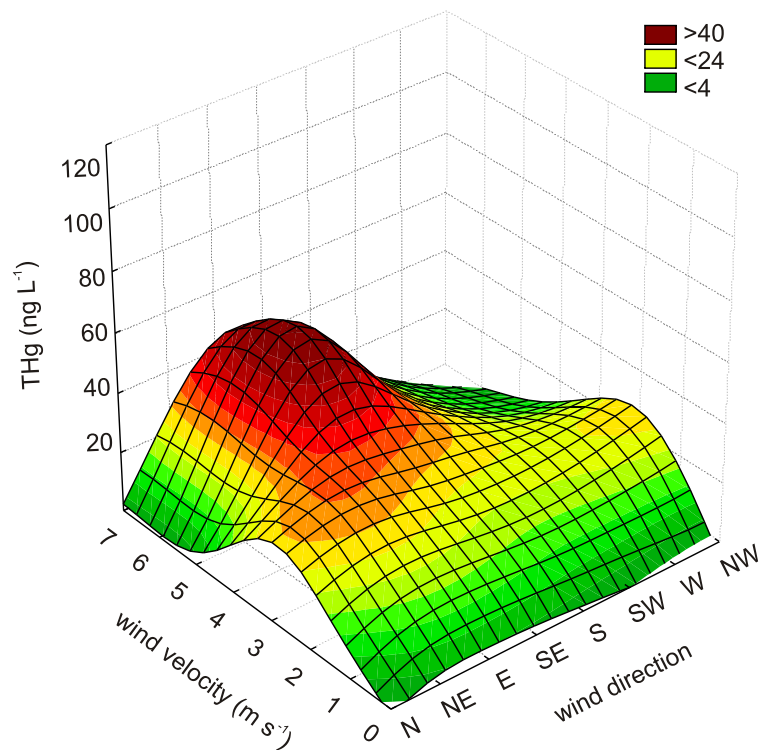


Fig. 5. The influence of speed and direction on wind on the concentration of THg in precipitation over southern Baltic.

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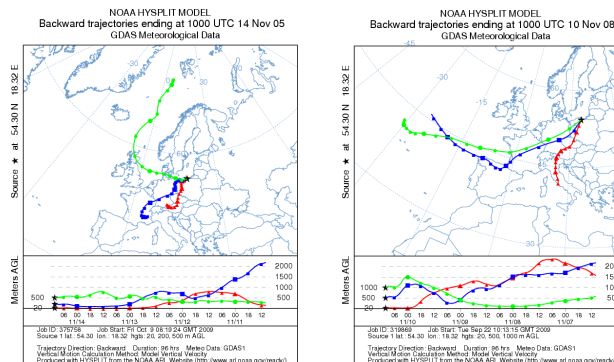
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Back trajectories for the heating season

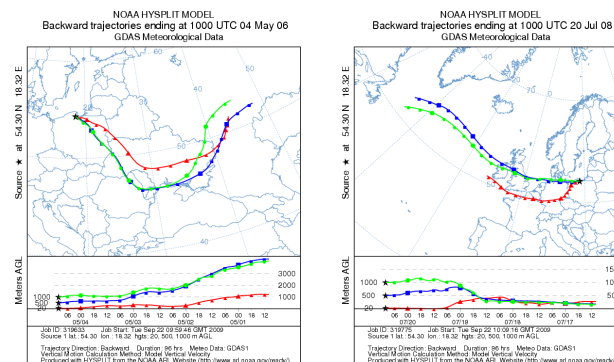


THg concentration (ng L⁻¹)

99.4

42.7

Back trajectories for the non-heating season



THg concentration (ng L⁻¹)

39.0

47.1

Fig. 6. Back trajectories (96 h) of air masses flowing to the southern Baltic.