

Original Research

An Evaluation of Endemic Exposure of Citizens Living in Near a Gdańsk Phosphatic Fertilizer Plant

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Abstract

The impact of the Gdańsk Phosphatic Fertilizer Plant “Fosfory” on citizens living near the plant was examined. Al, As, Au, Br, Ca, Cl, Cu, Dy, I, In, K, La, Mg, Mn, Mo, Na, S, Sb, Sm, U, V, Zn were determined by instrumental neutron activation analysis (INAA) in hair and toenail clippings collected from 22 persons living in the neighborhood surrounding the plant and from 11 people from the control district.

Essential differences in concentrations of elements for the citizens and the control group were evaluated using a non-parametric Mann-Whitney U-test. Significant differences between citizens of the factory neighborhood and the control group were found, for instance, for Mg, Sb, S and V, but it is hard to define if the factory impact exists, and whether it is the basis for these differences.

The study was also undertaken to indicate a correlation between hair and nail element content and differences between concentrations of these elements considering sex.

Keywords: phosphatic fertilizer plant, hair analysis, nail analysis, neutron activation analysis (NAA)

Introduction

Environmental and occupational risk assessment is one of the most discussed topics in modern science. It is very well known that many technologies contaminate the environment and may influence human health, both in workers and people living in the polluted areas [1-3]. One such technology is the phosphatic fertilizer industry. During the process of fertilizer production, elements like fluorine, uranium and lanthanides are released into the environment, and may have a pathological impact on occupational staff causing fluorosis, respiratory illnesses and other diseases. One element which is especially dangerous is uranium and its decay products (e.g. radon).

Radiation from radon and its offsprings produces a risk of lung cancer through inhalation over a long period of time [4].

Last year, in terms of the Copernicus project, the impact of the Gdansk Phosphatic Fertilizer Plant on its workers was examined. It was found that the concentration of most elements in hair, nails and air filters was higher for workers than for the control group, especially for elements typically originating from phosphate rocks, like rare earth elements (REE) and uranium [5]. What is more, an impact on the adjacent environment was found [6]. From this point of view, there is a probability that the factory can have an impact on Gdańsk citizens.

The aim of this project was to evaluate the impact of the Gdańsk Phosphatic Fertilizer Plant “Fosfory” on the inhabitants of the plant’s neighborhood.

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Experimental Procedures

Sampling and Sample Preparation

To assess the impact of the Gdańsk Phosphatic Fertilizer Plant on the citizens nearby, samples of hair and toenail clippings were collected from three of the most exposed districts of Gdansk: Nowy Port (NP), Przerobka and Stogi. The latter 2 districts were treated as one group (PS), because they are very close to each other (Fig. 1). The sampling sites are close to and leeward of the fertilizer plant. A total of 22 samples were collected from both groups. Hair and toenail clippings were collected using stainless steel scissors. Hair samples were taken from the head above the neck. Hair and toenail clippings were washed in accordance with the IAEA recommended procedure consisting of five successive washes: acetone – water – water – water – acetone each for 10 min in 25 ml of each solvent [7]. Additionally, an ultrasonic bath was used to improve washing procedure.

Control samples (C) of hair and nails were collected from 11 persons living approximately 15 km away from the plant where the potential impact was considered minimal.

Analytical Techniques

Neutron Activation Analysis (NAA) is a sensitive analytical technique useful for performing both qualitative and quantitative multi-element analysis of major, minor, and trace elements in samples from almost every conceivable field of scientific or technical interest [8-10]. Analysis of hair and nails was performed at the Interfaculty Reactor Institute in Delft, The Netherlands, at a 2MW swimming-pool nuclear reactor.

The analytical procedure can be divided into three steps:

- 1) preparing and irradiating samples
- 2) performing measurements, and
- 3) interpreting the results.

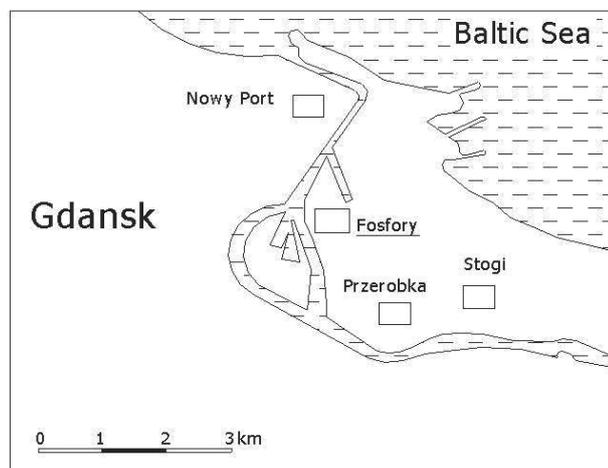


Fig. 1. Location of the "Fosfory" Phosphatic Fertilizer Plant and sampling districts.

The weight of the analyzed samples differed with type of sample: hair approximately 10-250 mg and nails approximately 10-100 mg. In step 1 sample material was packed in high purity polyethylene capsules. Comparators were added in order to measure the neutron flux during irradiation. The whole sample package was packed in an irradiation container, and irradiated by shooting it to a position close to the reactor's core. In step 2, the samples were unpacked and measured on either a well-type or a coaxial semi-conductor detector, depending on the amount of radioactivity formed. In step 3, the results were interpreted. The peaks were fitted, their peak areas and energies determined, and the neutron flux was determined by analyzing the comparators. Short-lived nuclides were determined first. Using a Fast Rabbit System, each sample was irradiated for 2 min in a neutron flux of $1.5 \cdot 10^{17} \text{ m}^{-2} \cdot \text{s}^{-1}$. After a waiting time of 1 min samples and comparator were measured for 5 min. After several days (5-7), the elements with longer half-life times were determined. After this waiting period samples were irradiated for 4 hours in a neutron flux of $0.5 \cdot 10^{16} \text{ m}^{-2} \cdot \text{s}^{-1}$. To determine the elements with middle half-life nuclides, all samples were measured 5 days after irradiation for 1 h. To determine the long half-life nuclides, the samples were measured again 3 weeks after irradiation for 4 h. After this third measurement, all 3 spectra of each sample were interpreted together.

Results

Quality Control/Quality Assurance

The accuracy of the method was checked by analyzing certified reference materials: GBW – 07601 (human hair) and NIST 1577B (bovine liver). As follows from the results presented in Table 1 within certified ranges or information values, 16 elements were fitted (As, Au, Ba, Br, Ca, Co, Cu, La, Mn, Mo, Na, Sb, Sc, Se, Sm and Zn) in human hair and 13 elements (Br, Ca, Co, Fe, K, Mn, Mo, Na, Rb, Sb, Se, V and Zn) in bovine liver. Fe and Hg concentration determined in the hair sample and Cl concentration in the bovine liver sample fitted within an interval of $\pm 20\%$ from the certified value. Results obtained for Ag, Mg and S in hair and Al and S in bovine liver samples show poor accuracy. This can be attributed to the fact that these elements were very close to the detection limit.

Sample Analysis

A statistical summary of the data obtained for hair and nail samples is presented in Tables 2 and 3. The high values of standard deviation and range in the case of hair and nail analysis suggest a large spread of concentration within all groups of citizens. This is due to a number of parameters – sex, age, dietary habits, the state of the environment in the residential area and occupational exposure influencing the concentration of elements in the examined media.

Concerning the skewness, it can be observed that most of the data is characterized by distribution differing from normal (the values of skewness differ much from "0").

Discussion of Results

To assess whether there are statistically significant differences in element concentrations in hair and nails between inhabitants of the factory neighborhood and the control district, a non-parametric Mann-Whitney U-test was used.

As follows from data presented in Table 4, statistically significant differences ($p < 0.05$) in concentrations of elements in nail clippings collected from citizens of districts close to the fertilizer factory and from people living in the control district were found for Mg and Sb (higher for the control district compared to the Nowy Port district) and for Br, Rb, Sb and Ti (higher for the control district in comparison with the Przerobka district). Moreover, statistically significant differences were found in the elemental composition of nail clippings collected from both of the potentially exposed groups of citizens. People from Nowy Port have higher concentrations of Br, while citizens of

Table 1. Results of reference materials analyses.

Element	GBW – 07601 (hair) [mg/kg]		NIST –1577B (bovine liver) [mg/kg]	
	Analysis (x±SD), N=6	Certified value	Analysis (x±SD), N=6	Certified value
Ag	0.061±0.016	0.029±0.0061	<d.l.	0.039±0.007
Al	19800±240	-	10.30±0.38	3 ^a
As	0.2578±0.0075	0.280±0.040	-	-
Au	0.00237±0.00010	0.0025 ^a	-	-
Ba	15.79±0.61	17±1.0	-	-
Br	0.295±0.019	0.36 ^a	9.788±0.066	9.7 ^a
Ca	3100±170	2900±200	126±16	116±4
Cl	27±13	35 ^a	2900±20	2780±6
Co	0.0607±0.0054	0.071±0.0078	0.2408±0.0057	0.25 ^a
Cu	12.8±5.6	10.60±0.70	164.7±1.4	160±8
Fe	44.2±2.1	54.0±6.0	183.4±1.1	184±15
Hg	0.4412±0.0094	0.36±0.050	-	-
K	<d.l.	20 ^a	9960±110	9940±2
La	0.0466±0.0025	0.0490±0.0080	-	-
Mg	548±41	360±29.8	637±24	601±28
Mn	6.47±0.32	6.30±0.48	10.61±0.25	10.5±1.7
Mo	0.073±0.036	0.073±0.012	3.50±0.16	3.5±0.3
Na	156.8±1.8	152.0±10.0	2420±24	2420±6
Rb	-	-	12.67±0.17	13.7±1.1
S	53900±1700	43000±200	9280±830	7850±6
Sb	0.0990±0.0052	0.095±0.012	<d.l.	0.003 ^a
Sc	0.00751±0.00032	0.008±0.0010	-	-
Se	0.634±0.013	0.6±0.03	0.726±0.016	0.73±0.06
Sm	0.01031±0.00090	0.012 ^a	-	-
V	-	0.03 ^a	0.1105±0.0072	0.123 ^a
Zn	192.5±2.3	190±4.9	123.1±1.1	127±16

a – information values

Table 2. Hair data statistics.

Element	Mean			Median			Range [mg/kg]			SD			Skewness		
	NP	PS	C	NP	PS	C	NP	PS	C	NP	PS	C	NP	PS	C
Ag	0.19	0.28	0.33	0.17	0.21	0.16	0.060–0.42	0.11–0.90	0.064–1.7	0.11	0.23	0.48	0.80	2.6	3.0
Al	15	6.7	9.8	10	5.1	8.3	4.7–41	2.4–19	5.2–21	12	4.9	4.7	1.4	2.2	1.4
As	0.056	-	0.028	0.028	-	0.026	0.0075–0.25	-	0.0070–0.080	0.073	-	0.019	2.2	-	2.5
Au	0.011	0.039	0.0058	0.0049	0.012	0.0075	0.0012–0.034	0.0016–0.14	0.0014–0.0082	0.012	0.051	0.0024	1.1	1.4	-0.77
Ba	2.8	2.5	2.8	2.9	2.2	2.0	1.7–3.6	1.4–4.9	1.3–6.6	0.54	1.0	1.7	-0.79	1.9	1.6
Br	2.7	13	18	1.5	0.51	0.98	0.25–14	0.14–115	0.50–144	4.0	36	43	2.8	3.1	3.0
Ca	2100	3200	2700	880	1800	650	140–5700	210–12000	150–9500	2100	3500	3700	0.49	2.2	1.2
Cl	800	530	580	750	140	520	40–1800	20–3500	31–2000	650	1100	580	0.42	2.9	1.5
Co	0.052	0.021	0.086	0.013	0.013	0.022	0.0040–0.24	0.0070–0.086	0.0039–0.68	0.073	0.024	0.20	2.0	2.7	3.2
Cu	13	18	13	13	15	10	8.0–22	8.4–29	7.0–33	4.0	7.7	7.5	1.00	0.48	2.1
Fe	16	6.6	14	11	6.0	6.8	6.3–50	3.2–10	3.0–62	13	2.6	18	2.0	0.29	2.4
Hf	0.013	0.015	0.016	0.0065	0.006	0.016	0.0041–0.067	0.0036–0.074	0.0040–0.030	0.019	0.021	0.0088	2.9	2.9	0.20
Hg	0.57	0.42	0.37	0.49	0.41	0.36	0.31–1.0	0.22–0.70	0.15–0.53	0.22	0.15	0.13	0.93	0.62	-0.36
I	0.37	0.43	0.25	0.22	0.23	0.16	0.11–1.3	0.095–2.2	0.060–0.82	0.35	0.62	0.22	2.2	2.9	2.0
La	0.014	0.011	0.027	0.0090	0.0053	0.025	0.0040–0.038	0.0030–0.046	0.0050–0.066	0.012	0.013	0.022	1.6	2.5	0.63
Mg	93	170	100	55	97	27	21–270	22–890	13–360	80	260	140	1.1	2.9	1.3
Mn	0.68	0.31	2.2	0.37	0.38	0.26	0.080–2.0	0.025–0.61	0.050–16	0.62	0.22	4.8	1.0	-0.20	3.0
Na	18	83	14	18	8.6	12	3.1–47	4.3–580	2.8–37	15	180	11	0.76	2.9	1.0
S	48000	49000	51000	48000	50000	51000	45000–52000	46000–51000	42000–57000	2000	1700	4000	0.28	-0.99	-0.83
Sb	0.095	0.022	0.031	0.032	0.014	0.024	0.0072–0.43	0.0038–0.073	0.017–0.063	0.14	0.021	0.016	2.1	1.7	1.2
Sc	0.0015	0.00075	0.00097	0.00090	0.00067	0.00085	0.00011–0.0042	0.00040–0.0016	0.00025–0.0023	0.0013	0.00033	0.00065	1.2	1.7	0.86
Se	0.32	0.27	0.30	0.33	0.30	0.35	0.14–0.44	0.070–0.40	0.070–0.45	0.089	0.099	0.14	-0.48	-0.85	-0.78
Si	3900	2300	3200	3100	1700	2900	60–10000	810–6700	60–7300	3000	1800	1900	1.1	2.2	0.65
Ti	5.8	1.9	3.3	3.4	1.7	2.2	2.1–17	1.0–3.5	1.1–7.8	5.3	0.85	2.5	1.6	0.74	0.93
V	0.059	0.015	0.017	0.020	0.012	0.015	0.013–0.35	0.0047–0.035	0.0067–0.052	0.10	0.010	0.013	3.0	0.99	2.4
W	0.053	0.029	0.037	0.041	0.022	0.029	0.018–0.13	0.011–0.065	0.010–0.089	0.035	0.019	0.024	1.4	1.1	1.2
Zn	210	210	210	180	200	190	120–480	120–300	160–290	96	67	42	2.7	0.080	1.0

Table 3. Nail data statistics.

Element	Mean			Median			Range [mg/kg]			SD			Skewness		
	NP	PS	C	NP	PS	C	NP	PS	C	NP	PS	C	NP	PS	C
Al	23	31	21	20	18	12	0.34-66	0.45-123	1.0-59	17	37	19	1.4	2.0	1.1
As	0.048	0.054	0.073	0.050	0.046	0.050	0.015-0.086	0.016-0.11	0.031-0.21	0.025	0.028	0.054	0.045	1.0	2.0
Au	0.0055	0.031	0.023	0.0019	0.0050	0.0080	0.00050-0.020	0.00088-0.18	0.0011-0.088	0.0063	0.056	0.032	1.4	2.4	1.4
Br	1.1	0.78	1.1	0.99	0.82	1.2	0.47-1.8	0.48-1.1	0.35-1.5	0.40	0.19	0.36	0.69	-0.33	-0.85
Ca	1300	1300	1400	1300	1200	1300	630-2000	650-2300	830-2100	400	560	410	0.27	0.56	0.33
Cl	1500	1000	1600	1500	980	1400	670-2800	300-1700	430-3500	660	550	1000	0.65	0.19	0.80
Co	0.035	0.030	0.051	0.027	0.031	0.031	0.0077-0.072	0.010-0.050	0.0050-0.18	0.022	0.012	0.063	0.43	-0.10	1.7
Cr	0.44	0.57	0.81	0.33	0.39	0.46	0.065-1.2	0.18-1.4	0.11-3.6	0.33	0.47	1.0	1.4	1.0	2.5
Cu	11	7.0	7.7	5.6	6.3	5.5	2.5-49	2.2-14	2.4-24	14	4.0	6.1	2.7	0.75	2.0
Fe	23	16	50	22	17	14	7.2-65	8.7-29	5.9-340	16	6.0	98	2.0	0.97	3.2
Hg	0.59	0.73	0.47	0.53	0.71	0.39	0.43-1.0	0.17-1.4	0.23-0.85	0.18	0.35	0.22	1.7	0.24	0.74
K	610	580	1100	600	510	1100	100-1500	54-1200	160-2300	390	390	710	1.2	0.60	0.32
La	0.040	0.060	-	0.030	0.030	-	0.011-0.16	0.012-0.34	-	0.041	0.099	-	2.8	3.1	-
Mg	130	160	180	130	140	170	50-160	110-280	98-330	31	51	64	-1.7	1.8	1.4
Mn	0.57	0.48	1.0	0.44	0.39	0.58	0.085-1.1	0.17-1.1	0.25-2.4	0.33	0.28	0.74	0.34	0.97	0.94
Na	590	520	970	370	320	780	210-1700	130-1100	220-2800	470	400	750	1.8	0.66	1.7
Rb	1.0	0.89	1.2	0.81	0.86	1.3	0.60-1.9	0.43-1.7	0.79-1.8	0.41	0.35	0.29	0.92	1.4	0.12
S	35000	34000	34000	34000	34000	35000	23000-51000	28000-49000	27000-41000	7300	6400	4200	0.91	1.5	0.051
Sb	0.029	0.042	0.85	0.024	0.038	0.95	0.013-0.065	0.022-0.067	0.016-1.6	0.015	0.017	0.58	1.5	0.55	-0.61
Sc	0.0030	0.0024	0.0034	0.0021	0.0024	0.0022	0.0011-0.010	0.00060-0.0046	0.00075-0.0076	0.0026	0.0011	0.0024	2.4	0.39	0.47
Se	0.49	0.46	0.49	0.49	0.49	0.48	0.35-0.58	0.32-0.56	0.39-0.65	0.069	0.076	0.079	-0.44	-0.70	0.77
Si	4700	6400	5500	5500	5800	4000	160-11000	270-21000	230-17000	4300	6700	5200	0.39	1.4	1.3
Ti	8.3	6.4	13	7.8	6.0	10	2.2-17	2.0-13	2.6-41	4.6	2.9	10	0.73	1.0	1.9
V	0.073	0.048	0.058	0.060	0.044	0.038	0.020-0.21	0.011-0.083	0.015-0.16	0.065	0.021	0.043	1.5	0.076	1.4
Zn	110	120	120	100	120	110	92-170	82-160	99-170	23	22	21	1.6	-0.065	1.3

the Przerobka district, in turn, have higher concentrations of Sb in nails.

In the case of hair samples, statistically significant differences between Nowy Port and the control district citizens were found for S and V content (S-higher, while V lower in control district's citizens hair). Significant differences between the Przerobka and the control districts were also found concerning Al, Au and La content (Al

and La were higher in the control district's citizens' hair). Moreover, statistically significant differences were found in the elemental composition of hair samples collected from both of the potentially exposed groups of citizens. Analytical data shows that citizens of the Nowy Port district have significantly higher concentration of Al, Fe, Sb, Ti and V in hair than people living in the Przerobka district.

Table 4. Statistical test for nail and hair data.

Element	Nails			Hair		
	NP – C	PS – C	NP – PS	NP – C	PS – C	NP – PS
Ag	-	-	-	0.67	0.50	0.31
Al	0.45	0.67	1.0	0.41	0.041	0.017
As	0.28	0.36	0.83	-	-	-
Au	0.11	0.89	0.16	0.97	0.038	0.20
Ba	-	-	-	0.16	0.86	0.084
Br	0.58	0.041	0.049	0.95	0.091	0.23
Ca	0.74	0.72	0.92	0.82	0.26	0.32
Cl	0.87	0.17	0.12	0.41	0.26	0.07
Co	0.79	0.62	0.89	0.67	0.18	0.62
Cr	0.45	0.83	0.53	-	-	-
Cu	0.84	0.94	1.0	0.55	0.14	0.14
Fe	0.87	0.89	0.40	0.14	0.36	0.0092
Hf	-	-	-	0.72	0.13	0.72
Hg	0.11	0.067	0.36	0.053	0.57	0.078
I	-	-	-	0.13	0.36	0.72
K	0.16	0.14	0.83	-	-	-
La	-	-	-	0.20	0.029	0.18
Mg	0.049	0.46	0.23	0.34	0.11	0.62
Mn	0.14	0.057	0.48	0.97	0.48	0.20
Na	0.14	0.11	0.67	0.49	0.65	0.94
Rb	0.14	0.015	0.70	-	-	-
S	0.95	0.65	0.78	0.022	0.13	0.11
Sb	0.0078	0.049	0.035	0.12	0.057	0.029
Sc	0.67	0.55	0.94	0.38	0.60	0.14
Se	0.72	0.67	0.50	0.82	0.28	0.20
Si	0.77	0.78	0.57	0.82	0.12	0.091
Ti	0.20	0.024	0.40	0.08	0.18	0.0049
V	0.90	0.92	0.89	0.026	0.62	0.020
W	-	-	-	0.22	0.50	0.057
Zn	0.39	0.53	0.28	0.36	1.0	0.70

Significant differences in the elemental composition of hair and nails found between citizens of the factory neighborhood and the control group are hard to explain. The elevated level of V concentration in hair from people living in the Nowy Port district may come from fertilizer production. However, it must be noted that V also has other sources in Gdańsk's environment: an oil refinery and a thermal – electric power station. None of the other differences can be explained by the fertilizer factory's impact. Elements characteristic of the phosphatic fertilizer plant, like La or S, were on a higher level in the control district's citizens' biosamples than in the potentially exposed districts, while other elements typical of the plant, like uranium and REE's, were under the detection limit. This is why the potentially existing impact was harder to detect. Most likely there are other sources that influence the elemental concentration of hair and nails: sex, age, dietary habits, medicine and occupational exposure.

Correlation of Element Levels Between Hair and Nails

A correlation of element levels between hair and nails is rather rarely discussed in literature. Usually only correlations for hair and blood element levels are presented. In the case of hair and nail element correlation, the most frequently noted correlations concern toxic or non-essential

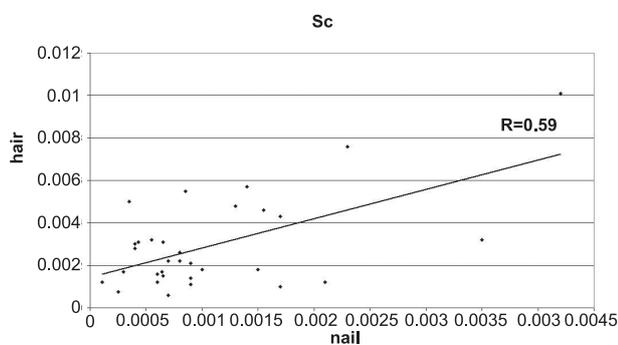


Fig. 2. Correlation between hair and nail Sc content.

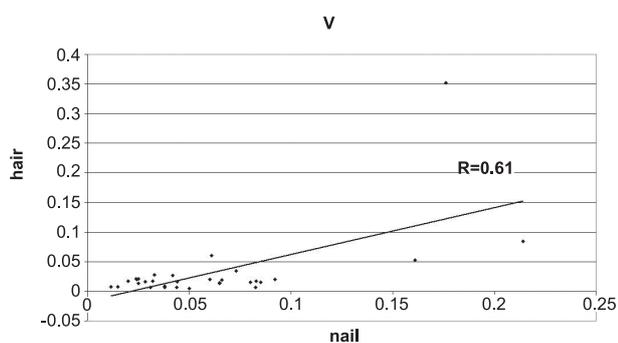


Fig. 3. Correlation between hair and nail V content.

elements. For example, Rodushkin et al. [11] found the strongest correlation for Hg, Cd, Pb, Zr, Hf, Sb and Bi. In turn, a moderate correlation for mercury concentration in hair and nail was observed (0.60) [12]. A similar observation was noted by Gerhardsson et al. [13], for Pb (hair to nail correlation coefficient = 0.52). In the case of our study, the Pearson moment correlation indicated correlations for elements which are not essential to humans (Sc) or which are necessary in small amounts, but an excess can cause health disorders (V). The correlations are rather moderate (Sc=0.59, V=0.61). However, after presenting data on the correlation diagram (Figs. 2 and 3) it can be observed that existing correlations are caused by "outliers". Some of the results can be considered as "outliers" not because of wrongly conducted analyses, but because of the fact that they decide about the existing correlation. After removing one of these points, there is no correlation between nail and hair element content (Sc=0.31, V=0.16).

Inter-Element Correlation

The strongest correlations between concentrations of elements in hair were found for Mg and Ca (0.97), Mg and Se (-0.85), Ca and Se (-0.83), Al and Ti (0.79) and Al and Sc (0.78). In the case of nails, the strongest correlations were found for: Sc and V (0.90), Na and Cl (0.88), Fe and Co (0.81), K and Cl (0.77), Mg and Ca (0.74). It seems that the correlation found between elements in hair and nails arise from similar properties, fulfilled roles in the organism and co-occurrence in the environment.

Variation in Elemental Concentration in Relation to Sex

It was found that sex influences the concentration of elements in hair and nails. On the basis of the Mann-Whitney U-test it was found that concentrations of Ca and Mn is significantly higher in female hair, while concentrations of Cl and Se is higher in male hair. Similar findings for Ca, Cl and Mn were observed earlier [13]. In the case of Zn and Se in nails, concentrations are statistically higher in female nails. The concentration of Ca, in turn, is higher in the case of men. It was interesting to find that Ca concentration is statistically higher in female hair but lower in their nails in comparison with men. The opposite situation occurred in the case of Se concentration – statistically lower in female hair but higher in nails.

Conclusion

It is difficult to state whether a factory impact exists based solely on the significant differences in the elemental composition of hair and nails found between citizens of the factory neighborhood and the control group. Based on the results, it can be said that there is no factory influ-

ence on the citizens or that the potentially existing impact cannot be determined by biomonitoring. The potential impact of the fertilizer plant on Gdansk is even harder to determine because the concentrations of elements characteristic of the fertilizer plant, like lanthanides and uranium, were under detection limits.

No correlation found between hair and nail element content and unclear situation with Ca and Se behavior in human body bring the question if hair and nail elemental concentration reflect the mineral status of the whole body. On the basis of this observation it can be supposed that there may be different mechanisms of incorporation of elements to hair and nails. Most likely, also time of hair and nail growth can be responsible for differences in element composition in the examined media.

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