

Evaluation of soil and agricultural products' contamination in the Spiš District of Slovakia

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Summary

The contamination levels of soil, feed, game, fish and other products of plant or animal origin by selected chemical elements were studied within the high-risk locality, along with their trends within 1991–2003. The evaluation results of 11 872 analyses suggested improving situation of the food chain contamination, with the exception of soil contamination by mercury. To address the prevailing problems, the region will remain a tightly monitored locality, the control and monitoring data will serve as the starting point for risk management.

Keywords

contamination; toxic elements; soil; agricultural products

The district Spišská Nová Ves has been negatively branded by mining and subsequent processing of complex iron and copper ores. There are three major pollution centres within the district: the industrial localities of Rudňany, Krompachy and Spišská Nová Ves. This district is listed among the environmentally most burdened regions of Slovakia because of the amounts of industrial pollutants emitted.

Heterogeneous contamination of the upper soil layers by mercury and other heavy metals (copper, lead, cadmium, zinc) mainly concerns the region of Krompachy where ores have been produced and processed [1].

In the past, the major share of air pollution within the burdened region was coming from the enterprise Kovohuty at Krompachy. Production activities declined within 2000–2002, and but minimum volumes of emissions of pollutants have been generated by the resumed production. There are additional sources of metallurgical and mineral raw materials processing industries, as well as enterprise and local heating systems which contribute to air pollution within the burdened region [2].

Mining, calcining operations and iron ore (and mercury) treatment plants (in the past) caused soil contamination around Rudňany, mainly by mercury, copper, arsenic, cadmium, chrome and zinc. Until recently, Rudňany represented one of

the Europe's largest sources of mercury emissions.

The aim of the paper has been to evaluate the actual concentrations and the trends of contamination by selected chemical elements of the soil, feeds and products of plant and animal origin.

MATERIALS AND METHODS

Time-related trends of mercury, cadmium, arsenic, lead, zinc, chromium and nickel concentrations in the soil, water, feeds, agricultural products and foods were evaluated within the boundaries of the District Spišská Nová Ves. A total of 11 872 samples were analysed within 1991–2003 [3]. Soil was sampled annually by Central Control and Testing Institute of Agriculture. Samples of feeds, feeding water and foods were taken by the staff of the Regional Veterinary and Food Administrations, and analyses were performed by laboratories of the State Veterinary and Food Institute in Košice.

Samples were taken as part of the regular area-wide control as well as part of the monitoring schedule. Their advantage is that they undoubtedly represent an updated information about the status of the pollution of the food chain, a fact of strategic importance from the aspect of risk ma-

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nagement. Some types of monitoring are repeated annually, with samples being taken from the same localities; as part of some monitoring schedule, samples are taken invariably at different locations, returning to the same places always after five years; still others focus on samples from localities as the need may be [4].

The contamination status of the different commodities was assessed in two 5-year cycles and one 3-year cycle. The cycles concerned the periods of 1991–1995; 1996–2000; and 2001–2003. The third cycle was completed in 2005.

The results were summed up and evaluated by the Centre of Foreign Substances Assessment established at the Food Research Institute, established by the Minister of Agriculture [5].

RESULTS AND DISCUSSION

A total of 5 594 samples were analysed within the first 5-year cycle, whereby 514 samples (9.2%) exceeded concentrations limits. The highest proportions of samples showing higher than permissible concentrations were identified for soils (263), mushrooms (89), bee honey and other bee products (57), game (51), fish (11), poultry, beef, milk, feeding mixtures, pig meat, and volume feeds.

During the second five-year cycle, 4 317 samples were analysed, and 213 (4.9%) were found to exceed the concentration limit. The majority of samples showing higher than permissible concentrations concerned those of soils (119), followed by game (35), fish (27), bee honey and other bee products (17), meat, poultry, cereals, drinking water, and bovine entrails.

Higher than permissible concentrations were only identified for as few as 38 samples (representing 1.9%) during the last, 3-year cycle. As before, samples of soils (20), cereals (10), game and mushrooms (4 each) accounted for the highest numbers of them. A total of 1 961 samples were analysed during the third cycle, with 1.9% of them showing higher than maximum permissible concentrations of the contaminants studied. Thus, the proportions of over limit samples were 3.0% smaller within 2001–2003 compared with 1996–2000, and even 7.3% smaller than identified within 1991–1995.

Soil

The most significant soil contaminants within the region monitored included mercury, copper and cadmium. The average contaminant concentrations measured for the different time periods were compared with each other. During the first cycle, higher than permissible concentrations of cadmium and mercury were measured overall for 36.7% and 50.8% of the soil samples, respectively. Soil contamination by cadmium was less intensive during the second cycle (2.0%), whereas that by mercury was more intensive (78.0%). Results of the third cycle analyses showed zero cadmium contamination levels. The proportions of mercury-contaminated soil samples dropped to 58.8%.

The average values of mercury concentrations measured in the soil samples highly exceeded the maximum permissible limit (0.3 mg.kg⁻¹): by 363.6% in 1991, by 401.5% in 1996, and by 59.4% in 2001. Compared with the preceding cycle, average values of mercury in the soil dropped by 1.03 mg.kg⁻¹ during the third cycle (Tab. 1). The most strongly contaminated soil samples were those from the localities Jamník, Rudňany,

Tab. 1. Overview of average concentrations of selected hazardous elements in the soil [mg.kg⁻¹].

Parameter	Monitoring period					
	1991–1995		1996–2000		2001–2003	
	average	deviation	average	deviation	average	deviation
chromium	2.23	11.18	1.07	2.00	0.90	0.90
nickel	5.37	8.60	2.94	6.40	2.78	3.20
copper	13.90	74.86	7.60	12.12	3.80	2.00
zinc	21.50	81.45	11.70	9.50	4.60	7.90
arsenic	2.93	23.80	1.12	1.60	1.00	0.00
cadmium	0.28	1.71	0.13	0.38	0.08	0.09
mercury	1.39	50.58	1.51	31.22	0.48	1.17
lead	15.80	51.12	11.90	18.30	6.80	5.20

Tab. 2. Overview of average concentrations of selected hazardous elements in commodities studied [mg.kg⁻¹].

Commodity	Parameter	Period of monitoring					
		1991–1995		1996–2000		2001–2003	
		average	deviation	average	deviation	average	deviation
fresh volume feeds	mercury	0.0084	0.0490	0.0014	0.0020	0.0018	0.0057
	cadmium	0.5000	3.3730	0.0270	0.0590	0.0120	0.0110
cereals	mercury	0.0038	0.0160	0.0015	0.0026	0.0010	0.0039
	cadmium	0.0270	0.1305	0.0240	0.0570	0.0850	0.1490
potatoes	mercury	0.0012	0.0059	0.0007	0.0019	0.0003	0.0009
	cadmium	0.0080	0.0295	0.0140	0.0180	0.0080	0.0180
beef	mercury	0.0031	0.0080	0.0028	0.0145	0.0016	0.0095
	cadmium	0.0120	0.0410	0.0110	0.0635	0.0080	0.0475
milk	mercury	0.0012	0.0049	0.0010	0.0029	0.0009	0.0005
	cadmium	0.0030	0.0099	0.0020	0.0055	0.0040	0.0075

Markušovce, Spišské Vlachy, Odorín, Vojkovce, Harichovce.

The comparison of the average cadmium values measured in the soil samples suggested a gradual reduction of contamination. Average cadmium concentrations of 0.28 mg.kg⁻¹ were measured in the soil within 1991–1995, representing 93% of the limit value [6], whereas the concentrations only reached 44.5% of limit within 1996–2000, and the value of 0.08 mg.kg⁻¹ Cd measured within 2001–2003 only represented 27.5% of the applicable limit concentration (Tab. 1). Samples with concentrations exceeding the set limit identified during the first two cycles were from localities Hrabušice, Vojkovce, Odorín, Smižany, Harichovce, Rudňany, Spišská Nová Ves, Markušovce, Spišské Vlachy. Soil contamination by cadmium became markedly reduced during the reference period. Within 2001–2003, cadmium concentrations in all soil samples tested already were below the maximum permissible concentration levels, and showed further abating tendency (Tab. 1).

Soil contamination by copper within 1991–1995 was found to be excessive (12 samples with values exceeding the limit); the same applied to arsenic (9 samples with values exceeding the limit), lead (9 samples with values exceeding the limit), zinc (6 samples with values exceeding the limit), chromium (2 samples with values exceeding the limit), and nickel (1 sample with value exceeding the limit). During the subsequent two cycles, the contamination levels by the above elements fell within the respective permissible ranges.

The most pronounced improvement in the soil quality was observed in case of zinc and copper:

the average values dropped by 79% and 73%, respectively. The smallest reduction of average values was found for nickel (improvement by 48%).

The passage of hazardous elements from the soil to plants is favourably influenced by low pH values of the soil. During the first cycle, the average pH value of the soils was 6.56 ranging from 3.90 to 7.80, with the lowest pH values measured for samples taken at the agricultural cooperative Rudňany. During the second cycle, the average pH value was 6.48 (pH range 4.00 to 7.60). The lowest values were measured for samples taken at agricultural cooperative Odorín. The last cycle showed average soil pH values of 6.47, with the range being 5.40–7.40, the most acid soils were those from Iliašovce and Jamník. Thus, a slight soil pH reduction could be noticed overall.

Raw plants sampling

Contamination of cereals by mercury showed a decreasing tendency over the reference period. As compared with 1991–1995, average mercury concentrations dropped to 73.7% within 2001–2003 (from 0.0038 mg.kg⁻¹ to 0.0010 mg.kg⁻¹), with the average values of mercury concentrations measured during cycle 3 only representing 3.3% of the limiting permissible concentrations (Tab. 2). None of the total number of 144 samples analysed showed mercury concentrations exceeding the permissible limit [7]. Elevated chromium (1 sample), nickel (2 samples) and cadmium (11 samples) concentrations were measured in samples of cereals from localities Spišská Nová Ves, Odorín and Spišské Vlachy.

Similarly as in cereals, potatoes too showed decreased average values (by 75% compared to 1991–1995, from 0.0120 mg.kg⁻¹ to 0.0003 mg.kg⁻¹ in 2001–2003, representing but 1.5% of the limit). None of the total number of 63 samples of potatoes analysed showed concentrations of the chemical elements studied exceeding the limiting permissible concentrations.

Raw materials of animal origin

The average contamination levels of beef by mercury ranged between 0.0031 mg.kg⁻¹ in 1991–1995 and 0.0016 mg.kg⁻¹ in 2001–2003, and showed a decreasing trend (Tab. 2). The values measured during the last cycle amounted only to 3.2% of the maximum permissible concentrations. No value exceeding the set limit was detected among the 156 samples analysed. The same applied to cadmium, with the average concentrations dropping by 12.2% within 1996–2000 and by 32.5% within 2001–2003 compared with 1991–1995. The average concentrations measured during the last cycle thus amounted to as little as 8.3% of maximum admissible concentrations. There was no sample with values exceeding the set limit among the total number of 87 samples analysed.

Similarly, the contamination levels of animal products by lead, arsenic, copper and nickel were within the ranges of admissible values. Only three samples could be identified with elevated

chromium concentrations during the first cycle, all taken at the localities Spišské Vlachy and Bystriansky.

None of the milk samples taken once a year within the reference period at the different agricultural farms showed elevated mercury concentrations. The average contamination levels have shown a slight continuing improvement since 1991. For the whole reference period, the lowest average values were measured within 2001–2003 (0.0009 mg.kg⁻¹), representing a reduction by 0.0001 mg.kg⁻¹ as compared with the period 1996–2000. Overall, the average concentrations dropped by 25% as compared with the first cycle, and amounted to just 9% of the limiting permissible concentrations. There was not a single sample out of the 164 milk samples analysed for which concentrations of the other chemical elements studied exceeded the limits.

With respect to cadmium, average concentrations increased again during the last cycle, from 0.0015 mg.kg⁻¹ to 0.0035 mg.kg⁻¹, i. e. by 133% as compared with the preceding cycle. This value however represents as little as 35% of the maximum admissible level.

Feeds

Contamination of volume feeds by mercury did not increase during the reference period. The average concentrations of mercury in fresh volume feeds dropped from 0.0084 mg.kg⁻¹ measured

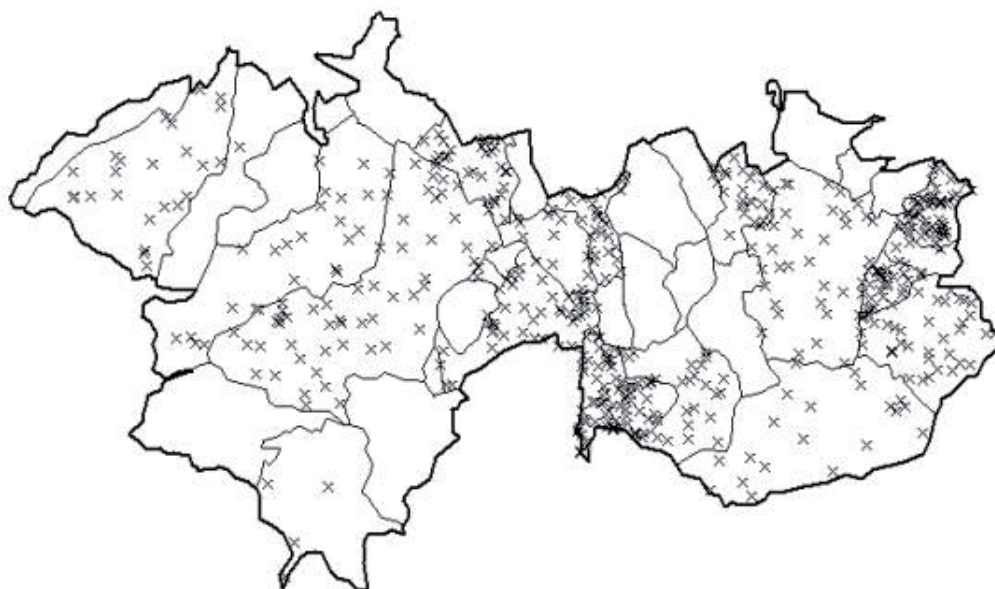


Fig. 1. Localities in the District Spišská Nová Ves with concentrations of hazardous elements exceeding the set limits, within 1991–1995.



Fig. 2. Localities in the District Spišská Nová Ves with concentrations of hazardous elements exceeding the set limits, within 1996–2000.

during the first cycle to $0.0014 \text{ mg.kg}^{-1}$ during the second cycle (by 83.3%). Mercury concentrations increased by 28.6% during the last cycle compared with the preceding one (Tab. 2).

Average cadmium concentrations measured in fresh volume feeds showed a decreasing tendency in the different cycles: compared to 1991–1996, the average values dropped by as much as 97.7% within 2001–2003. The average values measured during the last cycle represented but 1.2% of the maximum permissible value [8]. Permissible cadmium concentrations were exceeded during the first cycle in four samples taken at Spišské Vlachy and Bystrany. Two samples from Bystrany containing excessive lead concentrations were identified during the first cycle.

Regional development

Figs. 1 and 2 illustrate the improving situation in the District Spišská Nová Ves. Reduction of the contamination within the reference period is evident. Most samples with concentrations exceeding the set limits were found during the first cycle among samples from the municipalities Vojkovce, Kolinovce, Krompachy, Markušovce, Spišské Vlachy, Rudňany, Bystrany, Jamník, Hariachovce, Odorín and Spišská Nová Ves (Fig. 1).

During the second cycle, the number of samples with excessive concentrations dropped, as did the numbers of contaminated localities (Fig. 2). This mainly concerned the municipalities

Vojkovce, Kolinovce, Krompachy, Spišské Vlachy, Rudňany, Jamník and Odorín.

Concentrations exceeding the limits could be identified only occasionally during the last cycle, most of them in samples taken from the municipalities Jamník and Odorín. Contamination spikes were observed in samples from Smižany, Hrabušice and Mlynky.

Possible measures were always considered selectively for localities in which excessive contamination was identified. As a rule, the reasons for the contamination were explored and the competent control organization were advised on measures to be taken.

CONCLUSION

The assessment results of the contamination of the various commodities by chemical elements (Cd, Hg, Pb, Cr, Ni, Cu, Zn, As) conducted within 1991–2003 suggested that the contamination levels of the food chain in the District Spišská Nová Ves had a decreasing tendency. To address prevailing problems however, this region will remain subject of close monitoring as before.

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