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CHANGES IN PHYSICOCHEMICAL PROPERTIES OF SOILS IN THE AREA AFFECTED BY LIME INDUSTRY

ZMIANY WŁAŚCIWOŚCI FIZYKOCHEMICZNYCH GLEB W REJONIE ODDZIAŁYWANIA PRZEMYSŁU WAPIENNICZEGO

Abstract: Lime and cement industry belongs to the best developed branches of industry in Opole Voivodeship. The plants are located in eastern and central part of the province, in the vicinity of the city of Opole and the towns of Krapkowice and Strzelce Opolskie due to the location of the resources of calcareous raw materials (limestones and marls).The impact of lime and cement industry on the natural environment, especially soils, was studied by many authors, but these works do not concern the Province of Opole, which is one of the main centers of lime and cement industry in Poland for centuries. The article is an attempt to show changes in physicochemical properties of soils being under long lasting influence of alkaline dust emissions coming from the lime plant in Gorazdze. The research was conducted in the years 2010–2013. The range of the studies included field works in the areas adjacent to the plant, where 12 representative soil pits were arranged to collect samples for laboratory analyses. The following physicochemical parameters were determined in the samples: grain size distribution, reaction (pH), electrical conductivity and the content of calcium carbonate. The studies showed deacidification of the tested sandy soils resulting from alkaline dust deposition, which primarily concerned forest stands, which were characterized by the rise in reaction by 3–4 pH units. In the case of meadows, arable soils and wastelands, the pH values raised by 1–2 units. Moreover, alkaline falling dusts enriched the investigated soils in calcium carbonate. It concerns to the greatest extent the soil pits located nearest to the lime plant, where above 1% of $CaCO_3$ was found. The results of conductivity measurements proved low salinity of the investigated samples.

Keywords: lime dusts, proper rusty soils, pH, electrical conductivity, calcium carbonate content

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Introduction

The Province of Opole is an important center of cement and lime industry in Poland due to the presence of high-quality deposits of carbonate raw materials (limestones and marls) in the central and eastern parts of the region. The lime manufacture is the oldest branch of industry in Opole Province, concentrated in the area of Tarnow Opolski, Gorazdze, Gogolin and Strzelce Opolskie, where rich deposits of the Triassic limestones – raw materials for lime production – occur. At the end of the twentieth century, there operated six departments of lime manufacture, and these days there operate two, situated in Tarnow Opolski and Gorazdze.

By the end of the last century lime burning took place in shaft furnaces, and now mainly in modern regenerative Maerz type kilns. Lime plants produce quicklime (burnt lime) ground and in lumps, hydrated lime, calcium fertilizers (in the form of oxides, magnesium oxides and carbonates), calcareous aggregates, fodder and painting chalks, calcareous and glass flours, sorbents for flue gas desulfurization, limestone and special products.

The process of lime burning has a long tradition in the Opole Province dating from the 16th century, so the lime dust has impacted on soils of the Opole region for about 500 years, which is not without effect on their composition and properties. This is confirmed by the study of soils of Opole Province forest reserves, being under the influence of cement and lime industry, which showed increased magnetic susceptibility and the content of heavy metals and proved technogenic origin of the magnetic susceptibility, which was correlated with the content of metals [1, 2].

For decades, the main source of lime dust were shaft furnaces, which did not have any dust collection equipment and posed environmental hazard [3, 4]. According to the archive data, in the 90s of the last century lime plants emitted about 1000 Mg of dust per year, of which more than half came from shaft furnaces. The largest share in dust pollution of the atmospheric air had the lime plant in Gorazdze, hence the dust from this plant should be considered the most important from the point of view of environmental impact. Along with the increase in dust emissions, the increase in dust deposition was noted. Analyzing historical data it can be seen, that around the lime plants the allowable standard value of the fall of dust 200 $g \cdot m^{-2} \cdot yr^{-1}$ was significantly exceeded [5]. The impact on the quantity and quality of the falling dust was also close proximity to the cement plant that used raw materials coming from the same deposits.

The aim of the research, presented in this work, carried out in the years 2010–2013, was assessment of the impact of lime dust on the rusty soils in the vicinity of the lime plant in Gorazdze, by determination of changes in the physico-chemical properties of soils and diversity of features of genetic horizons. An attempt has also made to determine the extent of the impact of emitted lime dust in the selected transects.

Despite the fact, that the Province of Opole is one of the main centers of lime industry in Poland for about 500 years, the research into the impact of lime dusts on soils has not been conducted in this region so far, but mostly in the so called "White Basin" – the Swietokrzyski region.

Characteristics of the research object

The object of the study was the lime plant in Gorazdze, situated about 15 km to the south from the city of Opole, in Gorazdze village (Gogolin municipality). Nearby the plant (approximately 2 km in a westerly direction) the Gorazdze Cement Plant – the largest and most modern in Europe – is located. In the vicinity of both plants woodlands extend and in close proximity (around 5 km to the southeast from the lime plant), the Landscape Park „St. Anna's Hill" and nature reserves are situated.

The process of lime burning for decades was carried out in shaft furnaces. They were fired with the coke coming from, inter alia, Zdzieszowice Coking Plants. The fuel was introduced into the furnace together with the limestone and burned in direct contact with it. It was not until the end of the twentieth century that shaft furnaces were liquidated and two modern devices for lime burning: Maerz regenerative kiln, with a capacity of 600 Mg of lime per day and Gopex kiln – 350 Mg of lime per day. New kilns are fired with the coke oven gas or natural gas, and Maerz kiln is equipped with fabric dust collector, which resulted in the reduction in the amount of emitted dust.

According to archive data, the plant had a big share in the emissions of lime dust in the Opole Province, which amounted to almost 60%, wherein approximately 70% of lime dust emissions came from shaft furnaces [5]. According to the information provided by the lime plants, in the last decade dust emissions were high despite the modernization of technological processes. The highest values (from 81 to 157 $Mg \cdot yr^{-1}$) was recorded in the years 2007–2009, which was associated with increased, during this period, market demand for lime.

Research methods

Soil pits were located nearby the lime plant in Gorazdze, in the area of Strzelce Opolskie Forest District, the municipality of Gogolin. In order to choose the right places for the soil pits, the directions of the prevailing winds blowing in this region (W, NW, SW) and the distance from the plant were taken into account. The topographic map with a scale of 1 : 5000 was used. Soil pits were situated in relation to the plant in three transects of research:

– western (W), which included soil pits marked with symbols: G1, G2 (on grasslands), A3 (on arable soils), and F4, F5, L6, F7 (in forest areas), at a distance of 70 to 430 m from the plant;

– northwestern (NW), including soil pits marked with symbols: A10 (on arable soils), and F11 and F12 (in forest areas), at a distance of 300 to 470 m from the plant;

– southwestern (SW), which included soil pits W8 and W9, located on wasteland, at a distance of 80 to 120 m from the lime plant.

The total number of research points was 12, and the total number of soil samples collected at these points to a depth of 60 cm equaled 51. Representative samples were collected from each genetic horizon. After bringing to the laboratory, the samples were dried in trays to air-dry state, then crushed and homogenized in Retsch agate mill. Afterwords, in samples prepared this way four parameters that characterize the physicochemical properties of the soils were determined with methods commonly used in soil science [6, 7]:

– the particle size distribution – by Casagrande areometric method in Proszynski's modification (silt and clay fractions), and by sieving method (sand fractions);

– reaction (pH) – by potentiometric method, the mass ratio of the soil to the water in the suspension equaled 1 : 2.5;

– specific conductivity – conductrometrically, the mass ratio of the soil to the water in the suspension was 1 : 5;

– the content of calcium carbonate – by Scheibler volumetric method.

The tested soils were classified according to the latest international and Polish soil classsification systems [8–11].

Results and discussion

Profile structure and particle size distribution of soils

In the course of field research it was found, that in the tested area *Rusty soils* (WRB: *Brunic Arenosols*) occurs, the subtype of proper rusty soils, with the following structure of profile: Ol-Ofh-Ah-Bv-C (soils of forest areas), A1-A2-Bv-C (soils of grasslands) and ApBv-Bv-C (arable soils). It is known from the literature, that in natural conditions such soils are formed from the poor in nutrients, carbonateless sands. They are characterized by acid reaction with the dominance of the acidifying processes, *ie* leaching, washing, podsolisation, low sorption capacity and small buffering capacity [8–12].

As a result of grain size distribution analysis one granulometric group-sands, and three subgroups (loose sands, weakly loamy sands and loamy sands) were distinguished (Table 1). A characteristic feature of granulometry of the tested soils is the presence of sand fractions only (coarse sands) throughout the soil profile, to an average depth of 50 cm. The differences that occur in the composition of the granulometric subgroups arise from lithogenic properties of substrate (bedrock), relief and soil forming processes, affecting the soil substrate [8–11].

Table 1

Schematic layuot of grain size distribution in the tested soils

Particle size distribution arrangement		Number of soil pit
Homogeneous	loamy sand	G2
	sand	F5
	sand	W9
Heterogeneous, binary	sand/loamy sand	F4, F7, F11, F12, W8
	loamy sand/sand	A10
Heterogeneous, ternary	loamy sand/sand/loamy sand	G1
	loamy sand/sand/sand	A3
	sand/loamy sand/sand	L6

Reaction (pH)

The analyzed soils are hundreds of years exposed to the alkaline dust emissions. Dusts introduced into the atmosphere by the lime plant in Gorazdze are characterized by a very high pH values in the range of 11.49 to 12.54, which affects the course of pedogenic processes in the tested soils (Table 2) [5]. Alkaline reaction of dusts results from the predominance of calcium compounds, present as calcium oxide CaO (approximately 54.9%) and calcium carbonate $CaCO_3$ (approximately 40%). Other elements have a much smaller share in the dusts, which takes the following values (in terms of the oxide forms): 2% of SiO_2, 1% of MgO, 0.4% of Fe_2O_3, 0.2% of Al_2O_3 and 0.1% of SO_3.

As a result of the alkaline deposition, naturally acidic rusty soils underwent deacidification in the whole profile, in the case of soil pits situated on grasslands to a depth of 45 cm, on arable soils to 50 cm, on wastelands to 50 cm, and on forest soils up to 50 cm. In the case of forest soils (pits marked F4–F7, located in a westerly direction from the plant and F11 and F12 – in NW), the most transformed proved to be two genetic horizons: detrital Of (pH 6.0–7.25) and humic Ah (pH 7.07–7.27) (Table 2). Moreover, in some places unnaturally high pH values in *sideric* horizon (Bv), with pH values up to 7.06 and in the bedrock – to 6.16. Taking into account that the pH values of forest soils in Strzelce Opolskie Forest District range from 3.1 to 4.2 [1], the tested forest stands are characterized by an increase in pH by 3–4 units. The reaction of litter subhorizon Ol of the tested forest soils is lower, than detrital O_f, occurring below (pH 5.39–6.00), however the difference in pH values among tested points in vicinity of the plant reaches up to two pH units. An increase in acidity of Ol subhorizon reported in forest stands may be associated with higher concentration of H^+ ions, resulting from the decomposition of debris of forest undergrowth plants and pine needles [12, 13]. Only in the case of C horizon of forest soil pits L6, F7 and F11, the pH values were within the limits given for the rusty soils under coniferous forest stands, free from strong anthropogenic pressure.

Forest soil pits located to the west of the plant, show a tendency to decrease in alkalization deep into the soil profile, whereas there was no decrease in soil reaction with increasing distance from the plant, as regards especially humic and sideric horizons. This may be related to differences in the composition of the falling dusts, as well as a diverse species composition of undergrowth and different distance between tree crowns.

The reaction of A3 and A10 soil pits, located on arable land, in the distances of 220 m and 300 m from the plant, in line with prevailing winds, is characterized by pH values above 7.0. This applies to the entire depth of the soil profile of these pits. In A3 position a slight decrease in pH values with depth was noted. The difference between the highest A1 horizon (0–5 cm), and the bedrock C (45–55 cm) equals 0.14 pH units. Different results were obtained for A10 position, where pH values do not decrease with the depth, and the lowest value occurs in A1 surface horizon. Taking into account, that most arable soils in Opole Province have pH below 5.5 [1], and the optimal pH of the soil for plant cultivation should be in the range of 5.5–6.5 [10–12], the investigated case indicates the exceedance of pH value almost by 1 unit.

Soil pits located on grasslands, overgrown with grassy vegetation (L1 and L2) and on wastelands (W8 and W9), are characterized by an increased reaction throughout entire depth. In the case of meadow pits Ł1 and Ł2, located at the following distances from the plant: 70 m and 170 m respectively, in line with the prevailing winds, pH values of soil horizons vary in the range of 7.4–7.7. Wasteland stands W8 and W9, situated at distances: 80 m and 120 m from the emitter, are characterized by lower reaction of soil horizons, with pH values in the range of 7.0–7.5. The optimum pH values for grassy vegetation growth on mineral soils range from 5.5 to 6.5 [10–12], which indicates the exceedance of pH limit value from 0.5 to 1 pH unit in the tested soil pits.

Alkalization of soils is one of the best known and widely described in the literature results of the impact of cement and lime dusts on the environment. The falling dusts, deposited on the soil surface, are the reason for breakage soil protective barriers, arising from its buffer properties. Numerous studies have shown changes in functioning of a variety of soil buffers, which does not allow to maintain the soil pH in a natural range [11–14].

Particularly noteworthy are the work concerning chemical transformations of forest soils under the long-term deposition of alkaline dusts coming from cement and lime plants in the Swietokrzyski region of Poland [15]. The author showed strong alkalization of soils, manifested in changes in their buffering properties, and the content of such metals as: Ca, Mg, Mn, Al and Fe, which led to the transformation of plant communities. She also described the transformations which the alkaline falling dusts are subjected to in the upper soil horizons. It is the process of decomposition, which releases and mobilizes dust components into the soil profile, as well as the process of binding the dusts of hydrophilic properties in poorly soluble compounds. Surface horizons of forest soils (Of, Oh, Ofh) form the filter catching alkaline dust particles, which significantly reduces their migration to deeper horizons.

The content of carbonates

The content of calcium carbonate $CaCO_3$ in the tested soils ranged from 0.0 to 3.06% (Table 2). It was fund, that the amount of this component decreases with the depth of sampling in the soil profile, and depends on the distance from the plant, which may be related to the deposition of lime dusts.

Table 2

Physicochemical properties of the tested soils

Symbol of soil pit	Location of soil pit against the plant	Depth of sampling [cm]	Soil horizon	pH [-]		Content of $CaCO_3$ [%]	Electrical conductivity [$\mu S \cdot cm^{-1}$]
				H_2O	KCl		
G1	70 m W	0–5	A1	7.84	7.41	2.23	116.5
		25–45	C	8.07	7.72	0.10	50.1
		< 45	C	8.09	7.63	0.08	48.5
G2	170 m W	0–5	A1	7.84	7.44	3.06	145.7
		15–25	A2	7.87	7.57	1.39	115.1

Table 2 contd.

Symbol of soil pit	Location of soil pit against the plant	Depth of sampling [cm]	Soil horizon	pH [-]		Content of $CaCO_3$ [%]	Electrical conductivity [$\mu S \cdot cm^{-1}$]
				H_2O	KCl		
A3	220 m W	0–5 0–30 45–55	A1 Ap C	7.45 7.47 7.64	7.10 7.01 6.96	0.27 0.31 0.02	58.3 57.1 40.5
F4	230 m W	3–2 2–0 0–15 40–60 < 60	Ol Ofh Ah ABv C	6.11 7.15 7.68 7.26 6.92	6.00 7.15 7.16 6.67 6.16	— — 0.17 0.06 0.02	321.0 167.5 102.3 43.9 24.8
F5	280 m W	3–2 2–0 0–20 20–40 < 40	Ol Ofh Ah Bv C	5.99 7.26 7.76 6.73 6.82	5.88 7.25 7.27 5.95 5.92	— — 0.21 0.02 0	288.3 132.8 90.9 28.1 28.6
L6	330 m W	7–6 6–0 0–10 10–25 < 25	Ol Ofh Ah Bv C	5.78 6.57 7.70 7.28 6.09	5.46 6.65 7.22 6.75 4.94	— — 1.25 0.02 0.02	317.3 207.7 145.7 51.6 40.0
F7	430 m W	6–5 5–0 0–10 10–25 < 25	Ol Ofh Ah Bv C	5.57 6.69 7.77 6.82 4.80	5.42 6.50 7.23 6.07 4.31	— — 0.31 0.02 0.04	313.0 145.4 106.8 38.2 47.0
W8	80 m SW	0–5 0–25 25–40 < 50	A1 A2 Bv C	7.65 7.75 7.68 7.76	7.28 7.35 7.09 7.06	0.86 0.11 0.04 0.02	84.3 78.8 39.5 30.6
W9	120 m SW	0–5 10–25 25–40 < 50	A1 A2 Bv C	7.55 7.78 7.80 7.80	7.18 7.48 7.37 6.97	0.52 0.21 0.11 0	141.9 70.5 52.4 27.2
A10	300 m NW	0–5 25–35 < 50	A1 Ap C	7.67 7.92 7.91	7.22 7.44 7.34	0.82 0.35 0.02	96.0 66.4 36.9
F11	400 m NW	3–2 2–0 0–10 10–25 < 50	Ol Ofh Ah Bv C	5.39 7.16 7.41 5.62 5.12	5.39 7.16 7.07 5.13 4.53	— — 0.73 0.02 0	358.7 169.3 236 293.3 252.7
F12	470 m NW	3–2 2–0 0–10 10–25 < 50	Ol Ofh Ah Bv C	5.83 6.78 7.72 7.68 6.79	5.78 6.87 7.20 7.06 6.16	— — 0.79 0.06 0.04	276.7 140.9 115.5 45.9 29.8

In the top layer of all tested soil pits the content of $CaCO_3$ exceeded the value of 0.1%. In the case of grassland and wasteland it concerned the layer of 0–25 cm, arable soil 0–35 cm and forest soil 0–10 cm. As it is known from the literature, when carbonate content exceeds 0.5%, the saturation of the sorption complex with alkaline cations, mainly calcium ions, may take place in organic and organo-mineral soil horizons [15]. In the case of tested soils, $CaCO_3$ content above 0.5% was noted in the upper horizons, mainly turf (A1) and humic (A2, Ah, Ap), but it concerned only some of the tested pits. The greatest amount of $CaCO_3$ were accumulated in soils closest to the lime plant, *i.e.* G1 and G2 pits.

Electrical conductivity

The results of specific conductivity measurements indicate, that alkali emissions had much less impact on the salinity of the tested soils, than on reaction (pH values) and carbonate content. Analyzed soil samples were characterized by low specific conductivity values, much less than 1000 $\mu S \cdot cm^{-1}$ (Table 2).

The tested proper rusty soils of agricultural and forest land in the vicinity of lime plant were characterized by diverse values of conductivity. The results obtained for individual soil samples ranged from 24.8 to 358.7 $\mu S \cdot cm^{-1}$. The highest values were noted in the following horizons: litter Ol: 276.7–358.7 $\mu S \cdot cm^{-1}$ (forest pits F4–F7, F11 and F12), detrital Of: 140.9–207.7 $\mu S \cdot cm^{-1}$ (forest pits F4–F7, F11 and F12) and turf A1: 84.3–145.7 $\mu S \cdot cm^{-1}$ (soil pits G1 and G2 on grasslands, and W8 and W9 on wastelands). It was also noted the increased conductivity values in humic horizon of forest soils (90.9–236 $\mu S \cdot cm^{-1}$, soil pits: F4–F7, F11 and F12) and soils overgrown with grassy vegetation (70.5–115.1 $\mu S \cdot cm^{-1}$, soil pits: G2, W8 and W9).

For most of the tested soil samples the values of specific conductivity have been decreasing with the depth of the occurrence of horizons they were collected from. An exception was the soil pit F11, where the conductivity values of mineral horizons exceeded 250 $\mu S \cdot cm^{-1}$. The increased salinity of the entire soil profile of F11 location may result from the impact of both the alkali emissions, as well as other anthropogenic factors.

There was no the decrease of specific conductivity values with increasing distance from the plant. It was noted however, that the soil pits located in line with prevailing winds, *ie* in a westerly and north-westerly direction from the plant, were characterized by the highest salinity. Most of all it refers to the forest soil pits, showing the greatest salinity in the upper horizons. The increase in the conductivity of the upper soil layer (0–2 cm) in forest soils may result from atmospheric precipitation flowing down the tree trunks. Diverse values of conductivity of the tested soils and no relationship with the distance from the plant may also be the effect of differences in the composition of the falling dusts. This is confirmed by the research into the dusts from the lime plant in Gorazdze, which are characterized by very different values of the specific conductivity in the range from 355 to 7060 $\mu S \cdot cm^{-1}$, depending on the place of their sampling [5]. The values obtained for the dusts collected in a nearby cement plant in Gorazdze were within the range recorded for the lime plant.

Conclusions

1. Long-term deposition of alkaline dusts caused the chemical transformations of the tested soils. This concerns particularly reaction (pH), and – to a lesser extent – carbonate content and salinity.

2. Alkaline dusts were the reason for the change of pH values of the tested soils from acidic to neutral or alkaline in all transects of the reaserch (W, NW, SW).

3. The increased pH values was observed mainly in the upper soil horizons (detrital, turf) and humic horizons, as well as – to a lesser extent – in sideric horizon and bedrock, to a depth of about 50 cm.

4. Lime dusts enriched in calcium carbonate mostly turf and humic horizons of grassland soils, where up to 3.06% $CaCO_3$ at pH values above 7.41 were found.

5. There was a relationship between pH values and calcium carbonate content in the genetic horizons of the tested soil pits, *ie* an increase in pH values is accompanied by an increase in the content of $CaCO_3$.

6. The tested proper rusty soils have lost their natural properties and were transformed as a result of the impact of alkaline dusts emitted from the lime plant. Only the profile structure and particle size distribution were not changed – these soil characteristics remained typical of proper rusty soils.

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ZMIANY WŁAŚCIWOŚCI FIZYKOCHEMICZNYCH GLEB W REJONIE ODDZIAŁYWANIA PRZEMYSŁU WAPIENNICZEGO

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Abstrakt: Przemysł wapienniczy i cementowy należy do najlepiej rozwiniętych gałęzi przemysłu w województwie opolskim. Zakłady zlokalizowane są we wschodniej i środkowej części regionu, w sąsiedztwie miast: Opole, Krapkowice i Strzelce Opolskie, co wynika z występowania na tym terenie złóż surowców wapiennych (wapieni i margli). Wpływ przemysłu wapienniczego i cementowego na środowisko przyrodnicze, szczególnie gleby, był przedmiotem badań wielu autorów, ale prace te nie dotyczą województwa opolskiego, które od stuleci jest jednym z głównych ośrodków produkcji wapna i cementu w Polsce**.** W artykule podjęto próbę ukazania zmian właściwości fizykochemicznych gleb znajdujących się pod wieloletnim wpływem emisji pyłów alkalicznych pochodzących z zakładu wapienniczego w Górażdżach. Badania prowadzono w latach 2010–2013. Zakres badań obejmował prace terenowe w rejonie zakładu, gdzie założono 12 reprezentatywnych odkrywek glebowych, z których pobrano próbki do analiz laboratoryjnych. Dla każdej z próbek glebowych oznaczono następujące parametry fizykochemiczne: skład granulometryczny, odczyn (pH), przewodnictwo właściwe oraz zawartość węglanu wapnia. Badania wykazały wzrost odczynu badanych gleb piaszczystych będący efektem depozycji pyłów alkalicznych, co dotyczyło przede wszystkim odkrywek zlokalizowanych na stanowiskach leśnych, które charakteryzowały się wzrostem wartości pH o 3–4 jednostki. W przypadku łąk, gleb uprawnych oraz nieużytków wartości pH wzrosły o 1–2 jednostki. Ponadto wykazano, że opadające pyły alkaliczne wzbogaciły badane gleby w węglan wapnia. Dotyczyło to w największym stopniu odkrywek glebowych zlokalizowanych najbliżej zakładu wapienniczego, w których zawartość $CaCO_3$ przekraczała 1%. Wyniki pomiarów przewodnictwa właściwego wskazują na niski stopień zasolenia badanych próbek glebowych.

Słowa kluczowe: pyły wapiennicze, gleby rdzawe właściwe, pH, przewodnictwo właściwe, zawartość węglanu wapnia