Comparative analysis of soil magnetic susceptibility and concentration of rare earth elements in soil of problematic areas

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INTRODUCTION

The group of rare earth elements (REE) include the lanthanide elements, Sc and Y. Usually two groups of REE elements are distinguished. The first one includes the La, Ce, Pr, Nd, Sm, Eu and is called light REE. The remaining REEs are included in the group called heavy REE such. The important characteristic of all REE is that they have very similar physical and chemical properties (Hu et al., 2006). Moreover, all REE are often found in the same type of ores.
The REE and their compounds are very important for industry mostly because of their magnetic properties. Without the REE production of permanent magnets that are part of disc drives and many important electronic parts and batteries would be impossible (Jarosiński, 2016). Because of that, REE have been recognized as resources critical for today industry, because the lack of possibility of their substitution (Seredin and Dai, 2012). REE are characterized by limited amounts of ores, and moreover REE are usually highly dispersed in their minerals. Usually, REE are obtained from ores like bastnaesite, monazite and laterite. At present the majority of ores is located in countries such as China, USA, and India. In Poland, the natural resources of REE are rather small, and REE ores, such as: monacite, xenotile, apatite and zirconium are found mostly in Lower Silesia region, especially near Szklarska Poręba town, and near Białystok town, in a form of carbonatite (Kowalczyk & Mazanek, 1989).
Numerous studies were carried out so far that aimed at the analysis of potential secondary sources of REE. As it was analyzed, significant amounts of REE can be found in phosphogypsum, electrical and electronic waste or used catalytic converters (Jarosiński, 2016). The most important secondary source of REE is scrap electronic parts, and especially permanent magnets, $Fe_{14}Nd_2B$ or Sm_2Co_5. As it was found the content of Nd in this type of magnets can reach up to 24%, and Dy levels are up to about 5%.
What is important in highly industrialized areas significant concentrations of REE can be found in fly ash produced during the combustion of hard coal (Jarosiński, 2016;

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Smółka-Danielowska D., 2007). This observation can be important for southern part of Poland where substantial part of the industry is based on the coal extraction or coal combustion. As it was studied, after the dusts containing REE are deposited on the soil surface they are accumulated in organic and humic soil layers, similarly like other pollutants (Cao, et al. 2001). The rate of adsorption of REE in soil depends on pH and soil cation exchange capacity. The availability of REE usually increases with a decrease of pH and redox potential. Concentrations of REE in soil was also found to be dependent on a presence of organic material and chemical and physical properties of soil (Ran and Liu, 1993; Beckwith & Bulter, 1993). What is important the studies related to the analysis of REE concentration in soil that results from coal-based industry is still limited.

The important characteristics of REE concentration in soil that result from coal-based industry it that technogenic particles contain also admixtures of ferromagnetic elements as Fe, Co, or Ni, and reveal strong magnetic properties that can be measured and analyzed using soil magnetometry. As it was presented in numerous studies (Thompson et al. 1980; Strzyszcz et al. 1996; Petrovsky et al., 2000; Spiteri et al., 2005; Magiera & Zawadzki, 2006, 2007; Fürst et al., 2009; Zawadzki et al., 2009; Fabijańczyk et al., 2017) increased concentration of technogenic particles can be determined in soil using several geophysical methods. One of them is soil magnetometry that is a sophisticated technique of detecting and determining the level of potential soil pollution with anthropogenic dusts that are emitted by various types of industry (Ayoubi et al., 2013, 2018a, 2018b). Most of magnetometric studies were focused on the determination of soil pollution with heavy metals or Potentially Toxic Elements. Commonly, soil magnetic susceptibility κ is measured using the Bartington MS2 Magnetic Susceptibility System, equipped with various sensors (Dearing, 1994).

In the paper the possibility of using magnetometric measurements to delineate the areas with elevated concentrations of REE in soils of highly industrialized regions was investigated. To do so, three significantly different study areas were selected for field measurements, which were located in northern, and southern Norway, and in Upper Silesian Industrial Area. The analysis of concentrations of REE in soil was based on the chemical measurements and soil samples that were collected in study areas. Also, magnetometric measurements were carried out which allowed for investigation potential correlations between soil magnetic susceptibility and concentrations of REE in soil.

METHODOLOGY OF RESEARCH

At the beginning, three distinctly different areas were carefully selected, which could be called “problematic” because of higher level of anthropogenic pollution or a specific geological background. It is worth to mention here that all field measurements and laboratory analyses were performed in the frame of the Polish-Norwegian Research Grant entitled “Development of integrated geophysical/geochemical methods of soil and groundwater pollution assessment and control in problematic areas” (the acronym IMPACT) carried out in 2009-2014. The first one was located in Piekary Śląskie, Upper Silesian Industrial Area, south Poland, in the direct vicinity of the lead ore and lead battery processing factory, about 700 m away from a heap of metallurgical wastes. In this waste heap, high concentrations of sulphides of Hg, Be, Cu, Ag, Se, as well as many REE were found. Consequently, the soils of this area were subjected to pollution by dust generated by the sulphidation processes of the sulphides contained in the waste heap. Subsequently, these dusts were transported by winds to neighboring areas, including the field being investigated.

Second area was located in northern Norway, above the Arctic Circle near the border with Russia. This area is very polluted due to the nearby location of the mining industry that has been developing there for about 80 years. The main industrial facility was a Bjornevatn mine dealing with iron ore mining. During the mining of iron ores, significant amounts of waste are generated which is the cause of strong anthropogenic pressure on the environment (Bronder et al., 2010). Two main soil types were observed in the study area. Leptosols, shallow soils formed on hard rock or gravel material and have a low water retention capacity (Jones et al., 2010). In this area the magnetic signal is due to fine-grained primary sulphides and secondary fine-grained magnetite and/or maghemite (Magiera et al., 2018).

The third area was located in the southern Norway between Lyngdal and Moi villages. This area was not subjected to the significant local anthropogenic pressure though the geological background was composed of granite bedrock. In a consequence, soils of this area were characterized by significantly high magnetic background. Some studies also suggest that this part of Norway may be subjected to long-range pollution (Steinnes & Friedland, 2006).

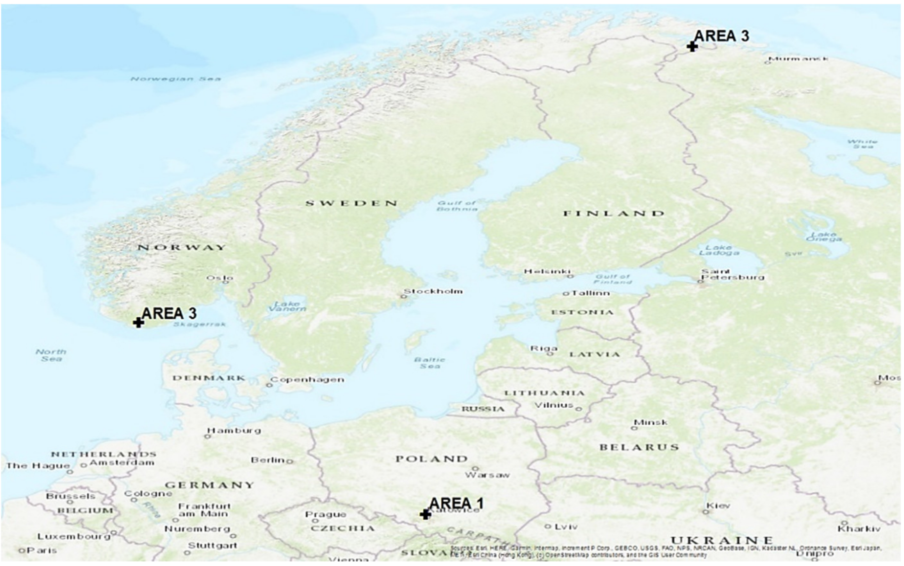

Fig. 1 Study area with marked location of measurement sites

At each study area about 15 soil cores were collected. Soil cores, 30 cm long, were taken to plastic tubes using a Humax sampler. Soil samples used to determine the content of selected REE in the soil were cut from a depth of 0 cm to 10 cm below the soil surface. Next, soil samples were dried, homogenized and sieved through a 2 mm sieve to separate the soil skeleton. Then, about 250 mg of the dried soil sample was digested with 50% (v/v) HNO_3, placed in a Teflon bottle, diluted to 108 ml and transferred to 15 ml vials for ICP-MS analysis.

Mass specific magnetic susceptibility χ was calculated using collected soil cores. Firstly, soil subsamples were cut from this part of collected soil cores, where maximum value of κ was measured. Next, magnetic susceptibility of these subsamples was measured using a MS2B Bartington sensor (Dearing, 1994).

Fig. 2 a) Humax sampler, b) 30 cm length soil profile taken using Humax sampler
Source: (Magiera, Zawadzki, 2006).

Fig. 3 Laboratory system with a MS2B Bartington device for magnetic susceptibility measurements
Source: (Magiera, Zawadzki, 2006)

The MS2B sensor had a resolution equal to 2×10^{-6} SI and its calibration accuracy was equal to 1%. The mass specific magnetic susceptibility χ of each soil subsample was calculated using soil subsamples with a volume of 10 cm^3. Consequently, the calculation of mass-specific susceptibility was done by multiplication by 10 cm^3 and division by the actual mass of the soil sample, irrespective of its actual volume.

RESULTS AND DISCUSSION

Concentrations of REE in soils were previously studied, and typical concentrations of REE in soil were determined (Ramos et al., 2016). These values, separately for Poland and Norway were presented in the Table 1.

Table 1
Typical concentrations of REE in soils of Poland and Norway

	Poland	Norway
	[mg/kg]	
Ce	5.5 – 72	5.7 – 253
Dy	0.4 – 5.1	0.8 – 9
Er	0.3 – 3.1	0.6 – 5.0
Ho	0.1 – 1.0	0.9 – 1.8
La	2.7 – 35	2.9 – 101
Lu	0.05 – 0.5	0.1 – 0.8
Nd	2.3 – 30	2.8 – 80
Pr	0.7 – 8.0	2.8 – 80
Sc	0.7 – 12	2.5 – 33
Sm	0.4 – 5.5	0.5 – 13
Tb	0.05 – 0.8	0.1 – 1.7
Tm	0.05 – 0.5	0.09 – 0.8
Y	14 – 30	-
Yb	0.3 – 3.4	0.6 – 5.8

Source: (Ramos et al., 2016)

As it was observed, in the area located near the battery processing factory (Area 1) measured average concentrations of all REE (Table 2) in soils were within the typical ranges observed in previous studies (Table 1). Similar observation was made in the case of the area 3, where the soils were developed on the granite bedrock.

In the area that was located in the vicinity of the Bjornevatn iron ore exploration factory. It was observed that the average measured concentrations of REE in this area were closer to a lower boundary of the interval of typical concentrations (Table 1). In the case of Pr, Ho, and Lu, the average measured concentrations in soil were even lover than a usually observed concentration that were determined in previous studies.

As it was previously observed during comparison with typical REE concentrations in soil, the lowest concentrations of all REE were observed in the area located in the vicinity of iron ore exploration factory. Simultaneously, in this area calculated values of mass magnetic susceptibility of soil were high, with maximum values reaching over 800×10-3 m^3/kg. Average measured mass magnetic susceptibility in this area was equal to about 306×10-3 m^3/kg. Such observation suggest that soil was potentially polluted or, what was also evident in this area, increased magnetic susceptibility of soil resulted from natural properties of geological substrata and presence of iron ores. Strong influence on the natural background values of soil magnetic susceptibility was observed in the area 3, where granite bedrock was present. The calculated values of mass magnetic susceptibility in this area were very high, up to 1600×10-3 m^3/kg. Such extreme values might suggest the extreme soil pollution, however in the area 3 anthropogenic pressure was low, and very high magnetic susceptibility of soil was a result of properties of granite rock. As it was observed, also the concentrations of REE in this area were the highest of all study areas. As it could be expected, geological bedrock in this area was characterized by some, increased amounts of REE.

Table 2
Descriptive statistics of REE concentrations in soil, and mass magnetic susceptibility in study areas 1 and 2

		Average	Minimum	Maximum	Q25%	Q75%	Std. Dev.
Area 1: lead ore and battery processing	χ	162	8	295	107	237	78
	Ce	39.31	31.09	50.16	34.15	44.26	5.52
	Dy	2.02	1.47	2.72	1.73	2.42	0.37
	Er	1.03	0.68	1.43	0.87	1.23	0.21
	Ho	0.36	0.25	0.5	0.3	0.43	0.07
	La	18.58	14.66	23.71	16.66	20.84	2.4
	Lu	0.12	0.09	0.17	0.11	0.15	0.02
	Nd	16.56	13.31	21.48	14.24	18.98	2.31
	Pr	4.37	3.49	5.73	3.8	4.95	0.62
	Sc	4.02	2.51	6.96	3.33	4.72	0.93
	Sm	3.14	2.42	4.18	2.76	3.64	0.47
	Tb	0.35	0.26	0.48	0.29	0.41	0.06
	Tm	0.13	0.09	0.19	0.11	0.16	0.03
	Y	9.44	6.21	12.98	8.17	11.07	1.82
	Yb	0.86	0.58	1.15	0.72	1.04	0.16
Area 2: iron ore exploration	χ	306	2	887	109	533	245
	Ce	8.04	1.21	21.49	5.37	10.05	5.24
	Dy	0.8	0.08	1.9	0.45	1.06	0.54
	Er	0.49	0.05	1.16	0.28	0.7	0.3
	Ho	0.16	0.01	0.39	0.09	0.22	0.11
	La	4.16	0.66	14.41	2.74	4.89	3.23
	Lu	0.06	0.01	0.15	0.03	0.09	0.04
	Nd	4.38	0.59	17.51	2.7	4.98	4.01
	Pr	1.08	0.15	4.17	0.67	1.2	0.95
	Sc	4.09	0.29	11.07	2.16	5.46	2.82
	Sm	0.86	0.12	3.12	0.5	1.13	0.73
	Tb	0.13	0.02	0.35	0.08	0.18	0.09
	Tm	0.07	0.01	0.17	0.03	0.1	0.04
	Y	4.19	0.47	9.55	2.41	5.93	2.64
	Yb	0.4	0.04	0.98	0.23	0.58	0.25

As it was observed in all studied areas statistically significant correlations were observed between mass magnetic susceptibility and the concentrations of all REE in soil. The strength of these correlations was the lowest in the area located near the iron ore processing factory. Such observation was related to very high concentration of iron ores that can be characterized by strong magnetic properties. In a consequence, the magnetic signal of REE ores or compounds could be too weak to be detected using measurements of soil magnetic susceptibility.

In the area with low anthropogenic pressure, where soils were developed on the granite bedrock, high and significant correlations between mass magnetic susceptibility and the concentrations of all REE were observed. Such observation suggested that soil magnetic susceptibility can be used as an indicator of increased concentrations of REE also in areas with low pollution, but with natural presence of ores.

Table 3
Descriptive statistics of REE concentrations in soil, and mass magnetic susceptibility in study area 3

		Average	Minimum	Maximum	Q25%	Q75%	Std. Dev.
Area 3: influence of granite bedrock	**χ**	986	129	1561	814	1236	369
	Ce	55.95	8.65	155.95	24.76	82.5	37.6
	Dy	2.51	0.43	5.1	1.23	3.16	1.37
	Er	1.3	0.22	2.56	0.69	1.57	0.71
	Ho	0.45	0.08	0.9	0.23	0.56	0.25
	La	19.15	3.47	53.92	9.91	25.58	11.86
	Lu	0.15	0.03	0.29	0.09	0.19	0.08
	Nd	20.08	3.29	55.7	10.43	25.42	12.21
	Pr	5.34	0.89	15.12	2.7	7.17	3.28
	Sc	2.6	0.63	5.65	1.47	3.58	1.37
	Sm	3.55	0.57	8.85	1.84	4.31	2.07
	Tb	0.42	0.07	0.86	0.21	0.51	0.23
	Tm	0.18	0.03	0.35	0.09	0.22	0.1
	Y	11.12	1.93	23.48	5.94	13.68	6.11
	Yb	1.12	0.18	2.18	0.59	1.34	0.59

In the vicinity of lead ore and battery processing factory between mass magnetic susceptibility and the concentrations of all REE were equal to about 0.5-0.6, and for most of REE were statistically significant. The reason for so evident correlations could be significant amounts of dust blown from the waste heap after battery post-processing. The distance from the study area to this waste heap was low, and equal to about 700 m.

Table 4
Pearson's and Spearman's correlation coefficients between mass magnetic susceptibility and REE concentrations in soil in study areas 1, 2, and 3; marked coefficients are statistically significant at α = 0.05

	Area 1: lead ore and battery processing		Area 2: iron ore exploration		Area 3: influence of granite bedrock	
	Pearson's	Spearman's	Pearson's	Spearman's	Pearson's	Spearman's
Ce	**0.36**	0.12	0.51	0.19	**0.70**	**0.68**
Dy	**0.64**	**0.63**	0.42	0.25	**0.79**	**0.75**
Er	**0.63**	**0.55**	0.29	0.32	**0.79**	**0.75**
Ho	**0.61**	**0.52**	0.35	0.27	**0.79**	**0.74**
La	0.34	0.17	**0.59**	0.10	**0.72**	**0.68**
Lu	**0.61**	**0.54**	0.17	0.25	**0.79**	**0.72**
Nd	0.28	0.21	**0.60**	0.14	**0.76**	**0.72**
Pr	0.21	0.16	**0.60**	0.16	**0.74**	**0.72**
Sc	**0.51**	**0.49**	-0.08	0.06	**0.48**	**0.53**
Sm	**0.37**	0.31	**0.58**	0.24	**0.77**	**0.71**
Tb	**0.58**	**0.51**	0.47	0.28	**0.79**	**0.73**
Tm	**0.58**	**0.52**	0.22	0.29	**0.80**	**0.74**
Y	**0.69**	**0.63**	0.37	0.29	**0.81**	**0.76**
Yb	**0.54**	**0.44**	0.18	0.22	**0.79**	**0.74**

CONCLUSIONS

The weakest correlations between soil magnetic susceptibility and concentrations of REE were observed in the vicinity of iron ore exploration factory in Bjornevatn (Area 2). There can be two possible explanations of such observation, one is low concentrations of REE in soil of this area, and the second is strong magnetic signal from the iron ores. In the area located near a factory of lead ore and battery processing (Area 1) concentrations of most of REE were significantly correlated with soil magnetic susceptibility. The highest correlations coefficients between soil magnetic susceptibility and concentrations of REE were found in the area where soils were developed on the bedrock with strong natural magnetic background with the influence of granite rock (Area 3). Analysing the above results it can be concluded that the origins of observed dependencies could be complicated or even indirect, and that these results based on macroscopic level measurements need to be checked by studies at microscopic scale. On the other hand, the obtained values of Pearson's and Spearman's correlations coefficients were often unsuspectedly high. However, more systematic studies including e.g. the use of geostatistical methods and geochemical indicators, especially carried out on anthropogenically polluted soils with REE are necessary to confirm the observed dependencies.

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Zawadzki, J., Fabijańczyk, P., Magiera, T. (2009). Geostatistical evaluation of magnetic indicators of forest soil contamination with heavy metals. Studia Geophysica et Geodaetica, 53(1), pp.133-149.

Abstract: Soil contamination with rare earth metals (REE) can have both direct and indirect connection with industrial activity and ore-exploration. In the studies conducted so far, the presence of rare earth elements in coal seams, coal combustion waste as well as fly ash was found. It is important that detailed studies of the REE content in soil were not carried out in Poland. Until now, in a few studies, a high content of cerium and lanthanum was found in relation to the average content of the torn ones in the world. This work focuses on the areas under the influence of the industry associated with the extraction and combustion of hard coal, but also with other types of industry. Analyzes of REE content in soil were conducted in selected areas of the Upper Silesian Industrial Region and Norway, located near the Bjornevatn mine. In study areas, soil samples were collected and used for chemical and magnetometric measurements. Firstly, concentrations of REE were determined, and after that soil samples were used to measure soil magnetic susceptibility. Finally, statistical analyses were performed in order to check the correlation between REE concentrations in soil and soil magnetic susceptibility.

Keywords: soil pollution, rare earth elements, magnetometry, magnetic susceptibility, industrial areas