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ATMOSPHERE AS A SOURCE OF TRACE METALS INPUT TO THE GDAŃSK BASIN *

Contents: 1. Introduction, 2. Materials and method, 3. Climatic conditions in the Gulf of Gdańsk, 4. Trace metals in the air, 5. Trace metals in precipitation and dry fallout, 6. The input of trace metals from the atmosphere into the sea; Streszczenie; References.

1. INTRODUCTION

The importance of the atmosphere as a source of trace metals for the aquatic environment is not generally appreciated. Only in recent years have papers appeared which take into account the atmosphere in the budget of trace metals [7, 22, 23, 26].

This study aimed to determine the content of cadmium, copper, mercury, lead and zinc in aerosols and fluxes of these metals in atmospheric precipitation and in dry fallout. The results helped to estimate the average annual discharges of these metals into the Gulf of Gdańsk and the Gdańsk Basin.

2. MATERIALS AND METHOD

Samples of aerosols of precipitation and dry fallout were collected at the meteorological stations in Gdynia and on the Hel peninsula, also at sea during research cruises from January 1975 to October 1976. Sampling was carried out according to the recommendations of the World Meteorological Organization [24] and the designation of the samples for the determination of trace metals. The samples of rainfall were preserved by means of nitric acid. Air and dry fallout samples collected on filters were acid digested before analyses. Flameless atomic absorption was used for all the determinations. Total mercury and its inorganic

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forms were determined by cold vapour technique [18]. The content of organic mercury compounds was obtained from the difference between the above two results. Cadmium, copper lead and zinc were determined using electrothermal atomization [5, 16, 25]. Measurements were made on a Beckman 1272 atomic absorption spectrophotometer equipped with a graphite furnace and deuterium backgroud corrector. The optimum instrument conditions were set previously for river waters [1].

The methods were calibrated with a series of inorganic standard solutions; such working solutions were diluted with rain water. So far, no reference materials are available for air and atmospheric dry fallout, therefore it was difficult to define systematic error of the method. It was also impossible to establish the accuracy of the whole analytical process covering sampling, their storage and the determinations, as both the physicochemical properties of the samples and the metal contents varied widely. The precision of spectrophotometric measurements ranged from 2 to 20% depending on the metal and the concentration. The detection limits for the Beckman 1272 atomic absorption spectrophotometer are as follows: 0,2 ng Cd, 10 ng Cu, 2 ng Hg, 5 ng Pb, and 0.2 ng Zn.

3. CLIMATIC CONDITIONS IN THE GULF OF GDAŃSK

During the research period weather conditions at the two stations were very similar (Figs. 1 and 2). Northerly and westerly winds were predominant at Hel, while at Gdynia they blew from W, NW and N [15]. The analysis of climatic conditions over the period 1951—1975 [14] showed that at both stations winds from W, NW, SW and S dominated. The only difference was due to the fact that during the research period the northerly component was predominant, while from 1951—1975, the southerly component was more frequently observed.

The average monthly wind velocities were also almost the same. The average annual wind velocity measured over many years was 4.5 $m \cdot s^{-1}$ at Hel and 4.3 $m \cdot s^{-1}$ at Gdynia [14].

There was less precipitation than normal during 1975 and 1976 which belonged to the warm years. Nevertheless, the climatic conditions at this time did not substantially deviate from the normal conditions of the research area. The geografical situation of Gdynia favours forced convection and convergence of air masses, which cause strong winds of varying circulation. Similarly strong winds, though of a different origin from those in Gdynia blow over open spaces of Hel. Considerable mixing of air masses takes place over the Gulf of Gdańsk and this is why the trace metals content in the atmosphere at Hel is similar to that at Gdynia. Both can be used to calculate the discharge of metals from the atmosphere to the sea in the coastal zone.

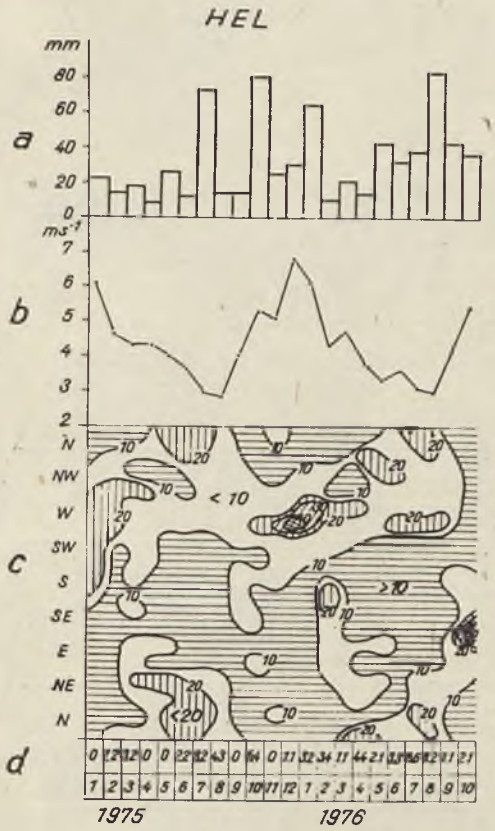

Fig. 1. Meteorological conditions at Hel: (a) total wet precipitation, (b) monthly mean wind velocity, (c) frequency (%) of wind directions, (d) frequency (%) of calm periods

Ryc. 1. Warunki meteorologiczne na Helu: suma opadów atmosferycznych (a), średnia miesięczna prędkość wiatru (b), częstość (%) występowania kierunków wiatru (c), częstość (%) występowania ciszy (d)

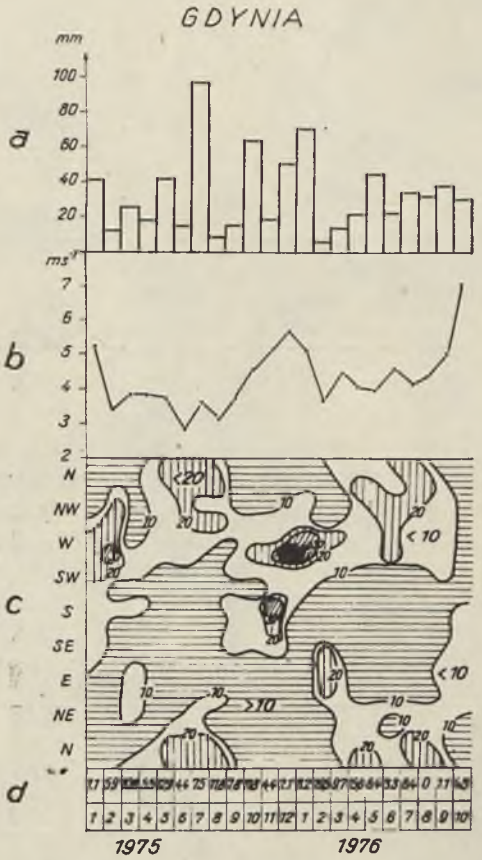

Fig. 2. Meteorological conditions at Gdynia: (a) total wet precipitation, (b) monthly mean wind velocity, (c) frequency (%) of wind directions, (d) frequency (%) of calm periods

Ryc. 2. Warunki meteorologiczne w Gdyni: suma opadów atmosferycznych (a), średnia miesięczna prędkość wiatru (b), częstość (%) występowania kierunków wiatru (c), częstość (%) występowania ciszy (d)

4. TRACE METALS IN THE AIR

The trace metal content in the air is a characteristic feature of the degree of pollution of the environment. Having compared selected data on trace metals in the air, it may be concluded that the trace metals contents in the atmosphere above the Gdańsk Basin (Table 1) are similar to those found over the North Atlantic and in nonindustrialized parts of the land (Table 2).

Table 1
Tabela 1

Trace metal contents in the atmosphere
Średnie stężenia metali śladowych w atmosferze

AIR POWIETRZE						
Sampling area Rejon poboru prób	No. of samples Nr próby	μg · m⁻³				
		Cd	Cu	Hg	Pb	Zn
Gdynia	46	0.09	0.18	0.02	1.50	0.03
Hel	41	0.06	0.04	0.02	0.87	0.02
Gdańsk Basin	21	0.07	0.17	0.01	1.41	0,02

PRECIPITATION OPADY ATMOSFERYCZNE							
Sampling area Rejon poboru prób	Sampling period Okres h	Rain Deszcz mm	μg · m⁻² h⁻¹				
			Cd	Cu	Hg	Pb	Zn
Gdynia	14 416	661	0.40	2.33	0.38	5.01	1.50
Hel	13 392	657	0.20	2.35	0.40	4.22	1.46
Gdańsk Basin	1 362	41	0.07	0.38	0.36	1.76	0.59

DRY FALLOUT SUCHY OSAD						
Sampling area Rejon poboru prób	Sampling period Okres h	μg · m⁻² h⁻¹				
		Cd	Cu	Hg	Pb	Zn
Gdynia	3 780	0.69	1.23	1.31	2.64	0.67
Hel	3 965	0.60	1.35	2.50	2.84	0.61
Gdańsk Basin	294	0.09	0.22	0.07	0.63	0.38

Lead is the exeption. Its higher concentration requires further study, primarily of a methodological character. At present increasing interest is being shown in organic lead compounds in the atmosphere. In papers dealing with this problem [5, 9], the proportion of organic lead compounds in the air is said to amount between 10 and 20% of the total lead concentration.

The aerosol component constitutes the original source of metals in the atmosphere. They are then washed out by precipitation and sorbed

Table 2
Tabela 2

Ranges and mean concentrations of trace metals in the near surface layer of the atmosphere

Porównanie zakresów i średnich stężeń metali w przyziemnej warstwie atmosfery

Area Rejon	$\mu g \cdot m^{-3}$					Source lit. Źródło bibl.
	Cd	Cu	Hg	Pb	Zn	
Gdańsk Basin	0.074	0.17	0.08	1.4	0.02	Table 1
New York Bight, wind NW-NE	1.4			0.20	0.057	[7]
New York Bight, wind NE-SE	0.052			0.044	0.032	[7]
Northern Atlantic	0.03—0.62	0.12—10		0.1—64	0—27	[6]
Gdynia	0.086	0.18	0.023	1.5	0.03	Table 1
Towns in Central Poland	0.002—0.05					[5]
Towns of the European Community						
— villages				0.5		[10]
— small towns				1		
— big cities				2		
— traffic centres				5—10		
Towns in Great Britain				0.4—7.4		[10]
Belgium			0.008			[9]
USA Tuxcon and Arizona districts	0.003	0.025		0.082—1.15	0.03	[3]
New York City	5.8		0.001—0.014	1.4	310	[7, 17]
Chicago	0.005—0.08		0.003—0.039			[9, 17]
Japan: rise fields			10	0.01—2.74		
towns			15			[9, 10]
Australia uninhabited areas				0.2—0.4		[16]
towns				0.5—9.5		

by the dry fallout. Under the climatic conditions of the Gulf of Gdańsk, where the total precipitation is quite high, metals are washed out to a very large extent. The enrichment coefficients of rain water in metals with respect to the air calculated on the basis of concentrations given in units of volume (Table 3), fall within the range $10^4 - 10^6$. In order to calculate these values for dry fallout, it is essential to know the exact mass of aerosols and dry fallout, and the height of the mixing layer. Studies taking into account the variability of this parameter have been the subject of a number of papers in which attempts have been made to estimate the rate of metal exchange between the atmosphere and the sea in the Gulf at Gdańsk [11—13].

Table 3
Tabela 3

Trace metal enrichment coefficients in rainfall versus aerosols

Współczynniki wzbogacania wody deszczowej w metale w stosunku do ich zawartości w aerozolu atmosferycznym

Area Rejon	Cd	Cu	Hg	Pb	Zn
Gdynia	$1.6 \cdot 10^5$	$2.4 \cdot 10^5$	$3.2 \cdot 10^5$	$1.2 \cdot 10^5$	$1.1 \cdot 10^5$
Hel	$6.6 \cdot 10^4$	$1.2 \cdot 10^6$	$1.3 \cdot 10^6$	$1.6 \cdot 10^5$	$1.5 \cdot 10^6$
Gdańsk Basin	$4.5 \cdot 10^4$	$1.1 \cdot 10^5$	$3.4 \cdot 10^6$	$4.9 \cdot 10^5$	$1.6 \cdot 10^6$

When calculating input of metals from the atmosphere to the sea the aerosol component has been mostly covered by rainfall and dry fallout.

5. TRACE METALS IN PRECIPITATION AND DRY FALLOUT

Systematic studies of trace metals in rainfall and dry fallout have been described in a number of papers, in which most attention was paid to cadmium [3], mercury [17], lead [4, 10], also natural and artificial radioisotopes [11, 19].

The flux of cadmium in the dry fallout over the Gulf of Gdańsk (Table 1) is approximately that found in previous years over Finland, 0.085 $\mu g \cdot m^{-2} h^{-1}$ and Sweden, 0.5—5 $\mu g \cdot m^{-2} h^{-1}$ [3].

The concentrations of mercury in rainfall at Hel and Gdynia fell within the range 0.1—2 $\mu g\ l^{-1}$, whereby the content of organic mercury compounds amounted on average to 67% of the total concentration of that metal. In Sweden the concentration of mercury in rain water was similar and come to 0:2 $\mu g\ l^{-1}$ [9] on average. The World Health Organization has given the natural concentration of mercury in rain water as from 0.05 to 0.5 $\mu g\ l^{-1}$ [9].

Lead concentrations in rainfall from the Gulf of Gdańsk area varied from 10 to 200 μg l^{-1}. For comparison the lead content in rainfall from the Geneva area ranged from 83 to 132 μg l^{-1} [4]. In Toronto the lead content in rain water was of the order of 8 μg l^{-1}, while the flux of lead in the dry fallout is from 2 to 3 orders greater [10] than that found in Gdynia and Hel (Table 1). The different proportions of lead in rainfall and dry fallout are probably the result of different climatic conditions and the traffic intensity in cities. Looking at the percentages of metals in rain water and dry fallout (Table 4) we see that in urban agglomerations cadmium and mercury appear mostly in the dry fallout, while cooper, lead and zinc are mainly found in rain water. Mercury and cadmium probably get into the atmosphere chiefly as fine dusts which may originate from the products of coal combution and from industry. Other metals, such as lead reach the atmosphere as volatile or soluble forms.

Table 4
Tabela 4

Partition (in percent) of trace metals between rainfall and dry fallout

Udział metali ciężkich w opadach deszczu i w suchym osadzie

	Cd	Cu	Hg			Pb	Zn
			total całkowita	organ.	inorg. nieorg.		
GDYNIA							
Rainfall Deszcz	36.7	65.4	22.5	17.8	25	65.5	69.1
Dry fallout Osad suchy	63.3	34.6	77.5	82.2	75	34.5	30.9
HEL							
Rainfall Deszcz	24.7	63.7	13.9	12.6	17.6	51.3	70.5
Dry fallout Osad suchy	75.3	36.5	86.1	87.4	86.4	48.7	29.5
GDAŃSK BASIN							
Rainfall Deszcz	45.7	63.3	83.9	89.5	53.1	73.6	60.8
Dry fallout Osad suchy	54.3	36.7	16.1	10.5	47.9	26.4	39.2

Over the sea the participation of cadmium and lead increases somewhat, while the percentage of organic mercury compounds in rain water increases considerably (Table 4). In the low atmospheric layers above the sea the metals are derived from the land and as a result of removal from the water. The source of organic mercury compounds in the atmosphere is likely to be the methylation of inorganic mercury by bacteria, which takes place to a large degree in the top layer of the bottom sediments. An estimation of the removal of metals from the

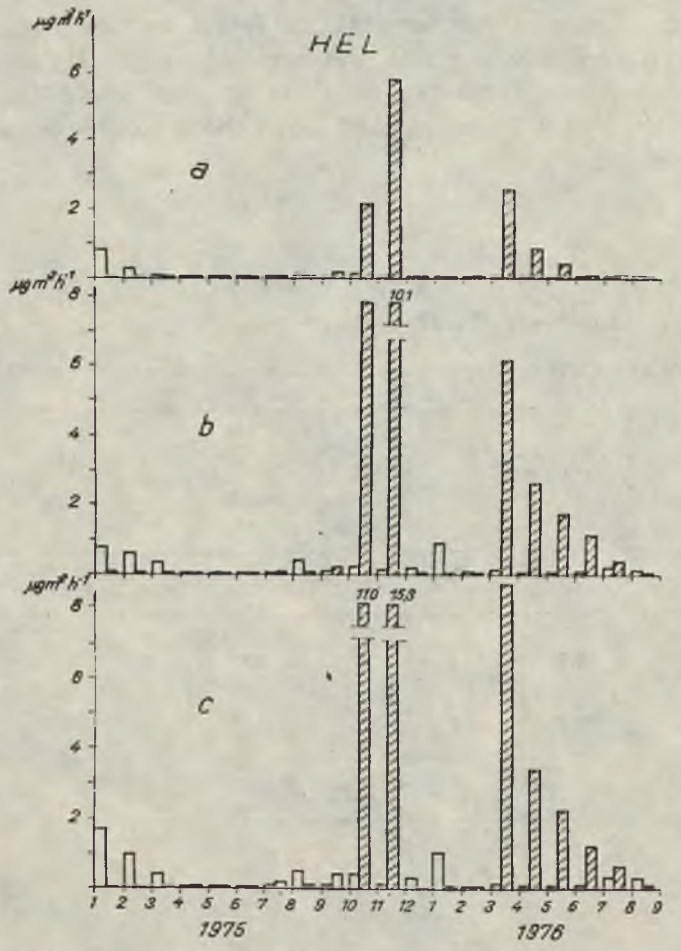

Fig. 3. Mean fluxes of mercury with rainfall (open columns) and dry fallout (dashed columns) at Hel: (a) inorganic mercury, (b) organic mercury compounds, (c) total mercury

Ryc. 3. Średnie strumienie rtęci w wodzie deszczowej (słupki puste) i w suchym osadzie atmosferycznym (słupki zakreskowane) w Helu: rtęć nieorganiczna (a), organiczne związki rtęci (b), rtęć całkowita (c)

sea to the atmosphere can be made knowing the metal concentrations in the surface microlayer of sea water, where there is a tendency for metals to accumulate [8, 20].

The greatest concentrations of mercury lead and cadmium in the atmosphere at Hel and Gdynia occurred in the winter and early spring (Figs. 3—6). This probably due to the use of coal as a fuel. The increase of lead and cadmium concentrations at Hel and in the low atmospheric layer, which was observed at Hel and over the Gulf of Gdańsk in the summer of 1975, may in part have been caused by a local horizontal exchange connected with the development of sea breeze circulation during this period [12, 13].

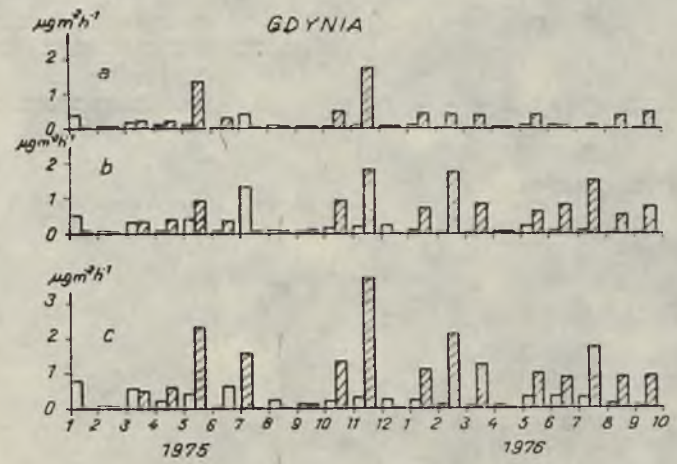

Fig. 4. Mean fluxes of mercury with rainfall (open columns) and dry fallout (dashed columns) at Gdynia: (a) inorganic mercury, (b) organic mercury compounds, (c) total mercury

Ryc. 4. Średnie strumienie rtęci w wodzie deszczowej (słupki puste) i w suchym osadzie atmosferycznym (słupki zakreskowane) w Gdyni: rtęć nieorganiczna (a), organiczne związki rtęci (b), rtęć całkowita (c)

The highest concentrations of copper in rainfall at both stations occurred in summer. In increase in zinc content in rainfall was observed at Hel and Gdynia in the autumn and winter of 1975 and at Hel during the summer of 1976 (Figs. 7 and 8).

6. THE INPUT OF TRACE METALS FROM THE ATMOSPHERE INTO THE SEA

On the basis of determinations of trace metals in rainfall and dry fallout, the potential annual discharges of cadmium, copper, mercury, lead and zinc from the atmosphere to the Gulf of Gdańsk and the Gdańsk

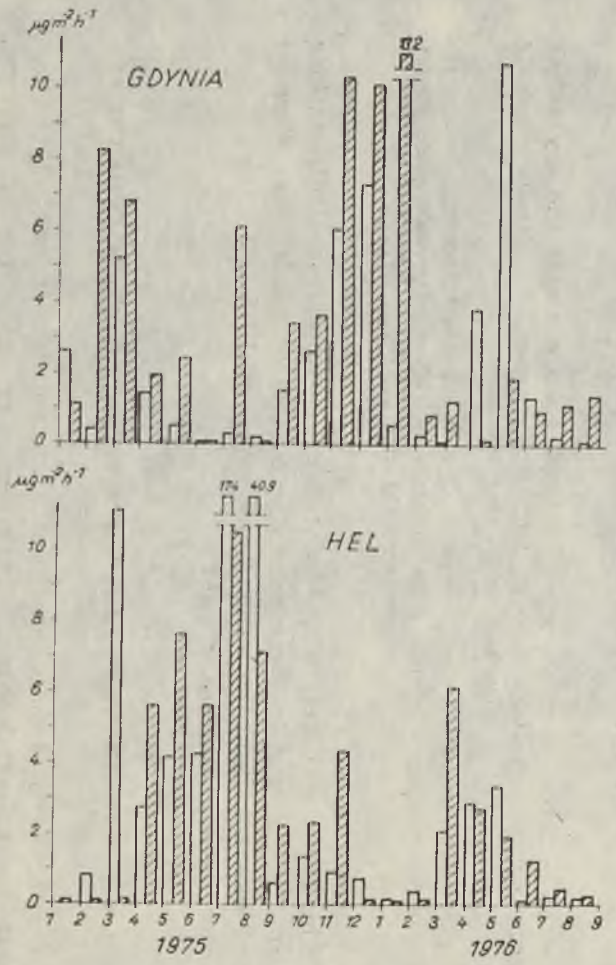

Fig. 5. Mean fluxes of lead with rainfall (open columns) and dry fallout (dashed columns) at Hel and Gdynia

Ryc. 5. Średnie strumienie ołowiu w wodzie deszczowej (słupki puste) i w suchym osadzie atmosferycznym (słupki zakreskowane) na Helu i w Gdyni

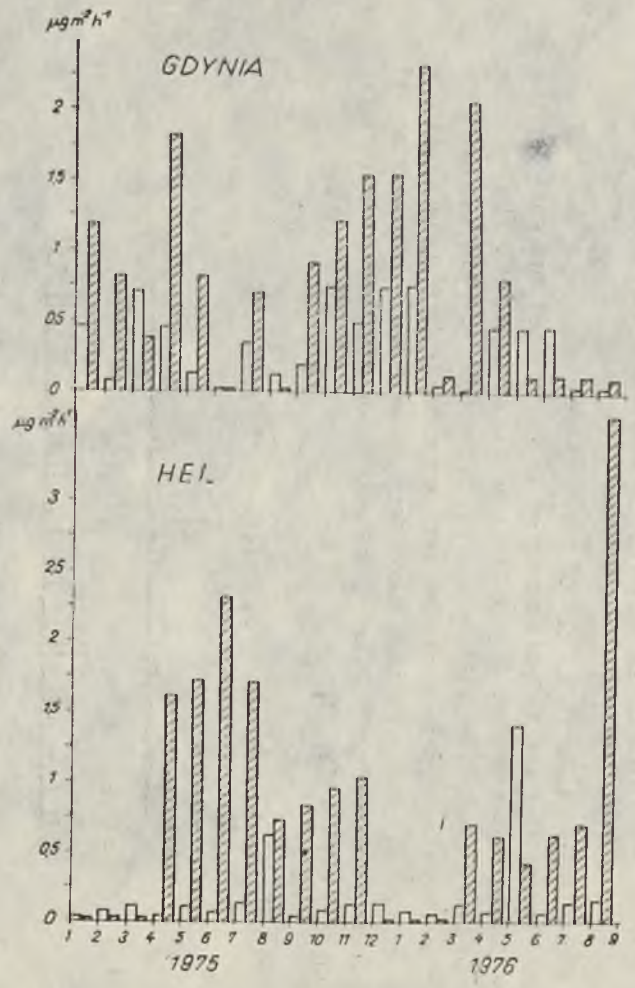

Fig. 6. Mean fluxes of cadmium with rainfall (open columns) and dry fallout (dashed columns) at Hel and Gdynia

Ryc. 6. Średnie strumienie kadmu w wodzie deszczowej (słupki puste) i w suchym osadzie atmosferycznym (słupki zakreskowane) na Helu i w Gdyni

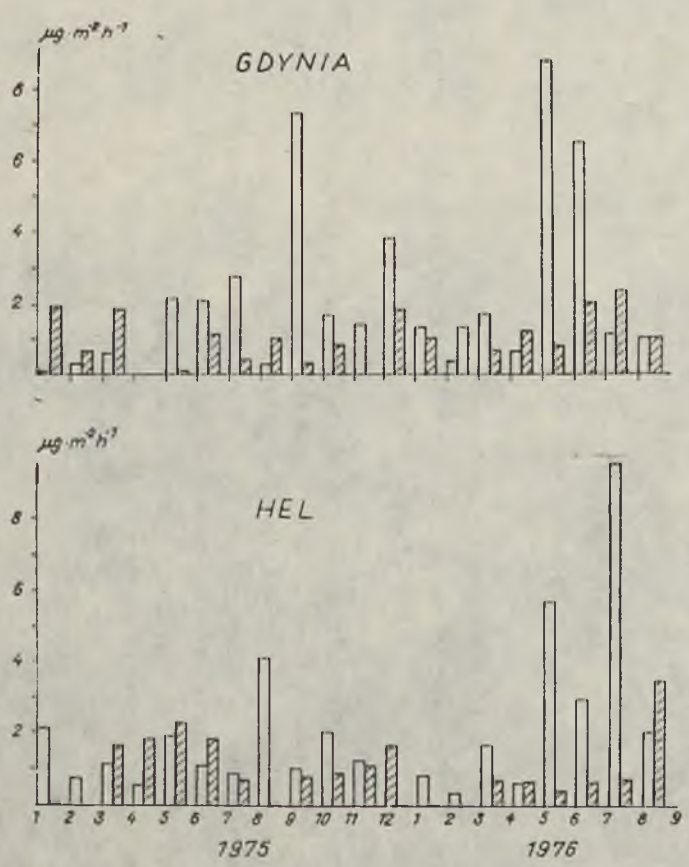

Fig. 7. Mean fluxes of copper with rainfall (open columns) and dry fallout (dashed columns) at Hel and Gdynia

Ryc. 7. Średnie strumienie miedzi w wodzie deszczowej (słupki puste) i w suchym osadzie atmosferycznym (słupki zakreskowane) na Helu i w Gdyni

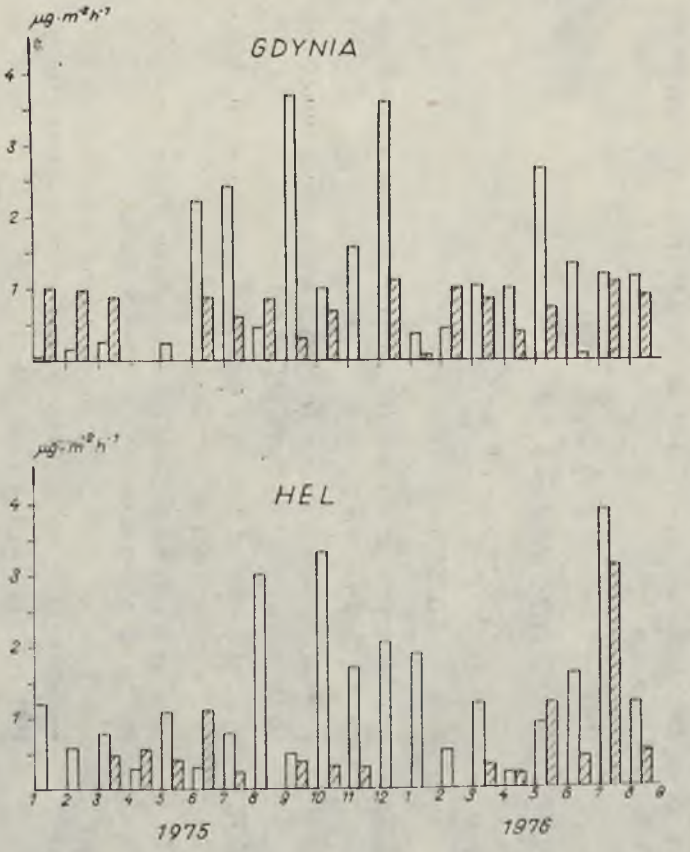

Fig. 8. Mean fluxes of zinc with rainfall (open columns) and dry fallout (dashed columns) at Hel and Gdynia

Ryc. 8. Średnie strumienie cynku w wodzie deszczowej (słupki zakreskowane) na Helu i w Gdyni

Basin have been calculated. The division of these areas as suggested by Trzosińska [21] was applied:

$$A_IBG = A_IZGW + A_IZGZ + A_IPZG$$

$$A_IZGW = c \cdot F_1S_{ZGW}$$

$$A_IZGZ = c \cdot F_2S_{ZGZ}$$

$$A_IPZG = c \cdot F_2S_{PZG}$$

where: A_IBG — annual input of metals (in tons) from the atmosphere to the Gdańsk Basin,

A_IZGW — annual input of metals (in tons) from the atmosphere to the inner part of the Gulf of Gdańsk,

A_IZGZ — annual input of metals (in tons) from the atmosphere to the outer part of the Gulf, of Gdańsk,

A_IPZG — annual input of metals (in tons) from the atmosphere to the open sea area of the Gdańsk Basin,

F_1 — the flux of metals in precipitation and dry fallout equal to the arithmetic mean of measurements at Hel and Gdynia ($\mu g \cdot m^{-2} \cdot h^{-1}$),

F_2 — flux of metals in precipitation and dry fallout equal to the arithmetic mean of measurments made at sea ($\mu g \cdot m^{-2} \cdot h^{-1}$),

S_{ZGW}, S_{ZGZ}, S_{PZG} — surface areas (km^2) of the inner (ZGW), and outer (ZGZ) parts of the Gulf of Gdańsk and open sea area of the Gdańsk Basin (PZG),

c — constant ($8.76\ 10^{-3}$) resulting from the calculation of units.

The comparison of atmospheric discharges should be treated only as estimates, as the time of sampling, their frequency, the distribution of sampling stations and climatic conditions, are of prime importance in such studies.

The results of calculations (Table 5) indicate that the atmosphere is a very serious source of metal input into the sea. As far as cadmium, mercury and lead are concerned, the discharge from the atmosphere to the Gulf of Gdańsk is greater than the inflow from the Vistula River (Table 6). A comparison of inputs from the atmosphere and the land into the Gulf of Gdańsk and Lake Michigan indicates similar percentages of the atmospheric input of copper in the total discharge of this metal. The small percentage of metal inputs from the atmosphere into the New York Bight is explained by the very large sewage discharge into the sea from the heavily industrialized agglomerations of New York City and Long Island [7].

Table 5
Tabela 5

Inputs of trace metals from the atmosphere to the sea
Zrzuty metali śladowych z atmosfery do morza

Area Rejon	Surface Powierzchnia km²	Atmospheric component Składowa atmosf.	tons.year⁻¹					Source lit. Źródło bibl.
			Cd	Cu	Hg	Pb	Zn	
Gulf of Gdańsk	$5 \cdot 10^8$	rainfall	6.4	46.5	15.2	117	38.2	Table 1
		dry fallout	12.6	25.9	31.8	59	19.6	
		total	19	72.4	47	176	57.8	
Gdańsk Basin	$1.8 \cdot 10^4$	rainfall	15	90.9	20	322	107	Table 1
		dry fallout	23	51.6	40	133	64	
		total	38	142.5	60	455	171	
North Sea	$5.8 \cdot 10^5$	total			30	1000		[22]
New York Bight	10^4	rainfall	8.2			400	540	[7]
		dry fallout	4.1			200	270	
		total	12.3			600	810	
Southern California Bight	10^4	rainfall				17		[7, 26]
		dry fallout				240		
		total			38	257		
Basin of Lake Michigan	$1.7 \cdot 10^6$	total		3200			3900	[23]

Table 6
Tabela 6

Comparision of trace metal inputs from the atmosphere and from the land
Porównanie zrzutów z atmosfery i z lądu

Area Rejon	Percentage of atmosferic input in total input Udział: atmosfera/ /całkowity zrzut					Ratio of inputs: atmosphere/land Udział: atmosfera/ląd					Source lit. Źródło bibl.
	Cd	Cu	Hg	Pb	Zn	Cd	Cu	Hg	Pb	Zn	
Gulf of Gdańsk	65	31	61	79	20	1.8	0.3	1.6	3.8	0.2	Table 5 [1]
New York Bight	15			13	8	0.01			0.15	0.09	[7]
Lake Michigan		45			87		0.8			6.7	[23]

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ATMOSFERA JAKO ŹRÓDŁO DOPŁYWU METALI ŚLADOWYCH DO BASENU GDAŃSKIEGO

Streszczenie

W pracy przedstawiono wyniki oznaczeń kadmu, miedzi, rtęci, ołowiu i cynku w aerozolach, wodzie deszczowej i suchym osadzie atmosferycznym. Próby pobierano na stacjach meteorologicznych na Helu i w Gdyni oraz na obszarze Basenu Gdańskiego w czasie rejsów badawczych w latach 1975—1976. Metody poboru prób opracowano zgodnie z zaleceniami WMO i ich przeznaczeniem do oznaczeń metali śladowych.

Rtęć całkowitą i jej formy nieorganiczne oznaczano techniką zimnych par. Zawartość organicznych związków rtęci otrzymano z odejmowania powyższych wartości. W suchym osadzie i w opadach atmosferycznych wynosiła ona średnio 67% całkowitego strumienia rtęci.

Pozostałe metale oznaczano metodą absorpcyjnej spektrometrii atomowej z zastosowaniem atomizacji elektrotermicznej. Wszystkie pomiary wykonywano na spektrofotometrze atomowo-absorpcyjnym Beckman 1272.

Stężenia metali śladowych w aerozolach w przyziemnej warstwie atmosfery (0,075 μg Cd/m³, 0,12 μg Cu/m³, 0,034 μg Hg/m³, 1,2 μg Pb/m³ i 0,024 μg Zn/m³) okazały się zbliżone do wartości znajdowanych nad innymi obszarami wodnymi i słabo uprzemysłowionymi obszarami lądowymi.

W aglomeracjach miejskich średnie strumienie metali przenoszonych z wodą deszczową były następujące: dla Cd — 0,30, Cu — 2,34, Hg — 0,39, Pb — 4,61, Zn — 1,48 $\mu g \cdot m^{-2} \cdot h^{-1}$; na morzu wartości te wynosiły dla Cd — 0,07, Cu — 0,38, Hg — 0,36, Pb — 1,76 i Zn — 0,59 $\mu g \cdot m^{-2} \cdot h^{-1}$.

Średnie strumienie metali w suchym osadzie na Helu i w Gdyni odpowiadały wartościom dla Cd — 0,65, Cu — 1,19, Hg — 1,9, Pb — 2,74, Zn — 0,64 $\mu g \cdot m^{-2} \cdot h^{-1}$; na obszarze Basenu Gdańskiego: dla Cd — 0,09, Cu — 0,22, Hg — 0,07, Pb — 0,63 i Zn — 0,38 $\mu g \cdot m^{-2} \cdot h^{-1}$.

W okresie pomiarów warunki meteorologiczne na Helu i w Gdyni były bardzo zbliżone i typowe dla obszaru Basenu Gdańskiego, co pozwoliło na wykorzystanie danych z obydwu stacji do obliczeń potencjalnego zrzutu metali do morza w strefie brzegowej. Średnie roczne dopływy metali na obszar Basenu Gdańskiego kształtowały się następująco: 38 ton Cd, 142,5 tony Cu, 60 ton Hg, 455 ton Pb i 171 ton Zn.

Stwierdzono, że dla Zatoki Gdańskiej atmosfera jest większym źródłem dopływu kadmu (1,8 raza), rtęci (1,6 raza) i ołowiu (3,8 raza) niż wody Wisły. Spływ rzeczny jest natomiast znacznie większym źródłem dopływu miedzi i cynku do Basenu Gdańskiego niż atmosfera.

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