Carpathian Journal of Earth and Environmental Sciences, August 2021, Vol. 16, No. 2, p. 507 – 512; DOI:10.26471/cjees/2021/016/195

DOES THE CEMENT AND LIME INDUSTRY POSE A THEAT FOR THE ENVIRONMENT – CASE STUDY: TRZUSKAWICA S.A. LIMESTONE PLANT, SWIĘTOKRZYSKIE MTS., POLAND)

Anna ŚWIERCZ
Institute of Geography and Environmental Sciences, Jan Kochanowski University in Kielce, Poland
e-mail: swierczag@poczta.onet.pl

Abstract: The aim of this research was to evaluate the influence of cement and lime dust on some properties of podzolic soils developed around a limestone plant located in Trzuskawica. The research was carried out on samples collected from four forest soil profiles in 2019. In the soil samples, particle-size composition, pH, total organic carbon, organic nitrogen, cationic sorption capacity, $CaCO_3$ content, and total concentrations of selected trace metals were determined. The analysed soils are characterised by particle-size composition corresponding to loose and slightly clayey sands. The surface horizons' pH is neutral and alkaline (pH_{KCl}> 6.5) which results from their enrichment in $CaCO_3$. The enrichment of soils by trace metals such as Pb, Zn, and Sr, especially in the surface horizons, was observed. Under the influence of alkalisation, a clear transformation of the physical and chemical properties of Haplic Podzol soils which lost their original features (acidic soils typical of oligotrophic habitats), was found. Although the cement and lime industry does not pose a major environmental threat today, the effects of past activities are still present in the soil cover.

Keywords: alkalisation, cement plant, physicochemical properties

1. INTRODUCTION

Poland is one of the leading cement producers in Europe (18.940 million tonnes in 2018 - data from the Central Statistical Office of Poland, 2018), taking the third place, behind Germany and Italy. Lime production amounting to 1,954 thousand tonnes (2018) places Poland in the fifth place. A significant part of these products is produced by the plants located within the so-called Białe Zagłębie in the Świętokrzyskie Mountains. The Białe Zagłębie houses cement plants in Nowiny and Małogoszcz as well as limestone plants in Bukowa and Trzuskawica. In 2017-2019, Trzuskawica S.A. limestone plant had the largest share in dust emissions into the atmosphere, producing over 132 thousand tons of dust per year. Despite numerous improvements in the reduction of dust emissions (fabric filters, cyclones, electrostatic precipitators), decades of operation of cement and lime plants in this area resulted in spectacular changes in ecosystems (Duszak et al., 2015, Świercz, 2005, Kozłowski et al., 2020). The cement and lime industry emits large amounts of dust into the atmosphere, which is not classified as a toxic substance, but affects the biological state of the environment (Mandre & Lukjanova, 2011, Dybey, 2013, Paal et al., 2013, Kumar et al., 2008). These are mainly emissions of alkaline pollutants which affect changes covering entire ecosystems. The immission of dust to soils violates their chemical properties, primarily causing a change in pH and species composition of living organisms (Mandre & Lukjanova, 2011, Paal & Degtjarenko, 2015, Barga-Więcławska & Świercz, 2015). It causes changes in a tree stand by reducing the efficiency of photosynthesis, transpiration, and the level of chlorophyll, as well as leads to remodelling and species change of entire phytocoenoses (Świercz, 2005, Kumar et al., 2008, Paal & Degtjarenko, 2015, Dąbkowska-Naskręt et al., 2006, Greszta et al., 2002). Along with prolonged exposure to alkaline dust, an effect similar to "liming" of soils occurs (Świercz, 2005, Nilsen, 2001; Ambulkar et al., 1995, Shivkumar & John, 1995).

The aim of the research was to assess the effect of cement and lime dust on some properties of Haplic Podzol and Albic Brunic Arenosol in conditions of reduced alkaline emission located in the vicinity of Trzuskawica S.A. limestone plant.

2. RESEARCH METHODS

Four soil profiles (Figure. 1) described in detail according to the methodology (including Bednarek et al., 2004) were selected for the research. During field works, the thickness of individual sub-horizons, grain-size composition, colour based on the Standard Soil Colour Charts acc. to Munsell, and the type of soil humus were determined. The soil samples for research were collected from each genetic sub-horizon. The samples were dried at room temperature to air-dry state. After drying, the mineral samples were ground in a porcelain mortar using a pestle and then sieved through a soil sieve with a mesh diameter of 2 mm. The organic samples were crushed with a Fitch laboratory grind. The following determinations were made in the samples (acc. to Karczewska & Kabała, 2008):

- a grain-size composition of earthy parts by sieve analysis and Casagrande's areometric method modified by Prószyński (Karczewska & Kabała, 2008),
- soil pH by potentiometric titration (pH H_2O, pH K_{Cl}),
- organic carbon (C_{org}) by Tiurin and Alten's methods modified for horizons of humus,
- total nitrogen (TN) by modified Kjeldahl's method,
- hydrolytic acidity Hh by Kappen's method in 0.5 M $Ca(CH_3COO)_2$ solution,
- sum of exchangeable alkaline cations (S) by Kappen's method in 0.5M NH_4Cl solution at pH 8.2.
- acidity Hw by Sokolov's method
- total content of selected trace metals (Cr, Cu, Ni, Pb, Sr, Zn) by AAS method (in a process of sample mineralisation - 1g taken from mineral horizons, 0.5g taken from organic horizons in aqua regia, dilution 1:5, AAS Unicam Solar 939)
- forest soil trophism index (FSTI) based on the weighted sum of indicators: dust fraction, floating parts, pH, cation exchange capacity expressed by a volume unit cmol/dm^3) and a C:N ratio (Brożek, 2001).

3. RESEARCH RESULTS AND DISCUSSION

The analysed soils were located in the immediate vicinity of Trzuskawica S.A. limestone plant (Figure. 1), between 300 m and 1.5 km from the dust emitter.

All tested profiles were made in the habitats of fresh coniferous forests and fresh mixed coniferous forests (Table 1). The tree stand is dominated by (approx. 60-year-old) Scots pine.

The analysed soils are deep (80-130 cm). Strong anthropogenic pressure did not cause changes in the sub-type of natural soils - all horizons are properly formed and have a thickness characteristic of Haplic Podzol soils and the presence of diagnostic horizons enabling their diagnosis in the field (Table 1, Figure 2).

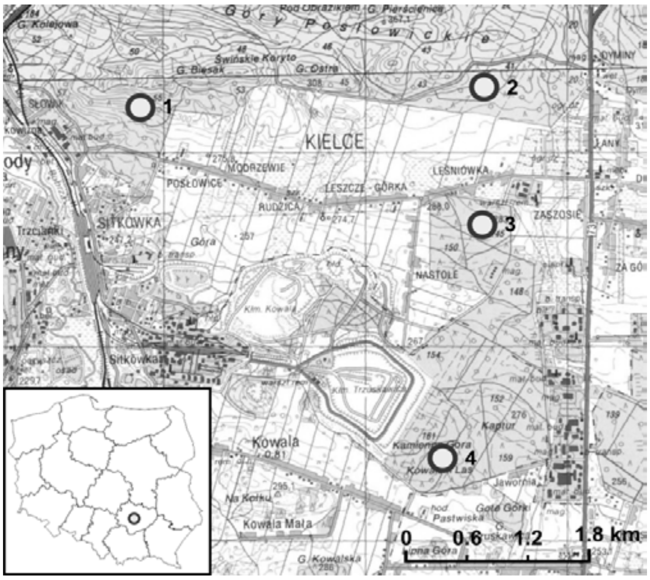

Figure 1. Location of sampling points (1-4 soil profiles) on the base of the topographic map.

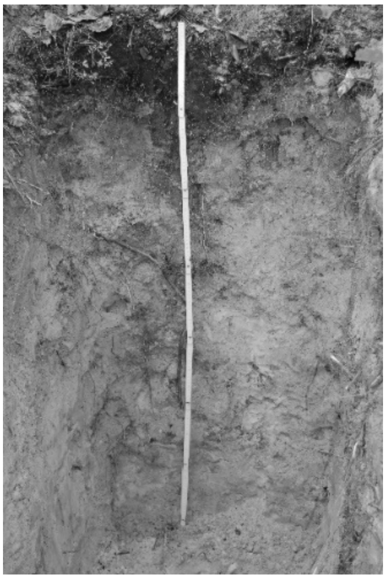

Figure 2. Haplic Podzol Ol-Ofh-Ees-Bhs-BC-C (profile no1) formed from sands.

Table 1. Location data of excavated soil pits. Names of soil horizons acc. to Polish Soils Classification 2008, types of soils acc. to WRB (World Reference Base) 2006 (PTG, 2008, IUSS, Working Group WRB, 2006)

Profile no	Location	Soil layer*	Latitude	Longitude	Height a.s.l	Soil type and subtype	Forest habitat type
1	Posłowice	Ol-Ofh-Ees-Bhs-BC-C	50.830932	20.561359	336.4	Haplic Podzol LWt	Bśw (fresh forest profile 1) and BMśw (mixed-conifer forest) with a predominace of pine in the stand
2	Posłowice	Ol-Ofh-AE-Bsv-Bv-C	50.832710	20.584274	320.0	Albic Brunic Arenosol RWbi	
3	Leśniówka	Ol-Ofh-Ees-Bhs-BC-C	50.818106	20.605292	277.2	Haplic Podzol LWt	
4	Kowalski Las	Ol-Ofh-AE-Bsv-Bv-C	50.800890	20.610113	274.1	Albic Brunic Arenosol RWbi	
*Ol-Ofh (Organic layer), A (Topsoil - mineral with humus), E (Eluviation layer - leached minerals and organic matter), B (Subsoil - deposited minerals and minerals salts), C (Parent rock, partially weathered parent material).							

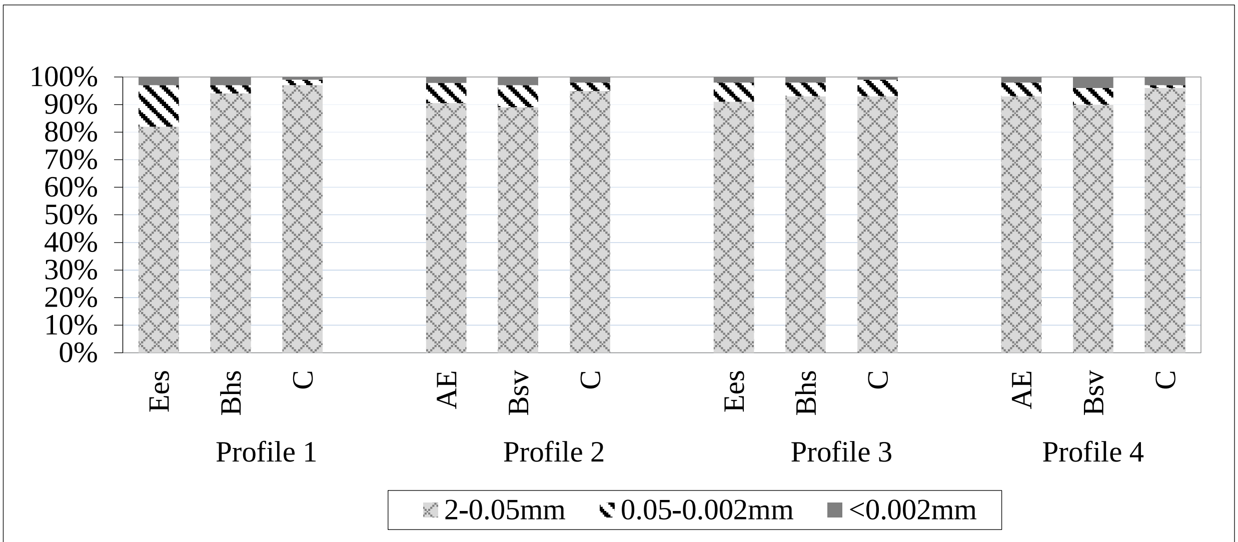

Figure 3. Grain size composition of the analyzed soils. (Explanations of soil layer as in Table 1)

The analysed soils have a similar particle-size composition. These are soils formed mainly of loose and slightly loamy sands with low content of clay fraction (Figure 3), which is a typical feature for Haplic Podzol soils. The results of research on the physical and chemical properties and the content of selected trace metals are presented in Table 2 and Figure 4, respectively.

Particularly noteworthy are the very high trophism indices calculated in the analysed soil profiles on the basis of the share of skeletal, floating (Ø <0.002 mm) and dust fractions, pH, humus decomposition degree and sum of alkaline cations, which indicate that the analysed soils are eutrophic and hypertrophic (Forest Soil Trophism Index in the range of 29.9-33.3; Table 2). Such an unnaturally high value of the FSTI - an increase in value by an average of 40% in relation to typical Haplic Podzol soils under pine stands for the analysed soils is mainly influenced by chemical properties (high share of exchangeable cations in a sorption complex and degree of decomposition of organic matter; Table 2).

Under the natural conditions of coniferous ecosystems, the Podzolic soils formed by sands are characterised by the FSTI in the range of 11-17, while the rusty soils with a grain-size composition similar to the analysed ones have the FSTI amounting to 14-17 (Świercz, 2005, Brożek, 2001). The most visible effect of the influence of cement and lime dust on the research area is the increased pH of KCl of soils, which was in the range of 4.85-7.62; while, the highest one was in the detritus sub-horizons and amounted to 7.15-7.62 (Table

Table 2. Selected chemical properties of soils.

Profile no	Soil profile	Depth	*FSTI	pH	pH	$CaCO_3$	Corg.	Nog.	Hw	Hh	S	V
		cm		H_2O	KCl	%			mmol(+)·kg $^{-1}$			%
1.	Ol	1	33.3	6.22	6.10	2.41	45.27	0.871	1.72	3.51	60.55	94.52
	Ofh	1-2		7.58	7.35	10.19	22.40	0.749	0.23	1.23	40.02	97.02
	Ees	3-10		7.14	7.10	1.20	1.312	0.147	0.02	0.48	6.81	93.42
	Bhs	20-30		6.00	5.90	0.35	0.243	0.032	0.2	1.23	2.77	69.25
	BC	30-40		5.82	5.13	0.00	0.125	0.010	0.23	1.51	1.13	42.80
	C	120-150		5.70	4.99	0.00	0.010	0.003	0.31	2.90	1.49	33.33
2.	Ol	2	31.2	6.20	6.09	12.55	48.33	0.891	1.17	9.81	55.74	85.04
	Ofh	1-3		7.20	7.15	21.30	22.43	0.711	0.2	1.42	52.41	97.36
	AE	5-15		7.15	7.50	5.0	0.961	0.069	0.04	0.48	17.28	97.29
	Bsv	40-65		6.80	6.22	0.00	0.070	0.007	0.15	1.18	1.39	54.09
	Bv	70-100		5.30	5.00	0.00	0.026	0.005	0.04	1.12	1.22	52.14
	C	120-130		5.15	4.97	0.00	0.010	0.003	0.64	0.98	0.97	49.74
3.	Ol	2	29.9	6.82	6.58	1.67	46.87	0.845	1.72	6.78	79.61	92.14
	Ofh	2-4		7.81	7.62	19.11	16.87	0.690	0.11	1.13	52.09	98.21
	Ees	10-15		7.73	7.21	2.52	1.091	0.037	0.04	0.98	12.43	92.69
	Bhs	30-45		6.34	5.11	0.22	0.098	0.016	0.2	1.31	2.29	63.61
	BC	100-110		6.11	5.00	0.00	0.065	0.012	0.14	0.96	1.56	61.9
	C	130-135		6.07	6.01	0.00	0.009	0.009	0.15	1.23	4.99	80.23
4.	Ol	3	30.0	6.66	6.53	6.81	45.33	0.912	1.82	4.88	81.77	94.36
	Ofh	1-2		7.84	7.42	19.92	19.21	0.653	0.09	1.33	56.53	97.70
	AE	5-15		7.11	6.75	5.16	0.909	0.082	0.18	0.88	14.99	94.46
	Bsv	15-25		6.75	6.48	0.30	0.082	0.015	0.25	0.59	1.92	76.49
	Bv	30-60		6.80	6.50	0.00	0.021	0.009	0.19	0.61	1.28	67.72
	C	90-120		5.34	4.85	0.00	0.009	0.005	1.31	2.66	1.69	38.85

*FSTI (Forest Soil Trophism Index)

2). Slightly lower values were recorded for the litter sub-horizon, but the values were higher than 6.09 there as well, which indicates their clear alkalisation. Similar observations were made by (Dąbkowska-Naskręt et al., 2006, Nilsen, 2001, Shivkumar & John, 1995, Canpolat et al., 2002).

Typical pH of the humus horizons of Podzolic soils is in the range 4.5-5.0 units (Świercz, 2005, Dybey, 2013, Dąbkowska-Naskręt et al., 2006). Podzolic soils do not contain carbonates and the analysed soils show their presence up to a depth of 45 cm (Table 3). The content of carbonates is significant and ranges from 0.22% (Bhs) to 21.3% in the detritus sub-horizon Ofh (profile 2). The parent material is free of carbonates, so $CaCO_3$ deposited in the surface soil horizons is anthropogenic (Dybey, 2013, Nilsen, 2001, Marsal, 2004).

The cement and lime dust contain mainly CaO (49-67%), with the content of approx. 45% in the raw material, i.e. Devonian limestones exploited from the Trzuskawica open pit (Świercz, 2005). Calcium oxide easily reacts with water to form $Ca(OH)_2$, which intensifies the effect of point alkalisation. Excessive amounts of calcium carbonate in soils reduces the absorption of iron, aluminium, boron, zinc, copper and manganese, causing retrogradation of their forms (Nilsen, 2001, Marsal, 2004).

Average monthly deposition of cement and lime dust in the vicinity of Trzuskawica S.A. limestone plant in 2019 was 20-35 g/m^2 and did not exceed the applicable standard. According to the regulation of the Minister of Environment (Journal of Laws of 2018, item 799), the permissible level of dust with a fraction of 10 microns - PM10 in the air is 50 g·m^3 (daily average), and the limit of permissible exceedances during the year amounts to 35 days.

Another noticeable soil feature is the high degree of saturation of soils with alkaline cations V (33.3-97.7%). The originally acidic and strongly acidic Podzolic soils enriched with anthropogenic carbonates changed their properties towards an increase in the share of alkaline cations in the S sorption complex (Table 3). This process is particularly visible in organic horizons. Typical physical and chemical properties for Podzolic soils are only found in parent rock horizons <90-100 cm.

Higher values in terms of pH, carbonate concentrations, and the degree of saturation with alkaline cations are characteristic of profiles located to the west of the dust emitter, which is consistent with the wind directions typical of this region (west, north-west). The content of soil humus and total nitrogen in the analysed soils is slightly lower (by 2-5%), but does not differ significantly from the average concentrations in Podzolic soils free from the influence of cement-lime dust (Table 2) (Greszta et al., 2002, Brożek, 2001, Marsal, 2004).

It is worth noting that in the soils exposed to excess liming, accelerated decomposition of soil humus may occur, which results in nitrate leaching (indirect nitrogen contamination of waters and their eutrophication) as well as total nitrogen loss in the horizons of overburden humus. Under extreme conditions, humus takes the calci-kseromull form being unnatural for oligotrophic forest habitats (Barga-Więcławska & Świercz, 2015, Ambulkar et al., 1995). Physico-chemical properties and (litter sub-horizon-detritus sub-horizon) structure allow for classifying all analysed humus levels to this type (Table 1, Figure 2).

Soils being excessively limed show an increase in the content of toxic trace metals: chromium, cadmium, cobalt, and sometimes lead, zinc and strontium (Świercz, 2005, Greszta et al., 2002, Ambulkar et al., 1995, Shivkumar & John, 1995). In this aspect, long-term deposition of trace elements contained in the cement-lime dust is particularly important and this problem is often raised in the literature (Canpolat et al., 2002, Guo et al., 2012, Bartkowiak et al., 2017, Ahmad et al., 2005). Accumulation of trace metals such as Pb, Zn, Sr was found in the analysed soils (Figure 4).

The highest amounts of all analysed metals were found in the surface parts of litter sub-horizons and detritus sub-horizons (Pb 90-358 mg·kg^{-1}; Zn 127-132 mg·kg^{-1}; Sr 112-176 mg·kg^{-1}; Cr 8-16 mg·kg^{-1}; Cu 8-16 mg·kg^{-1}, respectively). These values, although increased, do not exceed the permissible content of substances causing environmental risk for forest land according to the Regulation of the Minister of Environment of 2016 (Rozporządzenie Ministra Środowiska, 2016). The much higher content of trace metals in the tree litter indicates their anthropogenic character, and the grain-size composition of soils (mainly loose sands) lowers their buffer capacity.

4. CONCLUSIONS

The assessment of the impact of cement and lime dust emitted by Trzuskawica S.A. limestone plant showed that the surrounding it podzolic and rusty soils are characterised by the grain-size composition made of loose and slightly clayey sands, relatively low content of humus, high saturation of the sorption complex with bases as well as neutral and alkaline pHs, especially of surface horizons, and also a clear enrichment of these levels with calcium carbonate.

The analysed soils are characterised by high FSTI values, unnatural for autogenous sandy soils in coniferous forest habitats and calci-mull humus. The results of the research indicate a significant influence of the cement-lime dust emitter on the change of the physical and chemical properties of the analysed soils towards trophic growth.

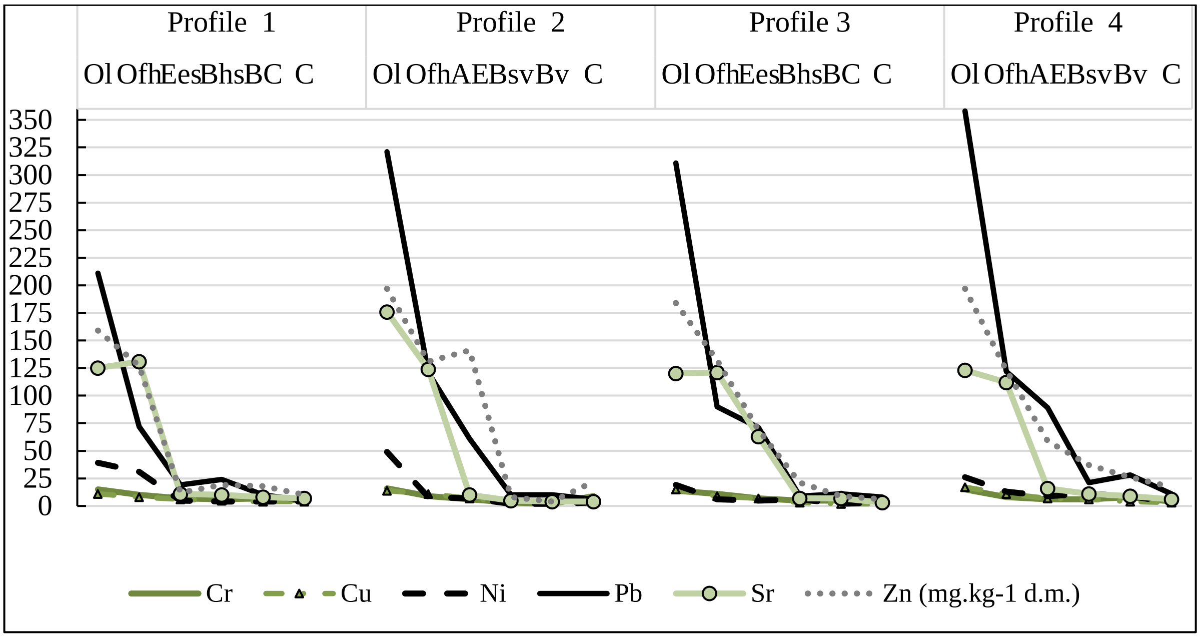

Figure 4. Trace metal content in the profiles of the analyzed soils (mg·kg $^{-1}$ d. m.).
*(Explanations of soil layer as in Table 1)

The reduced emission of cement and lime dust observed in recent years does not significantly affect the restoration of the original properties of Haplic Podzol soils under pine stands resulting from their profile structure, grain-size composition, and naturally acidic pH.

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Received at: 20. 05. 2021
Revised at: 09. 08. 2021
Accepted for publication at: 11. 08. 2021
Published online at: 13. 08. 2021

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