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MATHEMATICAL MODELLING FOR PARAMETRIC ASSESSMENT OF GENERAL TOXICITY OF WASTES CONTAINING METALS

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Abstract. The polynomial static predictive mathematical model was used to assess general toxicity of the selected groups of waste substances on the basis of heavy metal leached forms. The absolute value of waste substances was calculated for waste samples with metals determined by using TCLP (Toxicity Characteristic Leaching Procedure). Gradient factors for general toxicity equation in the form of $y(x_1, x_2, x_3, \ldots, x_n) = ax_1 + bx_2 + cx_3 + \ldots + nx_n$ were selected with the use of weighted mean, considering the weight of their toxicological properties. The value of y was calculated by adding up experimentally determined individual residual concentrations of elements, taking into account their weighted fraction in the total toxic load. Seeking the assessment criterion, it was assumed that the contents of metals leached from the tested wastes that may occur in toxic concentrations is hazardous for the components of biosphere when the value of general toxicity fulfils the relation: $y(x_1, x_2, x_3, \ldots, x_n) \geq 1$. Basing on computing for the assumed model of general toxicity for selected wastes and considering the verification of the model with reference to the toxicity criteria defined in TCLP, it was found that there was a coincidence between the achieved results.

Keywords: general toxicity of waste substances**,** toxic metals, mathematical modelling.

1. Introduction

Search for simple mathematical models to asses quickly the general toxicity of waste substances has become an issue of current interest of practical character. Literature data indicate two basic approaches to solve the issue of mathematical modelling of natural systems that can be found in present ecosystems (Jones and Bryan 1998). With the use of the so called descriptive and predictive approach, attempts are made to present the abstract systems in a mathematical way so that they can be the most similar to the real world of nature.

The aim of the descriptive approach is to use the mathematical model in order to achieve the largest amount of information about the way how the real system functions, in other words, to obtain the results of modelling with reference to basic experimental data. In case of predictive modelling, the main purpose is to obtain the most likely and representative experimental results describing a given phenomenon to use them as input data for the abstract mathematical modelling. At present, the most popular are predictive models (Zhang and Arhonditsis 2008). For instance, the Langmuir and Freundlich sorption models are typical predictive models used e.g. to describe events of sorption of pollutants in soil and sediments or sludge. They are most highly valued due to the fact that they match properly a theoretical adsorption curve to experimental data (Kinniburg 1986). In literature, you can find much more descriptive approach to the phenomenon of heavy metal sorption in the water-soil environment. The sorption on mineral and organic fractions is recognised most commonly and determined most frequently. The sorption on mineral fractions is described, among others, by the following models: Constant Capacitance Model (CCM), Diffuse Layer Model (DLM), Triple Layer Model (TLM) or Three Plane Model (TPM). The most frequently used are: CCM, DLM and TLM, which were included into the geochemical speciation code MINTEQ2, also in its complemented and enhanced form: MINTEQ3 (David and Allison 1999; Gustafsson 2000). In literature, you can find studies concerning mathematical modelling of remediation soils contaminated with heavy metals (Jankaitė 2009), transport of metals from the soil to tree seedlings (Baltrėnaitė and Butkus 2007), phytoextraction with e.g. maize (Chrysafopoulou *et al.* 2005), and simulation of the metal mobility in soils contaminated with waste sludges (Yahya and Abdulfatai 2007). Moreover, the literature raises the issues of metal bioaccumulation in sea benthic deposits (Clason *et al.* 2004) or in oceans by comparing complex models with e.g. a toxicokinetic one (Clason *et al.* 2004a).

2. Analytical part

The analytical procedure applied in TCLP was based on a method used in the USA: US 1311, 1990. According to these techniques, solid material particles (of soil, ground or solid waste) of diameter lower than 9.5 mm are required. With larger particles a sample is disintegrated by crushing, grinding or screening. A buffer solution of the aqueous acetic acid (L) in a ratio of liquid to solid material (S), respectively: (L/S) = 20:1 is used for leaching. A mixture prepared in this way is subject to leaching (shaking) during 18 ± 2 hours, then the phases are separated and the liquid is subject to further analyses. Samples of soil polluted with metals were taken from the depth of 0.0-20.0 cm from the area of the processing plant producing mineral pigments and from the area of the roundabout in the big municipal centre (samples marked in the table lists with indices 7 and 9). Samples of processing wastes (I and J) come from petrochemical industry. Sewage sludge samples were taken from two municipal wastewater treatment plants. Samples from the first treatment plant (D) were the raw sludge dehydrated on a press, and samples from the second treatment plant (E) were the sludges from fermentation carried out in the so called sludge digestion chambers. These sludges were dried at 105±1°C to obtain solid mass. In this study, the samples of waste substances were leached with a solution of pH = 4.93±0.05 prepared by adding 11.4 ml of glacial acetic acid and 128.6 ml of 1.0 M NaOH solution to 0.5 l of deionized water, and filling up to the volume of 2.0 l.

A method of inductively coupled plasma atomic emission spectrometry (ICP-AES) (Perkin Elmer Optima) was used to analyze leached metal forms in eluates. Three standards and a blank test were used for calibration. The standard samples were made in the matrix of the nitric acid(V)/hydrochloric acid, as defined by standard US EPA 6010 (US EPA 6010, 1986). The efficiency of the device was estimated while conducting the analyses by determining a method detection limit (MDL) and some quality control (QC) parameters. The results of maximum residue level (MRL) were in (mg/l) for: As – 0.5, Cd – 0.05, Ba – 0.04, Cr – 0.06, Pb – 0.3. While analyzing, some blank tests were made to check proper functioning of the ICP apparatus. At first calibration was checked with the standard averaging of the calibrated values. The level obtained ranged within the limits ±5% of the guidelines determined for this method. A trial of checking the interference for samples containing high concentrations of: Al, Na, Ca and Mg was carried out in order to ensure that the frequencies selected for this method observed no undetectable interference. The control standard checks (CSTD) under laboratory conditions were taken to verify efficiency of the apparatus and the methods used. CSTD values were within ±10%. Duplicated samples were analyzed and the analyte was determined within ±15% with reference to the original sample. External standards were added to the analyzed samples (1 mg/l for, respectively: Ag and Ba, and 2 mg/l for: Cr, Cd, As, Pb and Se). Other quality control parameters such as: laboratory spike blank (LSB), laboratory spike blank duplicate (LSBD), laboratory matrix spike (LMS), i.e. quartz and the laboratory matrix spike duplicate (LSMD) were included into the test.

3. Computational part

Standardised values for metals from the TCLP procedure were used to pre-assess the general toxicity of wastes in a static (i.e.: not including variations in values in time) predictive and polynomial model in the form of (1):

$$y(x_1, x_2, x_3, \ldots, x_n) = ax_1 + bx_2 + cx_3 + \ldots + nx_n \quad (1)$$

where: y – general toxicity of the tested waste [mg/l];

a, b, c, …, n – weighted conversion factors of limit values of metal concentration in the TCLP criterion (considering toxicological properties);

$x_1, x_2, x_3, \ldots, x_n$ – independent variables that present values of determined concentrations for the individual metals in waste samples acc. to TCLP procedure (US EPA 1992).

In order to calculate general toxicity, the published data from the publication (Mantis *et al.* 2005; Saeedi *et al.* 2009) and own studies where concentrations of heavy and toxic metals were determined according to the TCLP procedure were applied.

4. Results and discussion

Toxicity is typical of heavy metals, or (even more often) their respective speciation forms, consisting in causing function disorders or death of living cells, organs, or organisms after they get into their vicinity. Heavy metals and/or their speciation forms can show a toxic effect after being absorbed by different ways e.g. by diffusion, sorption, ion exchange, solution, co-precipitation, etc. (Rauckyte *et al.* 2009). Toxicity is an undesirable activity resulting from chemical, biochemical reactions and the complex of physico-chemical effects between the metal in its ionic form and/or its speciation form, which infiltrated the system, and the biological systems (i.e.: DNA, enzymes) (Rauckyte *et al.* 2009). Parameters that define the toxicity of heavy metals and make a part of the total load of wastes toxicity take into account labile speciation forms of these metals. In ecosystems, these forms occur in organic or mineral complexes mostly ($M(R)^{m+}_k$ where: M – metal, m+ - metal valence, R – organic or mineral ligand, k – organic ligand coordination number).

The test of toxic leaching consists in selecting the leaching reagents, assuming that they are much more aggressive than those occuring in natural conditions as elements of the ecosystems (US EPA Fed. Reg. 1986; Ure *et al.* 1993). A limit of toxic leaching procedure for each metal leached from soil or sludge was established at the level of one hundred times exceeding the maximum concentration of metal pollutions in potable water. E.g., for cadmium: 0.01 mg/l in potable water, and 1.0 mg/l – a limit value of toxic leaching; and for lead: 0.05 mg/l in potable water, and 5.0 mg/l - a limit value of toxic leaching (Rauckyte *et al.* 2009). The eluted or toxic forms of metals, e.g., lead, make as a rule app. 10-30% of total metal concentration determined in the concrete environmental components or in wastes (Rauckyte *et al.* 2006; Devesa-Rey 2010). With this respect, it was not possible to use data of metal total concentrations to undertake modelling attempts. The selection of gradient factors for the polynominal equation was based on permissible limit values for metals in TCLP procedure, presented in Table 1.

Gradient factors were selected by using weighted mean method, considering a number of metals with toxic concentrations basing on the criteria stated in TCLP procedures for the assessment of general toxicity of the indicated waste. On the ground of the achieved results, an attempt was made to indicate which of the tested wastes should be classified as wastes toxic to the environment and which ones can be treated as environmentally safe. The assessment of the toxicity level (y) for the indicated group of wastes was quantified with the assumption of the following criterion:

$y(x_1, x_2, x_3, \ldots, x_n) \geq 1$ – the tested waste is environmentally toxic;

$y(x_1, x_2, x_3, \ldots, x_n) < 1$ – the waste is not toxic.

It was assumed that the contents of forms of the leached metals with toxic concentrations in wastes is hazardous for the components of ecosystems when the value of general toxicity fulfils the following relationship $y(x_1, x_2, x_3, \ldots, x_n) \geq 1$. For instance, in order to make computations, concentrations of toxic metals [mg/l] determined in samples of processing wastes (I4 – I6 and I and J) (Table 2), waste sludges (U1, U5, U8 and D, and E) (Table 4), and soil samples (NH7-1, NH8, NH7-3 and 7, and 9) (Table 6) acc. to TCLP procedure, and the experimental data for one's own calculations were taken from reports (Mantis *et al.* 2005; Saeedi *et al.* 2009). Basing on the experimental data found in literature and own analytical studies, calculations were made for the assumed model of general toxicity, the results of which are presented in Tables: 3, 5, 7.

Table 1. Limit permissible contents of toxic metals according to the TCLP criterion (US EPA 1992; e-CFR [online] 2011)

Toxic metals	Pb	As	Ba	Cd	Cr	Hg	Se	Cu	Zn
Maximal permissible contents of toxic metals according to the TCLP criterion [mg/l]	5	5	100	1	5	0.2	1	15	25

Table 2. The level of metal concentrations [mg/l] in exemplary processing wastes determined in accordance with the TCLP procedure and used for one's own modelling attempts

Metal / Sample	As	Ba	Pb	Cd	Cr
I4[a)]	6.1	–	18.0	1.13	33.6
I5[a)]	3.5	–	13.7	1.40	32.0
I6[a)]	3.3	–	18.3	1.42	31.4
I[c)]	0.1	17.2	34.0	0.30	0.5
J[c)]	0.1	21.0	40.0	0.10	0.7

where: [a)] the experimental data were taken from report (Mantis *et al.* 2005); [c)] results of own co-author analytical studies (Richert *et al.* 2004).

Table 3. A list of calculated general toxicity values (y) for industrial wastes according to the assumed model $y(x_1, x_2, x_3, \ldots, x_n) = ax_1 + bx_2 + cx_3 + \ldots + nx_n$

Sample	Calculated general toxicity value $y(x_1, x_2, x_3, \ldots, x_n)$	Assessment of waste in accordance with the assumed criterion	Verified values of general toxicity[b)]
I4[a)]	17.95	y > 1, the waste qualified as toxic	58.83
I5[a)]	15.34	y > 1, the waste qualified as toxic	50.60
I6[a)]	16.51	y > 1, the waste qualified as toxic	54.42
I[c)]	16.32	y > 1, the waste qualified as toxic/non-toxic	52.1
J[c)]	19.86	y > 1, the waste qualified as toxic/non-toxic	61.9

where: calculations were based on the experimental data published in report [a)] (Mantis *et al.* 2005), [b)] made on the TCLP criterion (US EPA 1992; e-CFR [online] 2011); [c)] calculated on the basis of results of own co-author analytical studies. Gradient factors were selected on the basis of literature data and they were respectively for [a)]: As – 0.31, Cd – 0.06, Cr – 0.31, Pb – 0.31 and for [c)]: As – 0.0431, Pb – 0.0431, Cd – 0.0086, Cr – 0.0431, Ba – 0.8621.

Table 4. The level of metals concentrations [mg/l] in the exemplary waste sludges (from biological treatment of municipal wastes processing with the use of the activated sludge technology); determined acc. to TCLP procedure, which were later used for own modelling attempts

Metal / Sample	As	Ba	Pb	Cd	Cr
U1[a)]	23.2	–	24.0	1.06	20.0
U5[a)]	29.2	–	26.0	1.18	24.0
U8[a)]	33.1	–	26.3	0.97	23.7
D[c)]	< 0.5	1.8	0.3	0.10	0.2
E[c)]	< 0.5	2.2	0.6	0.10	1.2

where: [a)] experimental data were taken from the report (Mantis *et al.* 2005); [c)] results of own co-author analytical determinations (Richert *et al.* 2004).

Table 5. A list of calculated general toxicity values (y), according to the assumed model $y(x_1, x_2, x_3, \ldots, x_n) = ax_1 + bx_2 + cx_3 + \ldots + nx_n$ in the waste sludge samples

Sample	Calculated general toxicity value $y(x_1, x_2, x_3, \ldots, x_n)$	Assessment of waste in accordance with the assumed criterion	Verified values of general toxicity[b)]
U1[a)]	20.89	y > 1, the waste qualified as toxic	68.26
U5[a)]	24.62	y > 1, the waste qualified as toxic	80.38
U8[a)]	26.75	y > 1, the waste qualified as toxic	87.07
D[c)]	1.596	y > 1, the waste qualified as non-toxic	2.90
E[c)]	1.997	y > 1, the waste qualified as toxic/non-toxic	4.60

where: calculations were based on the experimental data published in report [a)] (Mantis *et al.* 2005), [b)] made on the TCLP criterion (US EPA 1992; e-CFR [online] 2011); [c)] calculated on the basis of results of own co-author analytical studies. Gradient factors were selected on the basis of literature data and they were, respectively, for [a)]: As – 0.31, Cd – 0.06, Cr – 0.31, Pb – 0.31 and for [c)]: As – 0.0431, Pb – 0.0431, Cd – 0.0086, Cr – 0.0431, Ba – 0.8621.

Table 6. The level of metals concentrations [mg/l] in the exemplary degraded soil samples determined in acc. to TCLP procedure and used for own modelling samples

Metal / Sample	As	Pb	Cd	Cr
NH7-1[b)]	–	17.39	0.38	0.24
NH8[b)]	–	8.21	0.46	0.20
NH7-3[b)]	–	1.49	0.34	0.15
7[c)]	13.0	0.50	0.30	–
9[c)]	< 0.5	1620.0	3.30	–

where: [a)] experimental data were taken from the report (Mantis *et al.* 2005); [c)] results of own co-author analytical determinations (Rauckyte *et al.* 2009).

Table 7. A list of calculated general toxicity values (y), according to the assumed model $y(x_1, x_2, x_3, \ldots, x_n) = ax_1 + bx_2 + cx_3 + \ldots + nx_n$ in the soil samples

Sample	Calculated general toxicity value $y(x_1, x_2, x_3, \ldots, x_n)$	Assessment of waste in accordance with the assumed criterion	Verified values of general toxicity[d)]
NH7-1[b)]	7.97	y > 1, the waste qualified as toxic	18.01
NH8[b)]	3.83	y > 1, the waste qualified as toxic/non-toxic	8.87
NH7-3[b)]	0.77	y > 1, the waste qualified as non-toxic	1.98
7[c)]	6.10	y > 1, the waste qualified as toxic	13.80
9[c)]	729.52	y > 1, the waste qualified as toxic	1623.50

where: calculations were based on the experimental data published in report [b)] (Saeedi *et al.* 2009), [d)] were made on the basis of TCLP criterion (US EPA 1992; e-CFR [online] 2011); [c)] calculated on the basis of results of own co-author analytical studies. Gradient factors were selected on the basis of literature data and they were respectively for [b)] Cd – 0.09, Cr – 0.45, Pb – 0.45 and for [c)]: As – 0.45, Pb – 0.45, Cd – 0.09.

Samples taken from the processing wastes (Tables 2, 3) in which barium was not determined were unambiguously assessed as toxic, which was additionally justified by verification. However, the samples in which barium concentration was taken into account for calculating general toxicity (samples I and J) were classified as toxic and during the verification as non-toxic due to the fact that neither of them exceed the value of 116. An important toxic metal characterized by the most significant effect on general toxicity of the discussed wastes is lead. Its concentration exceeds app. 3-8 times permissible value of 5 mg/l that is considered to be a threshold value. Chromium is another heavy metal with concentration exceeding 6 times its permissible toxic level. In waste sludges presented here, samples with determined barium and other leached toxic elements of very low concentration levels are not unequivocal either, considering the assumed criterion, but they are toxic due to the calculated general toxicity value and non-toxic due to verification (even if barium concentrations are not considered). However, the toxicity assessment of the three literature samples of waste sludge (U1, U5, U8) was mostly influenced by: arsenic (exceeded 6 times the TCLP standard), lead (5 times), and chromium (4 times).

The most important metal affecting the assessment of soil as toxic is lead with its concentration in the leached solution exceeding 1.6 (or app. 3.5) times permissible level acc. to TCLP. Arsenic is another metal that affected significantly the level of soil general toxicity acc. to the assumed criterion (exceeding 2.6 times the standard concentration given by TCLP), with trace concentrations of leached lead and cadmium. The only soil sample qualified as non-toxic in accordance with the calculations and verification was NH7-3, where none of the analyzed elements: Pb, Cd or Cr determined with the TCLP procedure exceeded permissible toxic concentrations. Soil NH8 was qualified as toxic waste in accordance with the calculated general toxicity value with the use of the assumed mathematical model, but after verifying - as non-toxic. Therefore, one can notice that the calculated values of general toxicity with the assumed gradient factors are higher than the verified ones.

5. Conclusion

1. The search for simple mathematical models in order to describe quantitatively the general toxicity of processing wastes, sewage sludge or contaminated soils basing on known permissible soluble forms of heavy metal speciation fractions is currently an essential issue, the solution of which is necessary to solve basic problems of environmental protection.
2. The static-predictive and polynomial model for the assessment of general toxicity of wastes might be a fast and simple method to achieve information on general toxicity on the basis of already possessed values obtained from e.g. tests of sequential extraction for the so called ion-exchanging and carbonate fractions which are comparable with the values achieved by using the TCLP procedure.

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