

## ANTHROPOGENIC AEROSOLS OVER THE COASTAL ZONE OF THE GDAŃSK BASIN

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### AEROSOLE POCHODZENIA ANTROPOGENICZNEGO W STREFIE BRZEGOWEJ BASENU GDAŃSKIEGO

W strefie brzegowej południowego Bałtyku i nad otwartym morzem zmierzono stężenia jonowych składników aerozoli oraz gazowych składników powietrza. W atmosferze nadmorskiej w aerozolach obecne były zawsze siarczany morskiego pochodzenia, niezwiązane z solą morską ( $\text{Na}_2\text{SO}_4$ ), które jako jądra kondensacji w chmurach odbijają promieniowanie słoneczne poza atmosferę. W maju 2003 w atmosferze nad Gdynią nie obserwowano siarczanu amonu w aerozolach, a za odbijanie światła odpowiedzialny był azotan amonu. W tym samym czasie, gdy w aerozolach nad Głębią Gdańską nie było azotanu amonu rolę tę pełnił siarczan amonu. Ze względu na wysoką wilgotność powietrza nad obiema stacjami aerozol był przesycony parą wodną. Nad Sopotem, w sierpniu 2003 roku obserwowano obie sole amonowe, a stosunek  $\text{NH}_4\text{NO}_3 / (\text{NH}_4)_2\text{SO}_4$  wynosił 1:3. Wilgotność względna powietrza atmosferycznego była wówczas powyżej punktu krystalizacji, aerozol był „mokry” i odpowiadał za odbijanie promieniowania słonecznego.

#### Summary

Concentrations of ionic components of aerosols and gaseous components of air in the coastal zone of the Southern Baltic were determined. In aerosols over the seawater sulphates of marine origin were always found, however, they were not related to the marine salt. Their role as condensation nuclei in clouds is emphasized with the particular focus on scatter of solar radiation behind the atmosphere. In May 2003, in the atmosphere over Gdynia, no ammonium sulphate was detected and ammonium nitrate was found to be responsible for the reflection of the solar radiation. At the same time when aerosols over the Gulf of Gdańsk were poor with ammonium nitrate the backscatter of light was made essentially by ammonium sulphate. Because of high  $R_h$  of the air over both stations aerosol was likely to be supersaturated with water vapor. Both ammonium salts were, in turn, observed over Sopot in August with the  $\text{NH}_4\text{NO}_3 / (\text{NH}_4)_2\text{SO}_4$  ratio equal to 1:3. The relative air humidity was above crystallization point. The aerosol was wet and considered to be responsible for the backscattering of the solar radiation.

## INTRODUCTION

Research in the final decade of the 20<sup>th</sup> century was focused very much on studies of chemical composition of aerosols, their transformations, and exchange fluxes through the sea-air interface as well as on processes which control aerosol transport [3]. Many studies were devoted to elucidate the mechanisms regulating the cycling of elements between the ocean, land and the atmosphere and to determine the effects of economic activities of the man and the effects of increasing human population in the world [4]. Considerable part of aerosols is emitted into the atmosphere directly by evaporation of droplets broken on the sea surface. Chlorides and sulphates comprise nearly 99.5% of marine aerosols and carbonates and other salts form the remaining share [6]. Clean marine air becomes transformed in the coastal zone due to fractionation and enrichment, what is manifested by an increased contribution of ions related to anthropogenic ( $\text{NO}_3^-$ ,  $\text{HNO}_3$ ,  $\text{NH}_4^+$ ,  $\text{NH}_3$ ) and terrigenous ( $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Cl}^-$ ) aerosols [18].

The changes in concentrations of greenhouse gases and their effect on the climate are already well known [14]. Recently, much attention has been focused on light absorption and light scattering phenomena by small submicron aerosols ( $d < 2.5 \mu\text{m}$ ) of anthropogenic origin composed mainly of ammonium nitrate, ammonium sulphate and carbon [8, 11, 21]. At high concentrations of the above constituents of aerosols their radiation properties may cause climate cooling of comparable effect to that of climate warming by  $\text{CO}_2$  ( $-1 \text{ W}\cdot\text{m}^2$ ) [13]. According to some evaluations [11] the effect of climate cooling due to the regional action of anthropogenic aerosols may be very pronounced. These phenomena are not yet fully recognized and understood. More detailed studies are required which involve radiation properties, chemical composition of aerosols in the atmosphere, corresponding processes which occur in the atmosphere and relative dependences on meteorological parameters.

A major goal of the work was to establish which compounds are being formed by particular components of air and aerosols in the atmosphere over the coastal zone of the Gulf of Gdańsk and the open Baltic Sea. An additional task was to establish to what degree the state of aerosols is determined by their origin, ammonium sulphate / ammonium nitrate ratio and the relative humidity. The measurements will be used to indicate major directions for further studies on the effect of aerosols of natural and anthropogenic origin on climate changes in a Southern Baltic scale.

## MATERIALS AND METHODS

Aerosol samples were collected at three measuring stations (Fig. 1): at the permanently used station located on a Gdańsk Deep in the point P1 ( $\varphi = 54^\circ 52' \text{ N}$ ,  $\lambda = 19^\circ 10' \text{ E}$ ) and at two coastal stations: in Gdynia and in Sopot. In the period May 8–15, 2003 parallel measurements on a Gdańsk Deep and in Gdynia, on a roof of the Institute of Oceanography, University of Gdańsk, were done. In the period August 18–23 samples were collected in Sopot, on a roof of the building of the Institute of Oceanology, Polish Academy of Sciences. In all cases the height at which measurements were carried out was the same: 20 m a.s.l. Samples were collected in the 12-hour cycles: between 8.00 a.m. and 8.00 p.m. and between 8.00 p.m. and 8.00 a.m.

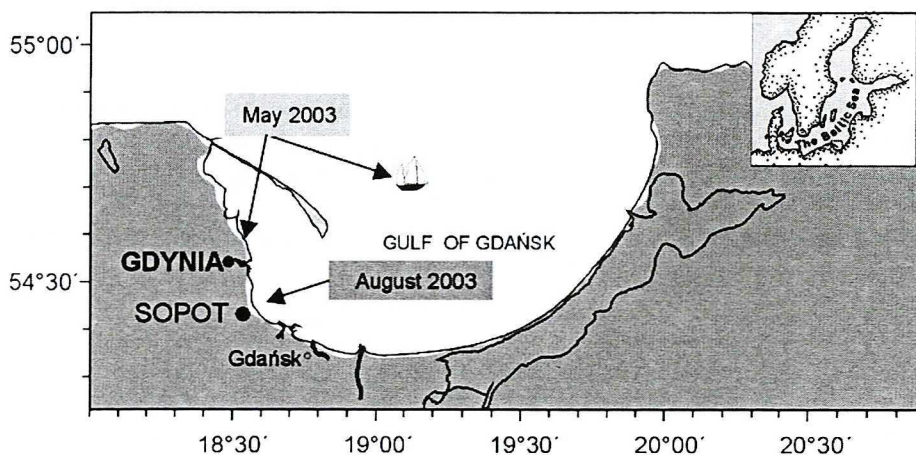


Fig. 1. Location of sampling sites and terms of sampling

At all stations concentrations of  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{NH}_4^+$ ,  $\text{Na}^+$  and  $\text{SO}_4^{2-}$  ions in aerosols and gaseous components in air were determined. The aerosols were collected with the use of filterpack. Each set contained three kinds of filters: teflon, nylon and Whatman 41, impregnated with 5% orthophosphoric acid p.p.a. The sampled air passed first through the pre-washed teflon filter collecting the largest particles (diameter  $> 1 \mu\text{m}$ ), then through the nylon filter where besides particulate fraction of small aerosols (diameter  $< 1 \mu\text{m}$ ) and gaseous constituents of air were retained. On the final Whatman 41 filter at the surface of the impregnating substance a gaseous fraction of ammonia was chemically absorbed. Additionally, gaseous ammonia, sulfuric acid and nitric acid vapors were chemically absorbed with the use of the corresponding denuders of the annular type. For impregnation the following substances were used: 5% orthophosphoric acid p.p.a., 2% calcium carbonate p.p.a. and 4% NaCl p.p.a.

At all sampling sites, filter packs and denuders were installed in parallel in a box securing them from meteorological conditions (rain, and strong wind) and connected to the air pump by polyethylene tubing. The air flow through the set of filters and denuders was regulated with a critical orifice as to provide its constant value  $3.2 \text{ m}^3 \cdot \text{h}^{-1}$  read by the gas meter.

After the sampling all filters were stored in airtight polypropylene bottles, in the refrigerator at the temperature  $+4^\circ\text{C}$ . Before the analysis the content of the filter was extracted via agitation in the ultrasonic bath for 45 min. Ions from teflon and Whatman 41 filters were introduced into deionized water and from nylon filters into carbonate buffer ( $0.75 \text{ mM NaHCO}_3 / 2.2 \text{ mM Na}_2\text{CO}_3$ ) (always  $0.014 \text{ dm}^3$ ).

Samples from denuders were transferred to the polyethylene vials and, as filters, were kept in the refrigerator until the analysis. The analyses of  $\text{NH}_4^+$  [12],  $\text{NO}_3^-$  [1],  $\text{SO}_4^{2-}$  [2] and  $\text{Cl}^-$  [9] ions were carried out by the colorimetric methods. Absorbance was measured with Perkin-Elmer 103 spectrophotometer. Chemical analyses for the presence of sodium were carried out by the atomic absorption method with flame technique. Detailed characteristics of the sampling method, applied analytical procedures and measurement statistics (bias, precision and detection limit) have been presented in [15, 16]. The sampling method and analytical procedures for the determination of macroelements, chloride, nitrate and sulphate in aerosols have been thoroughly discussed in [10, 17, 18].



## RESULTS AND DISCUSSION

Concentrations of sulfate, nitrate, ammonium, chloride ions and gaseous components of air ( $\text{NH}_3$ ,  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$ ) in the atmosphere over the coastal zone of the Gulf of Gdańsk (Gdynia and Sopot) and over the open sea (P1) in the two measuring periods are given in Table 1.

Both, over the marine station and over two land stations, the major components of the atmosphere were salts emitted from the sea surface (Table 1).

Table 1. Concentrations of main aerosol and gas species over Gdynia and Sopot coastal stations and over the Gulf of Gdańsk

Sampling site	Estymator	Concentration [ $\text{nmol m}^{-3}$ ]							
		$\text{Na}^+$	$\text{Cl}^-$	$\text{SO}_4^{2-}$	$\text{H}_2\text{SO}_4$	$\text{NO}_3^-$	$\text{HNO}_3$	$\text{NH}_4^+$	$\text{NH}_3$
Gdynia May 2003	n	13	13	13	13	13	13	13	13
	$\bar{x}$	29.6	119.6	29.7	13.4	17.7	43.6	22.1	33.7
	max	39.7	150.0	41.6	18.1	25.0	100.0	31.7	48.0
	min	23.3	96.0	11.9	9.3	10.3	3.5	14.2	2.4
	SD	4.8	19.4	8.0	2.2	4.7	36.4	5.5	12.3
Gulf of Gdańsk May 2003	n	13	12	12	13	13	13	13	13
	$\bar{x}$	40.9	137.8	31.9	23.6	0.7	2.3	16.2	14.3
	max	60.9	211.0	40.6	25.9	4.5	9.0	65.3	20.4
	min	24.7	90.0	20.9	19.8	0.0	0.1	3.2	9.1
	SD	9.0	31.2	6.6	1.8	1.2	2.7	15.8	3.0
Sopot August 2003	n	10	10	10	10	10	10	10	10
	$\bar{x}$	165.0	153.3	100.0	47.9	24.9	2.6	26.8	18.5
	max	378.6	197.7	129.5	117.3	48.3	7.4	45.0	32.3
	min	33.5	115.7	62.6	1.2	11.0	1.3	11.5	9.2
	SD	113.3	24.9	23.1	31.5	10.6	1.8	9.5	7.6

Symbols: n – number of samples,  $\bar{x}$  – mean value, SD – standard deviation, max – maximal value, min – minimal value.

In order to establish the effect of meteorological parameters on the change in chemical composition of air and to determine relations between particulate and gaseous fractions of the analyzed elements, the Principal Components Analysis (PCA) was used. This statistical method allows to easier understand complex phenomena described with many variables. In particular, it allows to decrease the number of variables to two or three, which are of the greatest significance and consequently to better understand the problem studied. In all cases studied in this work we distinguished two major factors affecting the processes occurring in the atmosphere over Gdynia, Sopot and Gdańsk Deep (Table 2).

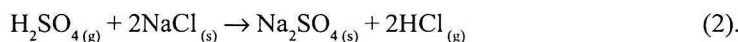
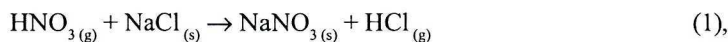
Table 2. Factor analysis of main aerosol and gas species and meteorological parameters in all measurement periods

	Factor loads					
	Gdynia May 2003		Gulf of Gdańsk May 2003		Sopot August 2003	
	Factor		Factor		Factor	
	1	2	1	2	1	2
H <sub>2</sub> SO <sub>4(g)</sub>	-0.14	0.39	-0.11	<b>0.89</b>	0.05	0.47
SO <sub>4(a)</sub>	<b>-0.98</b>	0.15	<b>0.87</b>	0.37	<b>0.60</b>	<b>0.66</b>
NH <sub>4(a)</sub>	<b>0.94</b>	0.14	<b>0.96</b>	0.014	<b>0.60</b>	<b>0.68</b>
NH <sub>3(g)</sub>	0.15	<b>0.73</b>	<b>0.63</b>	<b>0.61</b>	<b>0.78</b>	-0.03
NO <sub>3(a)</sub>	<b>0.90</b>	0.38	0.50	-0.43	0.01	<b>0.67</b>
HNO <sub>3(g)</sub>	0.17	0.45	0.10	-0.12	0.39	-0.52
Cl <sub>(a)</sub>	<b>0.85</b>	0.34	<b>0.68</b>	-0.45	<b>0.87</b>	0.21
Na <sub>(a)</sub>	<b>0.95</b>	0.12	<b>0.82</b>	-0.06	<b>0.88</b>	-0.26
T[°C]	-0.13	-0.08	0.20	<b>-0.82</b>	<b>-0.85</b>	0.38
Rh[%]	0.08	-0.10	-0.35	0.26	0.19	<b>0.86</b>
V <sub>w</sub> [ms <sup>-1</sup> ]	0.18	0.44	<b>-0.72</b>	-0.34	0.48	<b>-0.80</b>
Total contribution [%]	40	23	38	23	36	32

Marked values are > 0.6 and were treated as significant; factors 1 and 2 are of the extreme strength.

On the basis of the PCA analysis, in all studied cases a high correlation coefficient was found between sodium and chloride ions thus indicating their marine origin.

The major marine salt component (NaCl) may react with some gaseous components of the air e.g. sulfuric acid and nitric acid vapors to form sulfates and nitrates respectively [5]:

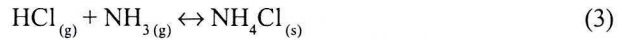


The formation of sodium sulfate in the air was confirmed by the relatively high correlation coefficients in the multifactor analysis (Table 2). On the other hand, when concentration of sodium ions in aerosols over Gdynia was low the reverse proportionality of statistical significance was found. The reason for this seems to be the fact that sodium

sulfate more readily forms large particles which show greater tendency to deposit [17]. The greatest contribution of  $\text{nssSO}_4$  and over 1.5 times higher  $\text{SO}_4^{2-} / \text{Na}^+$  ratio than the proportion of these ions in seawater suggest the anthropogenic origin of sulfate in the atmosphere over Gdynia in May 2003.

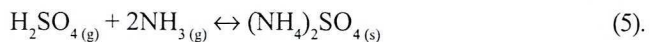
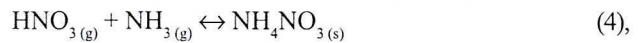
Sodium nitrate was identified in aerosols only in May 2003 over the coastal station in Gdynia, where contribution of marine aerosols is particularly high due the close-to-sea location. During other measuring periods (May 2003 over the Gdańsk Deep and August 2003 over Sopot) the concentration of nitric acid vapors was presumably too low (Table 1) to make the reaction (1) effective enough.

Ammonia which is always present in the atmosphere may react with gaseous HCl formed in reactions (1) and (2). The resulting ammonium chloride:



is relatively unstable and coexist with its decomposition products which in the gas phase, in contradistinction to water solution, appear to be essentially  $\text{NH}_3$  and HCl [20]. Ammonium chloride was identified in aerosols over coastal stations situated in Gdynia and Sopot as well as over the sea. Presumably several parameters, such as temperature, relative humidity of the air and concentration of chloride ions, played an important role in this respect. However, no relationship between its presence and meteorological conditions was found, even with multiple factor analysis used. Generally, low temperatures and high humidity favor the formation of large aerosol aggregates. Bearing in mind the fact that concentrations of ammonia and ammonium ion were comparable (Table 1) one can conclude that the reaction (3) occurred significantly both to the left and right.

Besides of the reaction (3) the ammonia present in the air may react with nitric acid and sulfuric acid vapors to form the corresponding ammonium salts:



Anthropogenic particles of aerosols, i.e. ammonium nitrate and sulphate are influencing the climate, cause the climate cooling, by being able to backscatter the solar radiation. The cooling caused by these particles is equal to the climate warming resulting from  $\text{CO}_2$  increase ( $-1 \text{ W}\cdot\text{m}^{-2}$ ) [7, 22]. Particles saturated with water are liable to backscatter up to two times greater portion of radiation than the “dry” aerosol particles. When aerosol contains ammonium sulphate beside ammonium nitrate, it is mixed internally and the water saturation point is stable [21]. The form of aerosol – wet or dry – is defined by the ratio of ammonium nitrate to ammonium sulphate that are present in the aerosol. From the experimental data [22] it was concluded that particularly wet aerosols exist when the  $\text{NH}_4\text{NO}_3 / (\text{NH}_4)_2\text{SO}_4$  ratio is above 2:1.

It is somewhat interesting that in May 2003, the concentrations of sulfates in aerosols over Gdynia and Gdańsk Deep and in August 2003 over Sopot, were always higher than those of nitrates and ammonium ion (Table 1). However ammonium sulfate was practically undetected in aerosols over Gdynia (Table 2). Instead, in the latter case, we identified the presence of significant amount of ammonium nitrate. Transportation of this species by the



wind from land (Fig. 2) with an average velocity lower than  $2 \text{ m}\cdot\text{s}^{-1}$  ( $0\text{--}9 \text{ m}\cdot\text{s}^{-1}$ ) suggests its local source of origin.

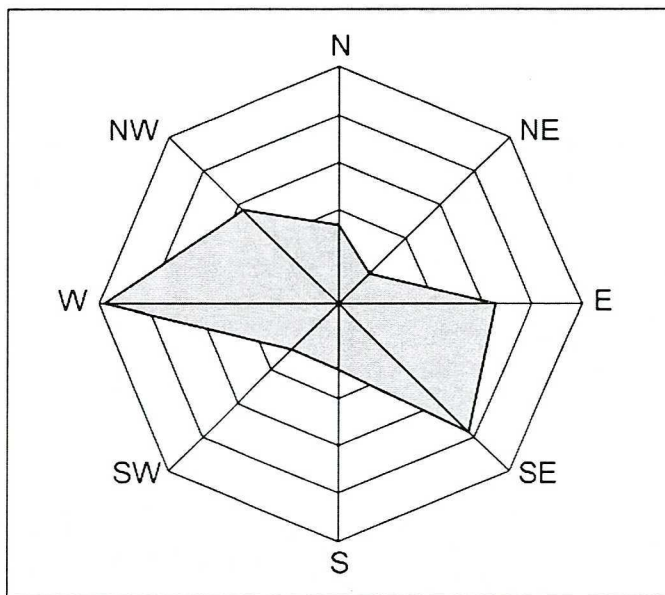


Fig. 2. Wind direction in Gdynia coastal station

Quite high temperature of air (on average  $12.8^{\circ}\text{C}$ ) and low relative humidity (on average 60.6%) were advantageous for evaporation of gaseous components from aerosols. This is confirmed by high concentrations of both  $\text{NH}_3$  and  $\text{HNO}_3$  compared to their ionic forms (Table 1) and by the reverse relationship between ammonium and sulfate ions concentrations (Table 2). According to Dougle et. al. [7] ammonia is a key component determining the formation of ammonium sulfate in aerosols. The same result was found in the multifactor analysis (Table 2). In the case when ammonium sulphate is not present in aerosols and the relative humidity of air reached the value of 62% aerosol becomes saturated with water [7]. This situation was observed in Gdynia in May 2003 and suggests that aerosol could be responsible for the light backscattering in this period.

In May 2003 when the concentration of nitrate ions in aerosols over the Gdańsk Deep was the lowest (Tables 1 and 2), both multifactor analysis and single correlations showed no presence of ammonium nitrate. However the presence of ammonium sulfate in aerosols was detected. The contribution of  $\text{nssSO}_4$  in the total mass of sulfate was 91.7% suggesting their anthropogenic origin, particularly when land winds of high speed (on average  $5.7 \text{ m}\cdot\text{s}^{-1}$ ) were dominating (Fig. 3). The  $\text{SO}_4^{2-} / \text{Na}^+$  ratio was higher in aerosols than in the water which independently confirms anthropogenic origin of sulfates. During the measurements over Gdynia the relative humidity of air ranged from 69% to 100%. According to ten Brink et al. [22] and Gebhart et al. [11], ammonium sulphate becomes to crystallize at humidity level 40–42% in aerosols without ammonium nitrate. One can therefore suppose that aerosols over Gdynia were supersaturated with water and responsible for the backscattering of the sunlight.

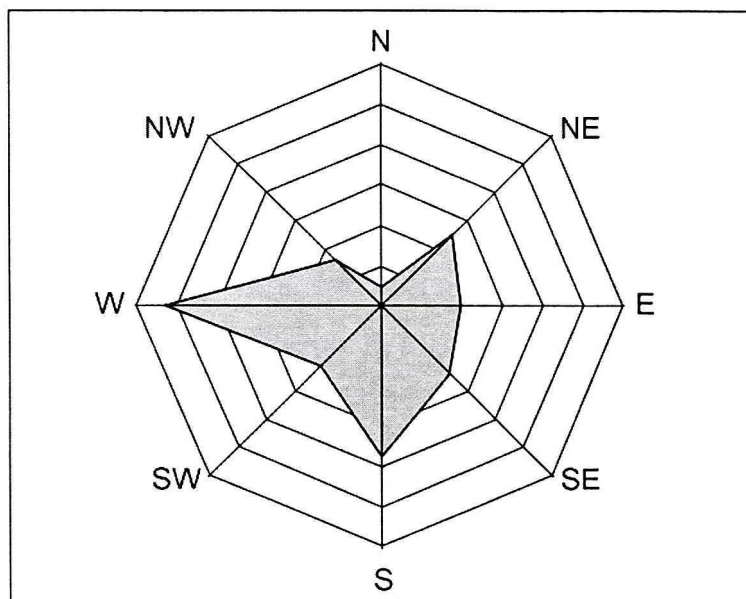


Fig. 3. Wind direction in Gulf of Gdańsk marine station

The presence of ammonium sulfate and ammonium nitrate was found in aerosols over Sopot in August 2003 and confirmed with large regression coefficients of the multifactor analysis (Table 2). Though the winds from the land were prevailing one can only suppose that both these compounds were of anthropogenic origin because the average wind speed ( $1.4 \text{ m}\cdot\text{s}^{-1}$ ) was low (Fig. 4).

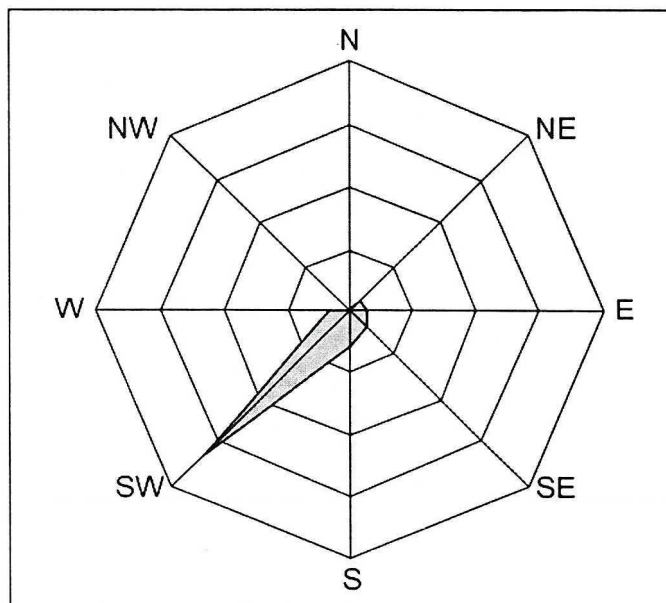


Fig. 4. Wind direction in Sopot coastal station



The contribution of  $\text{nssSO}_4$  was significant (90.1%) and the  $\text{SO}_4^{2-} / \text{Na}^+$  ratio in aerosols was higher than that in the marine water (0.92). The  $\text{NH}_4\text{NO}_3 / (\text{NH}_4)_2\text{SO}_4$  ratio was 1:3. At this ratio supersaturation of aerosols with water occurs when the relative humidity reaches the 61%, i.e. close to that one which is characteristic for ammonium nitrate [21]. During the period under discussion, the relative humidity of air was 77.9% and, as in May, both over the land and sea, the aerosols were supersaturated with water and were able to backscatter the solar radiation.

During short-term measurements carried out over Gdynia, Gdańsk Deep and Sopot it has never been found that ammonium sulfate and nitrate present in aerosols reached the crystallization point. The hydration degree of aerosols was always high and from our results no fact was observed indicating that aerosols of anthropogenic origin capable of backscattering the solar radiation were substantially decreased.

### CONCLUSIONS

- During the measurements carried out at three site stations sulfate ions were found to exist at the highest concentrations of all studied components of aerosols. In prevailing majority (90%) they were  $\text{nssSO}_4$ , indicating that they were of anthropogenic origin or they were formed in biochemical processes occurring in marine water, the latter being less probable.
- In May 2003 no presence of ammonium sulfate was found in aerosols of local origin over Gdynia and the sunlight scattering properties were attributed to ammonium nitrate.
- At the same time, ammonium sulfate was found to be responsible for reflection of sunlight over the Gdańsk Deep while ammonium nitrate was not detected. In both cases, the relative humidity of air was advantageous for high hydration of aerosols thus determining their state.
- The presence of ammonium nitrate and sulfate at the 1:3 ratio was found in the atmosphere over Sopot in May 2003. Because of the high hydration degree promoted by the high relative humidity they were responsible for backscatter of the sunlight.
- While studying major factors affecting the climate cooling one should take into account the change in chemical composition of aerosols paying particular attention to the relative amounts of ammonium, nitrate and sulfate ions which seem to be strongly responsible for the albedo of the Earth.

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