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ROLE OF INDUSTRIAL DUSTS IN THE DETERMINATION OF ATMOSPHERIC NITROGEN INPUT IN THE RYBNIK COAL REGION *

ABSTRACT: Nitrogen input from the atmosphere was investigated in a highly industrialized region. It amounted on an average to $15.26 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{growing season}^{-1}$. The main nitrogen carrier was dust – $11.90 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{growing season}^{-1}$, representing 78.73% of the total input. Field and experimental data have shown that the nitrogen contained in dust was predominantly in the dissoluble form – $10.52 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{growing season}^{-1}$. Nitrogen washed out of the dust and the independent nitrogen input with rain water jointly represented as much as 90.50% of the total nitrogen input from the atmosphere.

KEY WORDS: Nitrogen, rain water, solution, dust, aerosols, washing.

1. INTRODUCTION

Many papers dealing with the cycling of elements represent the view that rain water may get enriched with nutrients due to the contact with air-borne particles that fall down or are sedimented (Ingham 1950, Eriksson 1955, 1959, 1960, 1966, Stenlid 1958, Madgwick and Ovington 1959, Denaeyer-De Smet 1962, Carlisle et al. 1966, White 1969). The presence of dusts and aerosols in the atmosphere, and their interception by plants have often been recorded, and it has only been mentioned that elements can be washed out of them (e.g., Henderson et al. 1977). But there have been few papers quantifying these processes. Washing out of elements from deposited particles and aerosols was in general inferred indirectly by approximate calculations (Mayer and Ulrich 1974), or directly on the basis of experiments (Nihlgård 1970, White and

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Turner 1970). In other papers the role of dust as a carrier of elements is determined on the basis of its amount fallen or its concentration in the air, and element concentration in it (Adamczyk et al. 1978, Kasina et al. 1984). The enriching effect of dust on the rain water is defined as a washing-out potential, determined as the percentage of readily dissolved compounds in the dust dry weight (Maneck i and Skowroński 1984).

It could be expected that in industrialized areas, where dust content in the air is high, the washing-out process would account for a considerable proportion of the input of elements from the atmosphere. It is to industrial emissions that many authors attribute increased inputs of elements to ecosystems from the air (Mayer and Ulrich 1974, Zieliński 1984). In the case of nitrogen it is difficult to arrive at any unequivocal conclusions. For some investigators, who detected nitrogen compounds in industrial dust, and relatively high, of $20 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$, nitrogen input with rain waters, attribute it to close and distant industrial emissions. Others found equally high nitrogen input in unindustrialized areas — in Amazonia (Herrera and Jordan 1981), or in poorly industrialized areas where dust fall is small — in north-eastern Poland (Stachurski and Zimka 1984).

The present study is part of more comprehensive research into the cycling of elements in the pine forests of the highly industrialized Rybnik Coal Region. The aim of the study was to determine the amount and form of nitrogen input from the atmosphere. As the content of dust in the air is high, attention was paid particularly to the role this factor may play in the determination of nitrogen input from the atmosphere.

2. AREA, MATERIAL AND METHODS

Studies of nitrogen input from the atmosphere were carried out in the Rybnik Coal Region, at the village of Kamień, 5 km north-east of Rybnik, in the years 1981 — 1983. Since nitrogen is an important nutrient intensively used by plants, and its input from the atmosphere during the growing season no doubt has an effect on its budget in ecosystems, it was decided to collect samples from April to November each year.

In this period, 14 traps, to collect rain water and falling dust, were set up in an unwooded area. Each trap consisted of a 2 l glass collector and a polyethylene funnel, 10.5 cm in diameter (Fig. 1 a). To prevent nitrogen loss through gaseous NH_3 volatilization and development of microflora during the exposure in the field, chemically pure salicylic acid was poured into the collectors. Two types of filters were placed in each of the funnels. The upper filter was a synthetic fibre net 1 mm in mesh size. The lower filter — in the funnel outlet — was a Whatman GF/B type glass filter of a high retentive power ($1 \mu\text{m}$) and a high through-flow rate ($0.3 \text{ l} \cdot \text{minute}^{-1}$).

Owing to the method used, the dissoluble nitrogen form brought by rain waters or washed out by them from the dust, and the undissoluble form, remaining in the dust, were quickly separated in the field. It was thus possible to estimate not only the amount

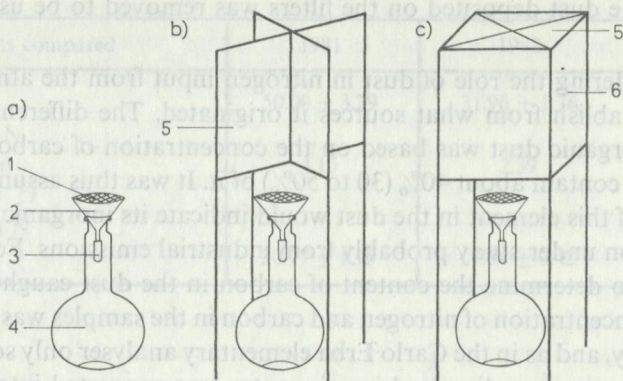


Fig. 1. A diagram showing the structure of a rain trap and traps modified for the experiment a — rain trap (control variant), b — experimental variant I with crossing vertical screens, c — experimental variant II with screens shielded from sides, 1 — net filter, 2 — polyethylene funnel, 3 — glass filter, 4 — glass collector, 5 — crossing vertical screens made of nylon cloth, 6 — nylon cloth side shield

of the element reaching the ground in solution form, but also the amount of dust fall and of the undissoluble nitrogen content in it.

The trap, its functioning and the advantages of using it in studies of this type have been described in detail in the paper by Stachurski and Zimka (1982).

Samples from traps were collected about every 30 days. Water was measured for each trap separately, and each time the glass filters were replaced with new ones of known weights. Since during their passage through the funnel a proportion of fine particles got stuck on the walls of the latter, funnels were also changed and weighed prior to and after the exposure in the field, with an accuracy to the nearest 0.01 mg. It was, therefore, possible to apply corrections to the amount of dust fall estimated from the sediment on the glass filters.

An experiment has also been carried out, the aim of which was to determine the effect of air-borne and falling dust particles on the input of dissolved nitrogen. To do this, near the main traps 9 additional traps, identical with them were set up, but: (a) 3 of them were used as controls (Fig. 1 a), (b) above three — vertical nylon cloth screens 50 cm × 50 cm, crossing each other, were fixed (Fig. 1 b), (c) above three — the same screens were fixed, but with a 50 cm nylon cloth band protecting them from the sides (Fig. 1 c).

The control traps trapped mainly those particles that were falling gravimetrically. The purpose of the screens used in variant I was to increase dust fall by intercepting aerosol particles circulating in the air. In variant II side shielding was used to prevent trapping particles whose movement was not vertical.

From all traps — control and modified for the experiment — samples were collected every month, following the procedure described above.

Glass filters were prior to field exposure and then after it (covered with sediment) dried at 65°C for 48 hours and weighed with an accuracy to the nearest 0.01 mg.

Subsequently the dust deposited on the filters was removed to be used for chemical analyses.

When considering the role of dust in nitrogen input from the atmosphere it was necessary to establish from what sources it originated. The differentiation between organic and inorganic dust was based on the concentration of carbon. Fine organic matter particles contain about 40% (30 to 50%) of it. It was thus assumed that a lower concentration of this element in the dust would indicate its inorganic origin – in the case of the region under study probably from industrial emissions. For this reason, it was necessary to determine the content of carbon in the dust caught.

Since the concentration of nitrogen and carbon in the samples was analysed by gas chromatography, and as in the Carlo Erba elementary analyser only solid samples can be analysed, the nitrogen dissolved in rain water was converted into the solid state.

The methods used in this study for converting water samples into the solid state (evaporation to a dry residuum), and for determining the total concentration of all the substances contained in one litre of the solution have been described by *Stachurski and Zimka* (1984).

The dust removed from the glass filters and the sediment left after the evaporation of solutions were homogenized and dried at 65°C for 24 hours. The material so prepared was used for making standard weighed amounts of about 0.800 mg with an accuracy to the nearest 0.001 mg, which were subsequently tested for nitrogen and carbon concentration of a Carlo Erba elementary analyser.

The concentrations thus obtained were the total of organic, ammonia, nitrite and nitrate nitrogen in the samples. Likewise, the carbon concentrations constituted the total of organic and carbonate carbon. There was only a very small standard error of estimation of the elements in samples by the elementary analyser, assuming values below 0.03%.

In their paper *Stachurski and Zimka* (1984) have described in detail the advantages of the gas chromatography used for determining nitrogen in samples of this type in studies of the cycling of elements.

Given data on the concentration in $\text{mg}\cdot\text{l}^{-1}$ of all substances in the solution, and the percentage concentration of nitrogen in the sediment left after rain water evaporation, one can calculate the concentration in $\text{mg}\cdot\text{l}^{-1}$ of this element in a sample, by multiplying one of these values by the other. Nitrogen and carbon per cent concentration in dust samples was obtained directly.

3. RESULTS

3.1. NITROGEN INPUT FROM THE ATMOSPHERE WITH RAIN WATERS AND DUST

In the study two forms of nitrogen input to ecosystems were taken into account: dissolved in rain waters and undissolved in dust. The former was considered important

Table 1. Input of total nitrogen dissolved in rain waters in the growing season (April–November) in the Rybnik Coal Region ($\bar{x} \pm SD$)

Elements compared	1981	1982	1983
Rainfall (cm)	50.00 \pm 3.29	31.96 \pm 1.76	39.20 \pm 1.96
Weighed average N concentration in solutions ($\text{mg} \cdot \text{l}^{-1}$)	4.15	2.39	3.37
Input of dissolved N ($\text{kg} \cdot \text{ha}^{-1}$)	20.77 \pm 1.15	7.64 \pm 0.42	13.22 \pm 0.92

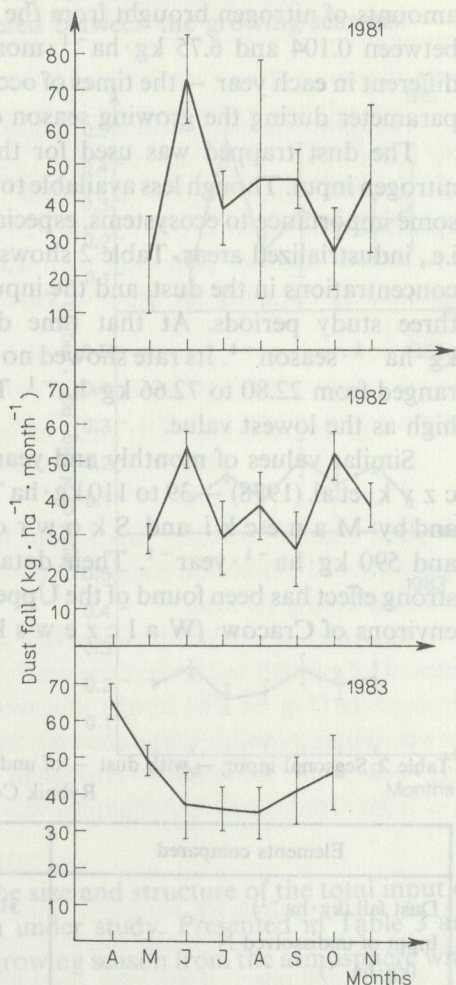
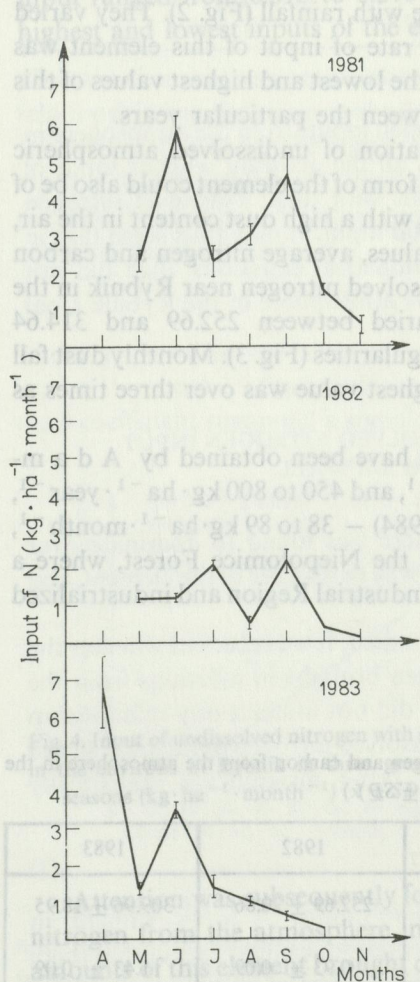


Fig. 2. Input of total dissolved nitrogen with rain waters from the atmosphere in the growing seasons 1981–1983 (April–November) in the Rybnik Coal Region ($\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$) ($\bar{x} \pm SD$)

Fig. 3. Dust fall from the atmosphere in the Rybnik Coal Region in the growing seasons 1981–1983 ($\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$) ($\bar{x} \pm SD$)

because of its availability to plants, and the latter was interesting on account of the high dust content in the air, characteristic of industrialized regions.

Nitrogen amounts brought from the atmosphere with rain waters in the area considered were rather considerable. Table 1 presents seasonal amounts of this element input in this way, its average concentration in solutions and amounts of rainfall in successive study periods. Dissolved nitrogen input ranged from 7.64 to 20.77 kg · ha⁻¹ · growing season⁻¹, thus being fairly variable. In the growing season of 1981 almost three times as much dissolved nitrogen was input as in the growing season of 1982.

During the studies, considerable variation was also observed in the monthly amounts of nitrogen brought from the atmosphere with rainfall (Fig. 2). They varied between 0.104 and 6.75 kg · ha⁻¹ · month⁻¹. The rate of input of this element was different in each year — the times of occurrence of the lowest and highest values of this parameter during the growing season differed between the particular years.

The dust trapped was used for the determination of undissolved atmospheric nitrogen input. Though less available to plants, this form of the element could also be of some importance to ecosystems, especially in areas with a high dust content in the air, i.e., industrialized areas. Table 2 shows dust fall values, average nitrogen and carbon concentrations in the dust, and the inputs of undissolved nitrogen near Rybnik in the three study periods. At that time dust fall varied between 252.69 and 314.64 kg · ha⁻¹ · season⁻¹. Its rate showed no seasonal regularities (Fig. 3). Monthly dust fall ranged from 22.80 to 72.66 kg · ha⁻¹. Thus the highest value was over three times as high as the lowest value.

Similar values of monthly and yearly dust fall have been obtained by A d a m c z y k et al. (1978) — 39 to 110 kg · ha⁻¹ · month⁻¹, and 450 to 800 kg · ha⁻¹ · year⁻¹, and by M a n e c k i and S k o w r o ń s k i (1984) — 38 to 89 kg · ha⁻¹ · month⁻¹, and 590 kg · ha⁻¹ · year⁻¹. Their data concerned the Niepolomice Forest, where a strong effect has been found of the Upper-Silesian Industrial Region and industrialized environs of Cracow (W a l c z e w s k i 1984).

Table 2. Seasonal input — with dust — of undissolved nitrogen and carbon from the atmosphere in the Rybnik Coal Region ($\bar{x} \pm SD$)

Elements compared	1981	1982	1983
Dust fall (kg · ha ⁻¹)	314.64 ± 45.73	252.69 ± 20.86	309.96 ± 18.95
Input of undissolved N (kg · ha ⁻¹)	1.78 ± 0.27	0.93 ± 0.09	1.43 ± 0.09
Average percentage of N in dust	0.71 ± 0.10	0.45 ± 0.04	0.58 ± 0.03
Average percentage of C in dust	10.63 ± 1.49	11.20 ± 0.90	11.60 ± 0.70

In the dust trapped near Rybnik relatively low carbon concentrations — 10.63 to 11.60% were found (Table 2). As it is known that organic matter particles contain about 40% of this element, it could be considered inorganic dust. This finding, combined with the fact that the region under study is highly industrialized, permits the presumption that the dust analysed probably originated from industrial emissions.

The amount of undissolved nitrogen input from the atmosphere varied during the growing season between 0.93 and 1.78 $\text{kg} \cdot \text{ha}^{-1} \cdot \text{growing season}^{-1}$ (Table 2). Thus the amount of undissolved nitrogen input in the growing season of 1982 was almost a half smaller than that input in the 1981 growing season. Monthly undissolved nitrogen input ranged from 0.098 to 0.276 $\text{kg} \cdot \text{ha}^{-1}$ (Fig. 4). The times of occurrence of the highest and lowest inputs of the element differed between the growing seasons.

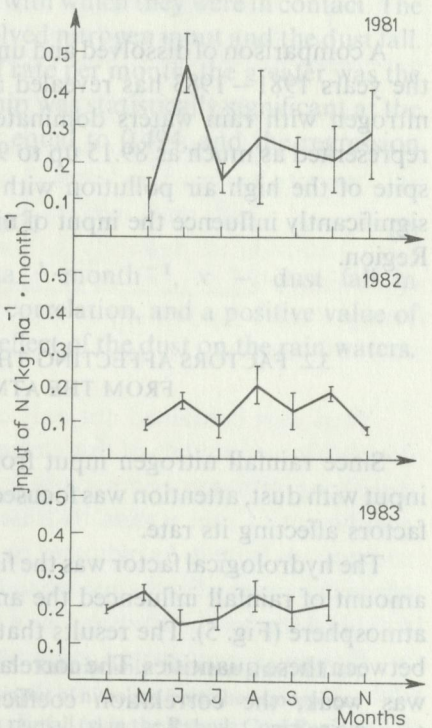


Fig. 4. Input of undissolved nitrogen with dust in the environs of Rybnik in three growing seasons ($\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$) ($\bar{x} \pm \text{SD}$)

Attention was subsequently focused on the size and structure of the total input of nitrogen from the atmosphere in the region under study. Presented in Table 3 are amounts of this element brought during the growing season from the atmosphere with rainfall and dust, and their respective percentages.

The amount of nitrogen input during the growing season was found to vary greatly from year to year, ranging from 8.57 to 22.55 $\text{kg} \cdot \text{ha}^{-1} \cdot \text{growing season}^{-1}$. Thus the highest total growing-season nitrogen input from the atmosphere was nearly three times as great as the lowest input.

Table 3. Total seasonal nitrogen input from the atmosphere – with rain waters and dust – in the Rybnik Coal Region

Elements compared	1981		1982		1983	
	kg · ha ⁻¹	%	kg · ha ⁻¹	%	kg · ha ⁻¹	%
Input of dissolved N with rainfall	20.77	92.11	7.64	89.15	13.22	90.24
Input of undissolved N with dust	1.78	7.89	0.93	10.85	1.43	9.76
Total input of N	22.55		8.57		14.65	

A comparison of dissolved and undissolved nitrogen inputs from the atmosphere in the years 1981–1983 has revealed a regularity. In all the growing seasons input of nitrogen with rain waters dominated over its input with dust. Dissolved nitrogen represented as much as 89.15 up to 92.11% of its total input. It may thus seem that in spite of the high air pollution with dust, due to industrial emissions, dust did not significantly influence the input of nitrogen from the atmosphere in the Rybnik Coal Region.

3.2. FACTORS AFFECTING THE SIZE OF DISSOLVED NITROGEN INPUT FROM THE ATMOSPHERE WITH RAINFALL

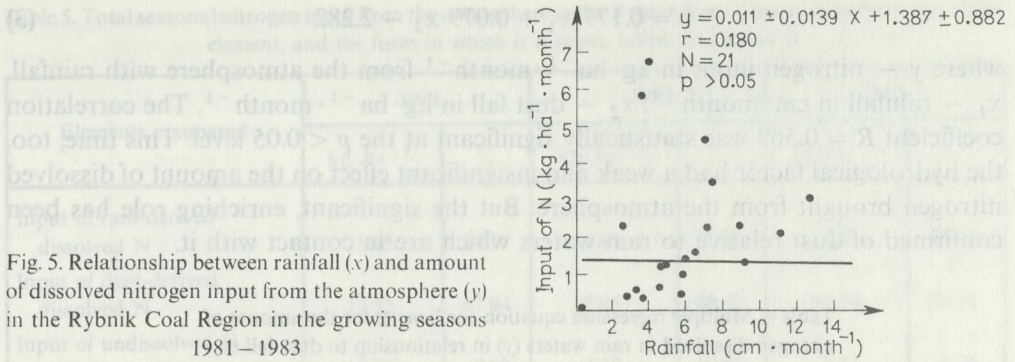
Since rainfall nitrogen input from the atmosphere was found to dominate over input with dust, attention was focused on the former way of transfer of the element, and factors affecting its rate.

The hydrological factor was the first agent to be studied. It was checked whether the amount of rainfall influenced the amount of nitrogen brought in solutions from the atmosphere (Fig. 5). The results that were obtained did not indicate any relationship between these quantities. The correlation between dissolved nitrogen input and rainfall was weak, the correlation coefficient, $r = 0.180$, being insignificant statistically ($p > 0.05$). The regression equation was of the form

$$y = 0.011 x + 1.387 \quad (1)$$

where y – dissolved nitrogen input in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$, x – rainfall in $\text{cm} \cdot \text{month}^{-1}$. When this factor was considered separately, one could expect that there was no definite effect of the hydrological factor on the amount of nitrogen input from the atmosphere in form of solutions.

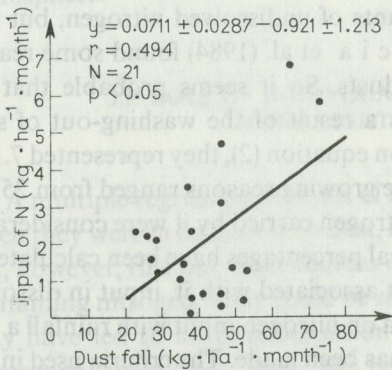
Because there was no indication that the amount of rainfall has an effect on the amount of nitrogen brought with it, it was decided to relate this parameter to the amount of dust fall. Particles present in the air and falling down in large numbers could



affect the chemical composition of the rain waters, with which they were in contact. The relationship was therefore analysed between dissolved nitrogen input and the dust fall. It has been found that the higher was the dust fall rate per month, the greater was the input of dissolved nitrogen (Fig. 6). The relationship was statistically significant at the $p < 0.05$ level, the correlation coefficient r being equal to 0.494, and the regression equation being of the form

$$y = 0.0711 x + 0.921 \quad (2)$$

where y — dissolved nitrogen input in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$, x — dust fall in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$. Statistical significance of the correlation, and a positive value of the b coefficient suggested a significant, enriching effect of the dust on the rain waters.



To check that the above supposition is true, the effects, of the hydrological factor and the dust factor on the amount of dissolved nitrogen input from the atmosphere were studied jointly (Table 4). A statistically significant relationship was found only in the case of dust ($p < 0.05$). The relationship was as follows: the higher the dust fall, the higher the input of nitrogen with rain water. The effect of the amount of rainfall has proved to be insignificant ($p > 0.05$). A multiple regression equation of the following form was obtained:

$$y = 0.177 x_1 + 0.079 x_2 - 2.282 \quad (3)$$

where y – nitrogen input in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$ from the atmosphere with rainfall, x_1 – rainfall in $\text{cm} \cdot \text{month}^{-1}$, x_2 – dust fall in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$. The correlation coefficient $R = 0.569$ was statistically significant at the $p < 0.05$ level. This time, too, the hydrological factor had a weak and insignificant effect on the amount of dissolved nitrogen brought from the atmosphere. But the significant, enriching role has been confirmed of dust relative to rain waters which are in contact with it.

Table 4. Multiple regression equation coefficients for the amount of nitrogen dissolved in rain waters (y) in relationship to dust fall (x_1) and rainfall (x_2) for three growing seasons – data from the Rybnik

Coal Region – $N = 21$

y – input of nitrogen dissolved in rain waters ($\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$),

x_1 – amount of rainfall ($\text{cm} \cdot \text{month}^{-1}$), x_2 – amount of dust fall ($\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$)

	$b_1 x_1$	$b_2 x_2$	a	R
	0.079	0.177	-2.282	0.569
SE	0.028	0.121		1.489
t	2.782	1.463		
p	<0.05	>0.05		<0.05

Thus dust contained not only certain amounts of undissolved nitrogen, but also dissoluble compounds of this element. G a r ś c i a et al. (1984) found some readily dissolving nitrogen compounds in industrial dusts. So it seems probable that the enrichment of rain waters by dust occurred as a result of the washing-out of such compounds from it. As indicated by the regression equation (2), they represented 7.11% of the dry weight. Since dust fall in the successive growing seasons ranged from 252.69 to 314.64 $\text{kg} \cdot \text{ha}^{-1}$, the amounts of dissoluble nitrogen carried by it were considerable.

By using the regression equation (2) theoretical percentages have been calculated of the nitrogen washed out of dust, and of that not associated with it, input in dissolved form. From these percentages and empirical data on nitrogen input with rainfall a new division of the total input from the atmosphere has been made. The criteria used in this division were the origin and form of nitrogen input to ecosystems (Table 5). The calculations resulted in a picture different from that obtained before (Table 3, 5). The factor determining the amount of nitrogen input in solutions appeared to be the dust, and more precisely the washing-out from it. Calculated from the data presented in Table 5, it constituted 73.76 up to 77.91% of the total input of nitrogen with rainfall. It is, therefore, dust that seems to be the factor determining the size of nitrogen input from the atmosphere, in undissolved form and in solutions, in the highly industrialized region considered.

Table 5. Total seasonal nitrogen input from the atmosphere in the Rybnik Coal Region, with the origin of the element, and the form in which it is input, taken into account

Elements compared	1981		1982		1983	
	kg · ha ⁻¹	%	kg · ha ⁻¹	%	kg · ha ⁻¹	%
Input of rain-derived dissolved N	5.45	24.17	1.69	19.72	2.92	19.93
Input of dust-derived dissolved N	15.32	67.94	5.95	69.43	10.30	70.31
Input of undissolved N with dust	1.78	7.89	0.93	10.85	1.43	9.76
∑ of dust-derived N input	17.10	75.83	6.88	80.28	11.73	80.07
Total input of N	22.55		8.57		14.65	

The earlier supposition, based solely on the classification of nitrogen input into dissolved and undissolved forms, assuming little importance of dust, has thus proved wrong. Of course dust was only carrying small amounts of undissolved nitrogen, but over 70% of the nitrogen input with rain waters originated from dust.

Thus the statement could probably be ventured that although the solution form of nitrogen transfer from the atmosphere predominated (89.15–92.11%), the main factor determining the size of input of this element was dust. In the Rybnik Coal Region it carried, in both forms, 75.83 to 80.28% of the total input of nitrogen from the atmosphere.

3.3. ROLE OF INDUSTRIAL DUST AS A SOURCE OF NITROGEN IN THE LIGHT OF EXPERIMENTAL DATA

A multiple regression analysis of the field data made probable the supposition that when they were in contact with dust, rain waters got enriched with nitrogen. One could not, however, rule out other sources of solution enrichment, namely emissions of gases containing nitrogen. They could be simultaneous with industrial dust emissions, which may have led to an overestimation of the role of dust in rainfall enrichment. The experiment mentioned above has been carried out to solve these doubts.

In the experiment the same traps were used as those applied in the field to trap dust and rainfall, with various shielded and unshielded screens fixed above them. The trap set-ups were modified so that they would diversify the amounts of dust caught. In variants I and II, and in the control set-up highly retentive Whatman glass filters were used for dust deposition. They trapped particles above 1 μm in diameter, that is, even very fine dust, aerosols and smoke. The results in the form of means of: dust fall, nitrogen concentration in dust, and nitrogen input with rain waters for the three experimental variants have been presented in Table 6.

Table 6. Nitrogen input with rainfall, rate of dust fall ($\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$), and nitrogen concentration in dust, determined during the experiment carried out in the environs of Rybnik ($\bar{x} \pm SD$)

Variant	Input of dissolved N	Dust fall rate	Percentage of N in dust
Control	1.30 ± 0.49	40.83 ± 5.72	0.76 ± 0.26
II (shielded)	1.56 ± 0.75	49.78 ± 8.96	0.62 ± 0.10
I (unshielded)	3.06 ± 1.41	99.17 ± 16.86	0.51 ± 0.07

It has been noticed that the screens fixed above the traps caused a considerable increase in the amount of dust trapped, as compared with the control variant. For the controls the amount trapped was $40.83 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$, and for variant I it was over twice as high, being equal to $99.17 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$. The difference in the amount of dust between these variants was statistically significant at the $p < 0.001$ level. In the case of variant II only a slight increase in dust fall was obtained in comparison with the control traps.

It has thus been found that unshielded screens fixed above the traps increased the amount of dust fall recorded by them. This increase probably resulted from the trapping of the dust fraction circulating in the air. Aerosol type particles stopped by them fell into the traps, thus causing an increase in the recorded amount of dust.

Experimental data indicated that a higher dust fall was accompanied by an increased dissolved nitrogen input. The monthly average input of this element with rain waters, as determined for variant I — $3.06 \text{ kg} \cdot \text{ha}^{-1}$ — was over twice as high as that estimated for the controls — $1.30 \text{ kg} \cdot \text{ha}^{-1}$. The difference between these quantities was statistically significant at the $p < 0.01$ level. In variant II a slight increase in dust fall was also accompanied by some insignificant increase in the input of nitrogen in the dissolved form.

On the basis of the results obtained from the experiment the potential effect of gaseous emissions on nitrogen input with rainfall was studied. Such emissions could have been synchronous with the dust emissions. So they may have blurred the picture of the relationship between nitrogen input in solution and the dust fall. The traps used in the experiment diversified only the amounts of dust trapped. For gaseous forms the access was the same in all variants. If gases had been the main factor responsible for the enrichment of rain waters with nitrogen, the input of dissolved nitrogen, estimated for the study period, would have been the same in all the three trap types. Yet in the experiment statistically significant differences in the amount of solution nitrogen input have been found between control and modified traps. Moreover, an increase in dust fall was always accompanied by a higher dissolved nitrogen input. It seems, therefore, that the gaseous emissions that occurred in the area under study did not exert a significant enriching influence on the rain waters.

The conclusions drawn on the basis of the data from the experiment were thus similar to those arrived at previously, based on field data — obtained from unmodified

rain traps. To see if they were true, the relationship was analysed between dissolved nitrogen input and the amount of dust fall (Fig. 7). A positive correlation between these parameters has been confirmed. The higher the amount of fallen dust, the greater the rainfall input of nitrogen. The regression equation was of the form

$$y = 0.0285x + 0.174 \quad (4)$$

where y — solution nitrogen input in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$, x — dust fall in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$, obtained in the experiment. The correlation coefficient, $r = 0.886$, was statistically significant at the $p < 0.01$ level. The coefficient of rainfall enrichment with dust, $b = 0.0285 \pm 0.0061$, was slightly lower than that obtained for field data — $b = 0.0711 \pm 0.0287$ (equation (2)), but the difference between them was not significant statistically ($p > 0.05$).

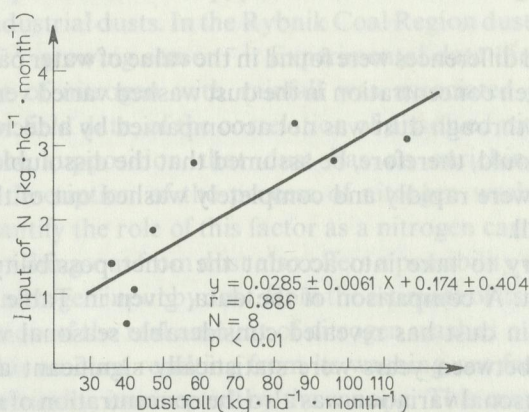


Fig. 7. Relationship between dust fall (x) and amount of nitrogen dissolved in rain waters, brought from the atmosphere (y) established on the basis of experimental data

Coefficients of rainfall enrichment with dust, present in both regression equations (2 and 4) — for field and experimental data, showed fairly great variation, which affected the correlation value. There were two possible causes of this variation. One of them could be a variable rate of the washing-out of nitrogen compounds from dust, depending on the amount of rainfall. The other — a variable concentration of these compounds in dust.

In the checking for the former possibility advantage was taken of the fact that in the experimental variants different amounts of dust were trapped. As a result, with similar rainfall volumes falling into traps, for the particular variants different values of water passage through a dust unit have been obtained. Assuming that the washing-out of nitrogen from dust depended on the amount of water passing through it, one arrives at the conclusion that an increased water passage should be followed by a lowered nitrogen concentration in the dust washed. For this reason, a comparison was made of the means, for one month, of rainfall, dust fall, water passage through 1 mg dust, and the concentration of nitrogen in the different experimental variants (Table 7). Great,

Table 7. Amount of rain water passing through dust and the concentration of nitrogen in the dust washed – experimental data for 1 month ($\bar{x} \pm SD$)

Elements compared	Control	Variant II (shielded)	Variant I (unshielded)
Rainfall (l · m ⁻² · month ⁻¹)	28.08 ± 0.56	21.59 ± 3.24	24.94 ± 4.99
Dust fall (kg · ha ⁻¹ · month ⁻¹)	32.40 ± 0.97	33.30 ± 3.33	77.13 ± 9.26
Water passage through dust (l · mg ⁻¹)	8.69	3.23	6.52
Percentage of N in dust	0.56	0.46	0.52

nearly two- and threefold differences were found in the value of water passage through a dust unit, whereas nitrogen concentration in the dust washed varied very little. Thus an increased water passage through dust was not accompanied by a decrease in nitrogen concentration in it. It should, therefore, be assumed that the dissoluble compounds of the element considered were rapidly and completely washed out of the dust even by small amounts of rainfall.

It was then necessary to take into account the other possibility – a variable nitrogen content in dust. A comparison of the data, given in Table 2, on dissolved nitrogen concentration in dust has revealed considerable seasonal variation of this parameter. Differences between years were statistically significant at the $p < 0.001$ level. Subject to intra-seasonal variation was also the concentration of nitrogen in dust, which can be seen when the rate of dust fall and the amount of nitrogen input with it are compared. Similar variations probably occurred in the amount of dissoluble nitrogen in dust. The possibility of washing-out nitrogen from dust would thus be determined only by the concentration of dissoluble nitrogen compounds in it.

The results of the experiment fully confirmed the field-data-based conclusions concerning the nitrogen-enriching effect of dust on rain waters. The presence of this element in rainfall was mainly the result of its washing-out from dust. Within the limits of the study, the process was complete and independent of the amount of water passing through dust. The coefficient of water enrichment by dust could thus be considered equivalent to the dissoluble nitrogen compound content in it.

The increased dust fall, observed in the experiment, due to the presence of screens fixed above the traps, intercepting air-borne aerosol particles, could be used as an extremely simplified model representing phenomena occurring in wooded areas and woods. Amounts of dust stopped by tree crowns provide an additional input of dissoluble and undissoluble nitrogen compounds to the ecosystem. It seems, therefore, that in the calculating of nitrogen budget for woodland ecosystems in highly industrialized areas this additional input of nitrogen, no doubt of considerable size, from the atmosphere, should be taken into account.

4. DISCUSSION

The forest ecosystem tendency, observed in Europe in recent years, towards eutrophication is attributed to the growth of industry and the resultant air pollution (Mayer and Ulrich 1974, Adamczyk et al. 1978, Jakucs et al. 1981, Zieliński 1984). In the Niepołomice Forest Adamczyk et al. (1978) have found amounts of dusts from large urban areas, and industrial emissions ranging from 450 to 800 kg·ha⁻¹·year⁻¹. These dusts also contained nitrogen compounds (García et al. 1984). Along with other readily dissolving substances they represented from 24 to 45% of dust weight (Maneck and Skowroński 1984). In the last decade, in Hungary Jakucs et al. (1981) found an almost twofold growth of nitrogen input with rainfall.

The results presented in this paper confirm these findings within the range of local emissions of industrial dusts. In the Rybnik Coal Region dust fall ranged from 252.69 to 314.64 kg·ha⁻¹·growing season⁻¹. Experimental data directly indicate that in that area the input of nitrogen with rainfall was associated with dust fall. Statistical significance, for field data, of the correlation of dissolved nitrogen input with dust fall also proves the supposition that dust has an enriching effect on solutions. A mathematical description of the process of nitrogen washing out of dust makes it possible to quantify the role of this factor as a nitrogen carrier. The calculation of the coefficient of washing-out from dust also offers a possibility of theoretically calculating the amount of nitrogen input by dust, from the estimate of its amount fallen. This allows the determination of the real amount of nitrogen washed out of the dust reaching the tree crowns, this amount resulting from its washing out from air-borne dust or dust falling gravimetrically, intercepted by the canopy. This may prove very significant in the calculation of the nitrogen budget in an ecosystem.

The establishment of the role of industrial dust in the determination of dissolved nitrogen input from the atmosphere, and the possibility of its direct quantification seem to be useful in studies of the cycling of elements.

By using a mathematically-calculated coefficient of washing-out from dust — 7.11% — it has been established that washed-out nitrogen represents 73.76 up to 77.91% (on an average 76.52%) of its total input with rainfall. Before they contacted dust, rain waters had thus been carrying as little as 1.69 to 5.45 kg·ha⁻¹ of nitrogen during the growing season (on an average 3.35 kg·ha⁻¹·growing season⁻¹), while the total input of dissolved nitrogen ranged from 7.64 to 20.77 kg·ha⁻¹·growing season⁻¹ (on an average 13.88 kg·ha⁻¹·growing season⁻¹). Similar yearly amounts of nitrogen input with rainfall have been obtained by Mayer and Ulrich (1974) — 25.4 kg·ha⁻¹, and Heinrichs and Mayer (1977) — 22.6 kg·ha⁻¹ — in the Solling district near the Ruhr Region, as well as by Zieliński (1984) — 20.8 kg·ha⁻¹ — for the Niepołomice Forest located in the zone of impact of the industrialized environs of Cracow and the Upper-Silesian Industrial Region, and Jakucs et al. (1981) for Hungary — 22.62 kg·ha⁻¹.

The above data show how important and decisive the role is of industrial dusts in determining the size of nitrogen input from the atmosphere. It seems that from the point of view of studies on the budget of this nutrient in ecosystems of considerable importance is the fact that they significantly increase the input of dissolved nitrogen, easily assimilated by plants.

5. SUMMARY

The influence of dust on the amount of total nitrogen input from the atmosphere was studied in a highly industrialized area.

The methods used made it possible to estimate dust fall and nitrogen concentration in it, as well as to determine the input of dissolved nitrogen. The investigations were carried out in the Rybnik Coal Region in the years 1981–1983.

In that period the amount of nitrogen input with rainfall during a month (Fig. 2) and during a growing season (Table 1) showed considerable variation. It ranged from 0.104 to $6.750 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$. Total dissolved nitrogen input per season varied between 7.64 and $20.77 \text{ kg} \cdot \text{ha}^{-1}$.

Variation has also been found in the monthly – between 22.80 and $72.66 \text{ kg} \cdot \text{ha}^{-1}$ – and seasonal – between 252.69 and $314.64 \text{ kg} \cdot \text{ha}^{-1}$ – dust fall (Table 2). Subject to variations was also the concentration of undissolved nitrogen compounds in dust – between 0.27 and 0.99% (Table 2). As a result of this variation, the monthly input of undissolved nitrogen ranged from 0.084 to $0.468 \text{ kg} \cdot \text{ha}^{-1}$ (Fig. 4), and the seasonal input – from 0.93 to $1.78 \text{ kg} \cdot \text{ha}^{-1}$ (Table 2).

The results indicated that in the input of nitrogen from the atmosphere nitrogen dissolved in rain water – 89.15 to 92.11% – dominated over its undissolved form – in dust.

Multiple regression was used to analyse the effect of the hydrological factor and dust fall on nitrogen input with rain water (equation (3)). It has been found that the amount of rainfall has no statistically significant influence on the input of dissolved nitrogen (Table 4). But statistically significant was the effect of dust fall on the value of this parameter (Table 4). It was positively correlated with dissolved nitrogen input. The rate of the washing-out of nitrogen from dust amounted to 7.11% of the weight of the latter (regression equation (2) – Fig. 6).

Using a calculated coefficient of the washing-out of nitrogen from dust, the percentage was theoretically calculated of dust-derived nitrogen in its input with rainfall – 73.76 to 77.91% . Dust played the dominant role also in the total input of nitrogen from the atmosphere in the industrialized region studied. The dissolved and undissolved forms carried by it represented from 75.83 to 80.28% of the total amount of nitrogen input (Table 5).

An experiment has been carried out which has fully confirmed an enriching effect of dust on rainfall, as well as its dominant role in determining the size of nitrogen input from the atmosphere (Fig. 7). It has demonstrated that the washing-out of dissoluble nitrogen from dust was complete and independent of the amount of water passing through it (Table 7). It has also proved that even simple structures, such as vertical screens fixed above traps, can increase dust fall, and thereby also the input of dissoluble nitrogen (Table 6, Fig. 7).

The relationships found during the study allow theoretical calculations of the amounts of nitrogen input from the atmosphere owing to air-borne and falling dust, intercepted by tree crowns, from their rough quantitative estimates, which may prove useful in the calculation of nitrogen budget for forest ecosystems in highly industrialized areas.

6. POLISH SUMMARY

Badano rolę pyłu w kształtowaniu dopływu całkowitego azotu z atmosfery w strefie silnie uprzemysłowanej. Zastosowana metodyka pozwoliła na oszacowanie opadu pyłu i koncentracji w nim azotu, jak również określenie dojsścia pierwiastka w roztworach. Badania prowadzono w latach 1981–1983 w Rybnickim Okręgu Węglowym.

W okresie tym obserwowano dość dużą zmienność dopływu azotu w wodach deszczowych, tak miesięcznego (rys. 2) jak i sezonowego (tab. 1). Wartości te zawierały się w granicach od 0,104 do 6,750 kg · ha⁻¹ · miesiąc⁻¹. Sumaryczne sezonowe dojsście pierwiastka w roztworach zmieniało się od 7,64 do 20,77 kg · ha⁻¹.

Stwierdzono też występowanie zmienności w miesięcznym – 22,80 do 72,66 kg · ha⁻¹ – i sezonowym – 252,69 do 314,64 kg · ha⁻¹ – opadzie pyłu (tab. 2). Wahaniom ulegały również koncentracje nierozpuszczalnych związków azotu w pyłe – od 0,27 do 0,99% (tab. 2). W wyniku występowania tego zróżnicowania miesięczny dopływ pierwiastka w formie nierozpuszczonej wynosił od 0,084 do 0,468 kg · ha⁻¹ (rys. 4), a sezonowy – 0,93 do 1,78 kg · ha⁻¹ (tab. 2).

Z otrzymanych danych wynikało, iż w dopływie azotu z atmosfery dominowało jego przekazywanie w postaci rozpuszczonej w wodach deszczowych – 89,15 do 92,11% – nad formą nierozpuszczoną – w pyłe.

Zanalizowano metodą regresji wielokrotnej wpływ czynnika hydrologicznego i opadu pyłu na dopływ azotu w wodach deszczowych (równanie (3)). Stwierdzono, iż wielkość opadu deszczu nie oddziałuje na dojsście pierwiastka w postaci rozpuszczonej w sposób statystycznie istotny (tab. 4). Natomiast istotny statystycznie wpływ na wartość tego parametru miał opad pyłu (tab. 4). Był on dodatnio sprzężony z dopływem azotu w roztworach. Wymywanie tego pierwiastka z pyłu kształtowało się na poziomie 7,11% jego masy (równanie regresji (2) – rys. 6).

Na podstawie otrzymanego współczynnika wymywania azotu z pyłu obliczono teoretycznie udział pierwiastka pochodzenia pyłowego w dopływie w wodach deszczowych – 73,76 do 77,91%. Również w całkowitym dojsście azotu z atmosfery w tym uprzemysłowanym regionie pył odgrywał dominującą rolę. W formie rozpuszczonej i nierozpuszczonej niósł on ze sobą 75,83 do 80,28% całkowitej ilości dochodzącego pierwiastka (tab. 5).

Dokonany eksperyment potwierdził w pełni wzbogacający w azot wpływ pyłu na wody deszczowe i jego dominującą rolę w kształtowaniu dopływu pierwiastka z atmosfery (rys. 7). Wykazał, że wymywanie jego rozpuszczalnych form z pyłu było całkowite i niezależne od wielkości przepływu wody (tab. 7). Udowodnił także, iż nawet najprostsze formy przestrzenne (pionowe ekrany) umieszczone ponad pułapkami powodują zwiększenie opadu pyłu i, co się z tym wiąże, zwiększenie dopływu azotu w postaci roztworów (tab. 6, rys. 7).

Stwierdzone w pracy zależności pozwalają na teoretyczne obliczanie ilości azotu dochodzącego z atmosfery dzięki pyłowi opadającemu i unoszącemu się, a przechwytywanemu przez korony drzew, tylko na podstawie ich ilościowego szacunku – co wydaje się nie bez znaczenia przy obliczaniu bilansu azotu w ekosystemach leśnych na terenach silnie uprzemysłowanych.

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