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THE MECHANISMS OF AIRBORNE PARTICULATE TRANSPORT TO SEA VIA ATMOSPHERE TAKING BALTIC FOR EXAMPLE *

C o n t e n t s: 1. Introduction, 2. Structural transport model, 3. Experimental data concerning the operation of the particulate mechanisms of transport; Streszczenie; References.

1. Introduction

One can expect that none of the changes in the atmospheric air composition, originated by human activities, will remain without an effect on the substances present in sea waters. For certain substances of industrial origin the mechanism of their transport form continent to the sea via atmosphere may play the predominant part in the ocean sedimentation cycle. Thus, e.g., according to E.D. Goldberg [20] the share of atmospheric component in the transference of oil products from continent into the sea is 90×10^{12} g per year, whereas the direct oil pollution sources of the sea, connected with the seaborne transports of oil cargoes and industrial waste, conjointly contribute by merely 2.1×10^{12} g annually. Even though part of those gases may undergo oxygenation while in the atmosphere, still their substantial amounts — by Author's suggestion — are transferred into the sea with the precipitations as well as due to the sorption of aerozol particulate matter over the air-sea interface. There is a convincing proof of the share of atmosphere in the transport of heavy metals into the sea, in the form of results obtained on the examples of strontium-90 and cadmium [23, 33]. The atmospheric transport of these metals to the Baltic and the North Sea respectively recently exceeded the amounts contributed by river discharges nearly by four times. Further, the important rôle of atmosphere in the transportation of heavy metals and other toxic substances is also confirmed by the findings assessed by a number of other authors during their investigations [10, 27].

Considering the disturbances in quasi-stationary stability due to the progressing changes in atmospheric air composition, attention was drawn to the mechanism of gas exchange between sea and atmosphere, even at

an earlier date. Studies were concerned, in particular, with the carbon
dioxide exchange [3, 12, 24]. Apart from gases, much attention was also
devoted to the transference of aerosol particles [7, 22, 23, 35]. In addition,
there was a rising interest in the micro-scale mechanism of exchange,
connected with the direct atmosphere-sea interaction [4, 16, 28].

All the same, the sea water area presents a serious problem from the
point of view of the full budget of the toxic substance exchange, taking
place there. None of the components of that budget can be neglected, even
though apparently insignificant. The matter at stake are the effects
connected with the cummulation of the particular constituents in time and
their influence on the long-term loss of quasi-stationary equilibrium.
When thus approaching the problem, one must also take into consider-
ation the complexity of aerodynamical composition of the atmosphere
itself, being the medium responsible for transportation of toxic substances
from the continent into the sea. We actually miss a satisfactory knowledge
of phenomena in this aspect so far. At the same time, the lack of a
more complete recognition is perceived with respect to the concentration
dynamics of the various toxic substances at various levels in the atmos-
phere. Additionally, this state is affected by the lack of data on the output
capacity and layout in space of air pollution sources. In general, the effect
of insufficient knowledge of the complex mechanism of the dispersed
phase transportation from above the continent to the sea, in both the lower
and upper tropospheric scale, must be deemed negative. Continuous meas-
urements of microelement concentrations must be carried out at various
levels in the atmosphere, in dry fallout and atmospheric precipitations
above the sea to explain this mechanism. Besides, the possibility to com-
pare these values against the concentrations of identical elements, simul-
taneously to be measured in sea water of the investigated area — especially
in its surface film — may be of essential importance. It is absolutely
necessary to have such measurement data material to hand, if we are
to understand the mechanism of particulate matter transport through the
air-sea interface. Again we can examine the airborne transfer from the
pollution sources to the water area with regard to the macro- and me-
zoturbulent structure of the atmosphere.

2. Structural Transport Model

It is proposed to review here, in general, the structural model (Fig.
1) explaining the importace of the particular constituents of transport
of the toxic substances passing from the atmosphere into the sea. The
model shall be dealt with exclusively from the view-point of the dynamics
of the airborne particle transport in the direction normal to the sea sur-
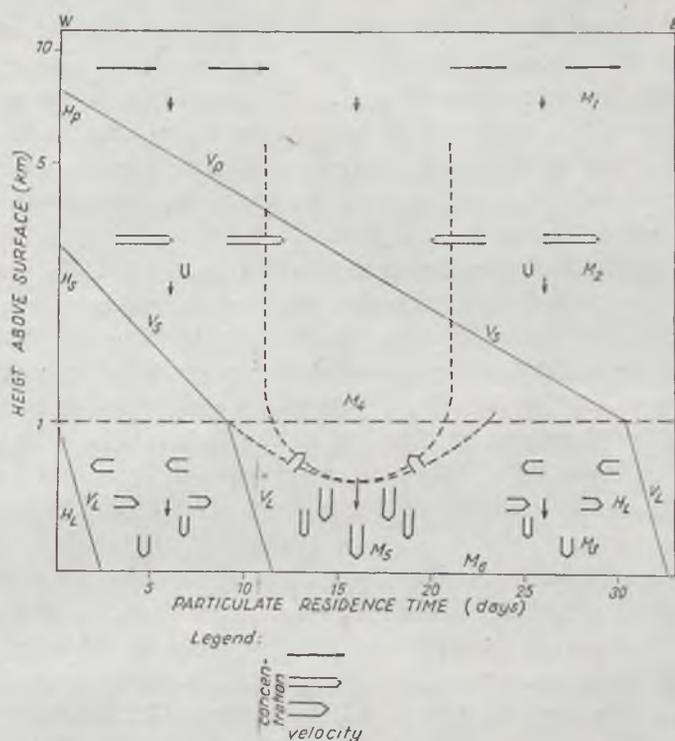
Fig. 1. Conceptual framework of the pollutants transport
air-to-sea mechanisms acting in the troposphere

Ryc. 1. Koncepcyjny schemat troposferycznych mechaniz-
mów przenoszenia zanieczyszczeń do morza

face. Let us assume here that the physico-chemical changes, if any, of the particle populations in the atmosphere will not substantially affect the mechanism of transport itself. This model will result even from the very circulation pattern of the atmosphere, since the zones of circulation, connected with the general atmospheric circulation, cannot remain without effect on the spreading of pollutants of industrial origin. The transport occurs between the surface of emission and a closing layer, i.e. the lower bound of thermal inversion. As for the processes occurring in the tropospheric scale, it is the tropopause that forms such closing layer. The structure of the mixing processes in general scale of each hemisphere is determined by the height and quasi-stationary character of the tropopause while the energy comes from the motions of smaller scale. Together with this there exists a connection between the spatial scale of the mixing processes and the scale of time.

First of all, then, we assume the action of the mechanism of admixtures spreading on a planetary scale (M_1). This mechanism is generally typical

for its zonal direction of transference, affecting the manner of substance spreading in the atmosphere, mainly in the upper tropospheric layers and perhaps in the lower stratosphere. The geostrophic movement causes the horizontal transfer component to be the dominant one and to take the westerly direction in temperate climatic zones. The substances transported at that level can travel around the globe, without significant falling out into the lower tropospheric layers.

The mechanism of transference, marked by symbol M_2, reaches much lesser altitudes that does mechanism M_1 and it reaches into the lower tropospheric layers only. Upon the whole, the character of transference is retained and the occurring disturbances do not change the general character of transport. Mechanism M_2 is typical for its spreading range, usually restricted to the given synoptic region, or else it is related with the natural synoptic circulation type. The concentrations of transported admixtures at that level should depend on the output of pollution sources present in the given region.

The boundary layer of the atmosphere, typical for its most differentiated turbulent structure, deserves special mention. Right now much attention is paid to it in literature on the subject [1, 8, 11], especially when determining the coefficients of heat and mass transport from the point of view of pollutants spreading, as does, for instance, G.T. Csanady [8] for the Ekman layer. We in our schematic approach, should like above all to draw attention to the importance of the very scale of processes. The processes of spreading in this layer predominantly should be in the nature of the mechanism of local-scale transport (M_3). Actually the transport extent in horizontal directions resembles much more closely the vertical turbulent mixing scale than it does in the cases of M_1 and M_2. The lesser scale of turbulent spreading in comparison with M_1 and M_2 — while assuming a direct effect of the emission sources on the air composition — causes the presence of highest concentrations in that layer. The considerable share of coarse-grained fractions also does not remain without effect on the magnitude of the particulate matter transport on the sea surface.

If we mark by symbols H_p, H_s, and H_L the thickness of atmospheric layers covered by the planetary, the synoptic and the local-transport scale respectively, the assessment is that H_p H_s H_L is limited by the height of near gronud inversions. Usually H_L height will oscillate between several hundreds and about one thousad metres on an average. It appears reasonable in this context that if we assume the level of non-divergency to occur at a height of about 600 mb, we also adopt it as the upper level of mechanism M_2 operating. Obviously, the tropopause level should determine the H_p height.

The average size of transported particles and their residence time in

the atmosphere, (τ) will depend on the height of levels covered by the transport mechanism. Fig. 1 presents the curves corresponding to the approximate mean values of τ for the three atmospheric levels covered by the action of mechanisms M_1, M_2 and M_3 respectively. The mean value for τ_3 for the ultimate turbulent layer (Fig. 3) was adopted from the conclusions following from theoretical analysis [3], as well as from own investigations carried in the Baltic Sea [16, 17]. When determining τ_1 and τ_2 for the upper and lower tropospheric layers respectively, covered by the action of M_1 and M_2 mechanisms, it was possible to base them on the source literature data. E.g., according to W.M. Burton and N.G. Steward [5] the τ value, determined from lead-210 to polon-210 ratio, is about 40 days, for a 9 km level. We obtained a more or less identical value for τ_1, assuming the velocity of particle fallout at H_p and H_s heights to be $v_p \simeq v_s$ respectively and to constitute half of the velocity v_L which is approximate for H_L height.

Apart from the action of M_1, M_2 and M_3 mechanism, there are two other mechanisms present in the atmosphere, connected with the cloud formation and precipitation-forming processes. The first of them (M_4) is a rainout mechanism of admixtures connected with the nucleation and condensation processes of water vapour in clouds. In this mechanism decisive are the microphysical processes connected with the type and composition of clouds. At the same time an interaction between M_1 and M_2 as well as M_3 and M_4 mechanisms takes place. The share of M_1, M_2 and M_3 in the action M_4 in this instance will depend on the cloud development scale of the vertical cloud distribution. When their vertical development scale is large enough, all the above mentioned transport types may undergo the rainout process. The characteristic scale of spreading in the horizontal plane in such case is restricted mainly to the synoptic region in question; this also refers to the share of M_1 component, retransformed into M_4. The efficiency of M_4 however, is connected with the amount of the precipitations. During their fallout there is yet another mechanism active, that of washout, designated by symbol M_5 in Fig. 1. The action of the latter however, is merely local in its extent, due to the thickness of the air layer covered by this process. Apart from the amount of precipitations, their type is also of importance in this respect.

The rôle of mechanism M_4 is in a way similar to that of atmospheric inversions. This is a cathegory of systems which cummulate pollutants and constitute sui generis secondary voluminous sources in the atmosphere.

Inasmuch as inversions are concerned, the level of admixture cummulation can take place depending on the direction of their vertical transport and it will be found both below and above the inversion. The type

and dispersive composition of the investigated fraction spreading in the atmosphere is also of some importance. Mechanism M_4 normally exceeds the inversions with respect to its cummulative effectivity, especially when its action is of a large vertical span. Due to the advantageous composition of convection currents in such systems, the extent of the action of mechanism M_4 becoms largely extended, reaching considerable heigts and at the same time also widely including the of convergency around the cloud base. In the model proposed in Fig. 1 the typical average thicknesses of the atmospheric layers covered by the action of mechanism M_4 are roughly taken into account, at the occurrence of vertically well developed clouds as well as the low clouds. On the other hand, τ_{4+5}, the duration time of rainout and washout processes is disregarded. This time may vary for M_4 and M_5 mechanisms and it will oscillate within the range of $\tau_2 \geqslant \tau_{4+5} \leqslant \tau_3$.

The conclusion drawn from the model under review appears to be that gradually as we pass from the higher-scale mechanisms to local-scale hydrodynamical processes, the importance of the vertical transport component tends to increasse. Ultimately, at the lowest atmospheric layer, in the sublayer spread close to the ai-sea interface, we find mechanism (M_6) of the small-scale transport processes present there, which is directly responsible for the exchange of airborne particulate matter to be transfered to the sea surface. This is a very complicated mechanism, which in its aspect of aeroolic mass exchange has recently drawn particular attention from many authors [2, 9, 16, 18, 34].

The problem pursuant to the idea of conjoint action of a number of separate mechanisms of substance transport in the air-sea system is the quantitative assessment of share of the particular components of this complex structure. The lack of homogeneity in the aerodynamical structure, especially when high wind gradients are present both vertically and horizontally, renders the prediction of particle movements considerably difficult. The difficulties encountered in this respect were already discussed in literature [29]. At the present stage of researches an exact determination of magnitude of the particular components is hardly probable. Still, a general recognition of the problem is quite possible. An important prerequisite to gain such a recognition is the availability of data on the distribution in space and output of sources.

Value F_0 — of the total flux of toxic admixtures directed towards the sea in its first approximation can be described in the form of a system of equations of the following type

$$\overline{F_0} = \sum_i <q_i> H_i \tau_i^{-1} \tag{1}$$

where q_i — concentration and τ_i — residence time in the atmosphere of transported substances for the characteristic height of M_i. Index i in this

formula refers to a successive transport mechanism. Considering the non-stationary character of the concentration distribution, one should correspondingly rely on equation [19]:

$$\overline{F_o} = \sum_i H_i \left(\Lambda_i <q_i> - K_{ii} \frac{\partial^2 <q_i>}{\partial z^2} \right) \qquad (2)$$

where $\Lambda_i = \frac{1}{\tau_i}$ and K_{ii} — atmospheric eddy diffusivity for the axis of i. Practically however, when the processes are averaged beyond longer lapses of time, one can rely on equation (1) as well.

3. Experimental Data Concerning the Operation of the Particular Mechanisms of Transport

Mechanism M_1 covers by its action the atmospheric layers, in which least concentrations of industrial substances usually occur. Also one can assume that the horizontal dispersion of admixtures is homogeneous there. The action of mechanism M_1 is best proven by the findings into the spreading of artificial radioactive polutants entered into the higher atmospheric layers during the tests with nuclear and thermonuclear weapons [21, 26, 32]. An analysis of experimental data material confirms the zonal character of transport paths of radioactive aerosols in the lower stratospehere and in the upper and medium tropospheric layers. Moreover, those paths run around the whole of the globe, an assertion made when identifying the reappearence of radioactive clouds above the observation regions. Such a scale of substance spreading to a certain degree also pertains to the industrial admixtures entering the atmosphere at the near ground atmospheric level. An example in this respect is furnished by the DDT, now generally present on our globe [36], or other stable substances manufactured by present day industry.

Therefore the planetary extent of dispersion, typical for M_1, explains the general background of pollution making its appearance. In spite of low concentrations, this background is likewise a magnitude that may not be disregarded in the budget of exchange of the components of various substances in the sea areas particularly when they happen to be a semi-closed basin, as in the case of the Baltic Sea. Ample proof is furnished by the already mentioned investigation results on the airborne transport of strontium-90 into the Baltic [23]. In 1970 the concentration of this isotope in the Baltic waters was 0.7 pc. l^{-1} on an average, and according to the summations made, the total fluvial discharge of this isotope into the Baltic maximally accounted for about 0.2 pc. l^{-1}. Hence the data represent a process of uniformly distributed fallout of old dusts from the upper tropospheric layers, derived from the dusts stored in the lower stratosphere.

This is an example of the action of mechanism M_1. Now this component due to the low concentrations of transported particles typical for this scale of dispersion, as often as not will be detected only with the use of most sensitive methods based on radioactive tracers. Still, its share in the budget — even when ludging by the quoted data — can be quite considerable in its summaric effect.

If we in turn consider mechanism M_2, its existence is also proved by numerous observation findings. For instance, it is interesting to note the assessment [30] of highest concentrations of pesticides in the equatorial zone of the Atlantic, where air masses are transported from the industrial districts located north. Similarly, satelite observations furnish us with materials confirming the spreading of smogs from above the industrialized areas, with the propagating systems of meteorological Highs.

Another proof of the admixtures spreading along the northern path of the Atlantic-European synoptic region is furnished by the investigations concerning the sea salt nuclei above the Baltic [13, 14, 17]. An analysis of experimental data actually resulted in the conclusion that in the composition of sea-origin condensation nuclei above the Baltic there is yet another — the ocean aerosol component, apart from the local one. The share of the Atlantic component in the collections of transported airborne particulate matter is confirmed by a number of statistical and physical characteristics, including the dispersive composition of particles, the short-term oscillations of their concentrations in the near-water atmospheric layer, (Fig. 2), residence time, size of mother droplets and others. In spite of the relatively large distances of transport, the dominant concentration of this advection component in the Baltic Sea can prove both the output capacity of the coast source regions and the efficiency and range of action of mechanism M_2 itself. The findings based on this natural index suggest an obvious conclusion also with respect to the share of toxic substances being injected into the atmosphere by the West-European industries — in the total budget of air pollutants above the Baltic area. The westerly and south -westerly direction of air mass advection, which is dominant above the Baltic, favours even such a transfer of particles.

An example of the local mechanism calling attention in our investigation results from the oscillations in lead concentrations in the air above the Gulf of Gdańsk. The problem of lead transport from air into the sea water has not yet been sufficiently explained. R.S. Cambray et al. [6] have investigated the magnitude of lead fallout from the air into the North Sea. This fallout during the 1972—1973 period varied between 0.76 and 2.4 g cm^{-2} annually in dry fallout and it was 1.65 to 4.9 g cm^{-2} annually in precipitations. Ch.S. Martens et al. [27] investigated the oscillations in lead concentration above the Gulf waters in the San Francisco region,
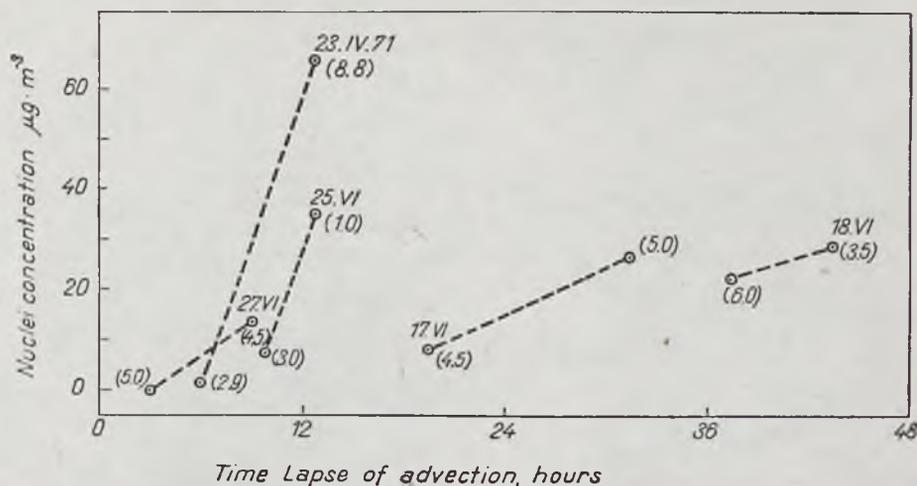
Fig. 2. The concentration increase of sea-salt nuclei in the near water air layer on the Baltic in periods of the maritime air advection from the Atlantic Ocean areas. (Figures in brackets means the wind speed in m. s^{-1} measured from board on the r. vessel when sampling the airborn particles)

Ryc. 2. Wzrost stężenia jąder soli morskiej w powietrzu przywodnym na Bałtyku w okresie adwekcji mas powietrznych znad Atlantyku. (Liczby w nawiasach oznaczają prędkość wiatru w m. s^{-1} zmierzoną z pokładu statku n.-b. w czasie pobierania próbek aerozolu)

also trying to explain the effect of particle size on the decreasing lead concentrations in the air.

In order to explain the effect induced by the mechanism of local factors on lead transport from the atmosphere into the sea, part of the study program under report was devoted to measurement of lead compounds in the Gulf of Gdańsk area. Measurements were taken during 1974—1975 from a research vessel in the Gulf of Gdańsk waters and parallelly at two shore measurement stations as well. One of those stations was located on the Hel Peninsula, in the central part of the Gulf, the other one in Gdynia, closely adjoining the Gulf shore. Air was filtered through fibrous filters at a 7 ÷ 15 m a.s.l. elevation height. At the same time dry fallout was collected during rainless periods by exposure of collecting tanks in the form of polyethylene trays filled with bidistilled water. Total lead concentrations were determined by the atomic absorption method, using a flame attachment.

Pursuant to these investigations, the course of oscillations in the lead concentrations present in air was recorded, as presented in Fig. 3. The curves thus obtained demonstrate high fluctuations of lead concentrations in air. The dominant maximum falls in with the second half of the heating
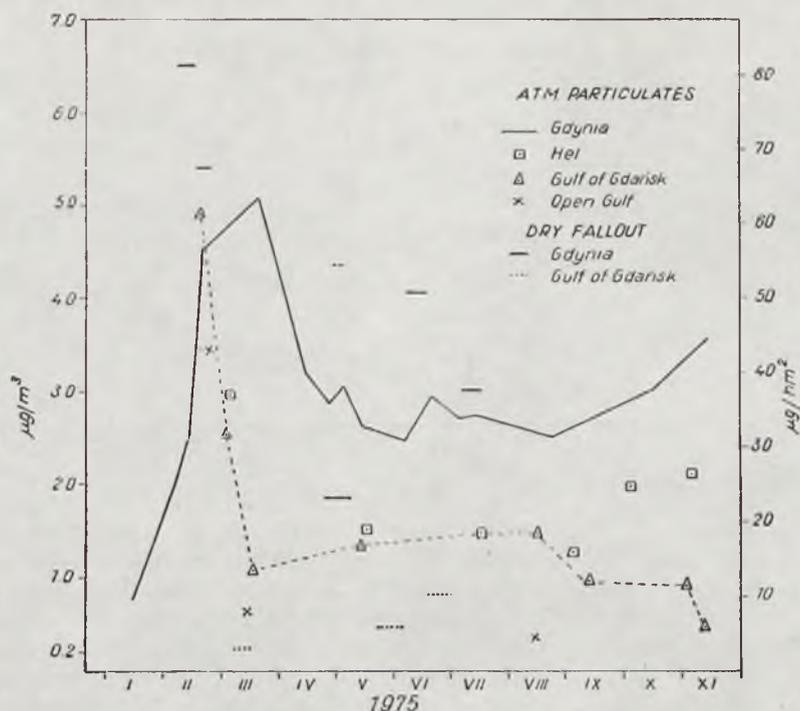
Fig. 3. Temporal trends of lead concentration in air and dry fallout in
the Gulf of Gdańsk area

Ryc. 3. Lokalne wahania stężeń ołowiu w powietrzu i w suchym osa-
dzie w rejonie Zatoki Gdańskiej

period. Again, the much lesser concentrations recorded on the Hel and
the Gulf stations during the summer season clearly indicate the existence
of yet another pollution source, i. e. motor cars.

A comparison of lead concentration curves against the frequency of
onshore winds prevailing during the measurement periods in Gdynia (Fig.
4) has proved that those winds, partly in the nature of a sea breeze, per-
form their part as a local air ventilation mechanism in the Gdynia area,
and at the same time are responsible for the airborne lead transport
above the Gulf of Gdańsk. A confirmation of this fact is also found in the
anticorrelation between the lead concentration fluctuations curve for the
Gulf of Gdańsk versus an analogical curve obtained for Gdynia. During
the more intense periods of air ventilation above Gdynia town, with
the lead concentrations dropping there, there is a corresponding rise in
lead concentrations in the near-water atmospheric layer over the Gulf.
This process is a typical example of an action of mechanism M_3, and its
extent is of a typically local character.
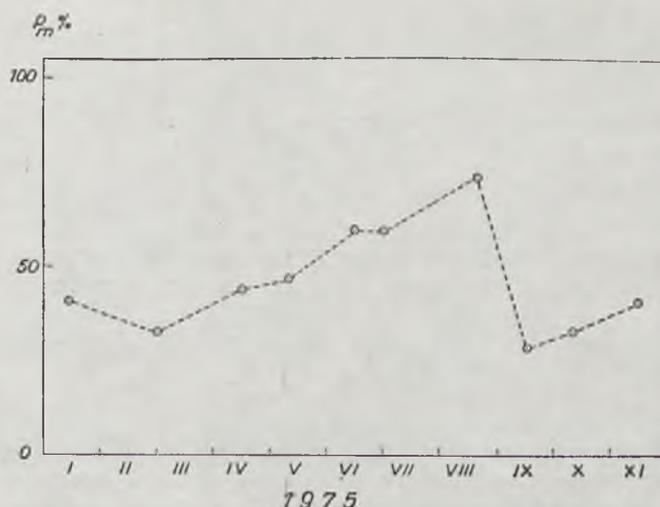
Fig. 4. Frequency of on-shore winds at Gdynia during
measurement of lead in the daytine
Dots correspond to the mean aerosol sampling dates.

Ryc. 4. Częstość występowania wiatrów z kierunków odmor-
skich w okresie pomiarów ołowiu w Gdyni w porze dziennej.
Punkty odpowiadają średnim datom pobierania prób aerozolu.

The important rôle of precipitations in the airborne transport of toxic substances to the sea is confirmed by both the numerical data quoted above and similar example [6, 31] referring to heavy metals and other constituents. To judge by the assessment from various authors, the wash-out in conjunction with the rainout of particles are most prominet of all the mechanisms. Still, it must be noted that the precipitations will not take place at one time troughout the whole area, such as the Baltic Sea, but they are rather sporadical in space and time. On the other hand, the dry fallout normally is a continuous process, taking place within the whole sea area. Consequently, in the final effect the importance of dry fallout may be quite substantial. It must be added that the sampling method of precipitations for chemical analyses, now widely applicable, in the majority of cases does not prevent from accidental sampling of dusts falling out from the air, when collecting precipitation water. That is why the results of such sampling in principle can be loaded with errors and one may doubt whether or to what degree they may be taken to represent the action of mechanisms M_4 and M_5 exclusively. This difficulty is enhanced by the absence of verified data on the height of precipitations above the open seas — an objection which may be referred to the Baltic Sea just as well.

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MECHANIZMY PRZENOSZENIA DOMIESZEK AEROZOLOWYCH DO MORZA
VIA ATMOSFERA NA PRZYKŁADZIE BAŁTYKU

Streszczenie

Bałtyk, jako akwen prawie zamknięty, przedstawia szczególnie ważne zagadnienie z punktu widzenia pełnego bilansu zachodzącej w nim wymiany substancji toksycznych. Nie wolno zaniedbać żadnej składowej tego bilansu, nawet jeżeli to będzie składowa o pozornie nieznacznym tylko udziale. Chodzi tu bowiem o efekty związane z wpływem kumulujących się zmian składu na długotrwałe zachwianie równowagi quasi-stacjonarnej.

W związku z takim stanem rzeczy analizuje się model strukturalny wyjaśniający rolę poszczególnych składowych przenoszenia do morza rozproszonych w atmosferze faz. Dokonuje się oceny działania szeregu mechanizmów przenoszenia, w tym o zasięgu planetarnym, jak i charakterystycznym dla naturalnych synoptycznych typów cyrkulacyjnych, którego wskaźnikiem są jądra soli morskiej przenoszone znad Atlantyku nad Bałtyk.

Szczególną uwagę zwraca się na działanie lokalnych mechanizmów przenoszenia substancji toksycznych z atmosfery do morza. Przykładem występowania takiego mechanizmu jest przenoszenie ołowiu z powietrza do Zatoki Gdańskiej. Z analizy wynika bowiem, że wiatry z kierunków odmorskich, częściowo o charakterze bryzy, odgrywają rolę lokalnego mechanizmu wentylacji powietrza w aglomeracjach miejskich i przenoszenia domieszek nad obszary morskie.

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