EVOLUTION OF HEAVY METALS POLLUTION FROM COPSA MICA R. LĂCĂTUŞU*, ANCA-ROVENA LĂCĂTUŞU**

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Abstract

This paper presents a synthetic study on the dynamics of the phenomenon of heavy metals pollution, which took place over 25 years in the Copşa Mică. The main results of research in the pollution of soil, vegetation, animals and the main effects recorded have been reviewed. The first investigations carried out (Răuță et al., 1987, Vădineanu et al., 1991, Toti et al., 1993) outlined an area affected by pollution about 180,750 ha, of which a highly polluted area of 21.875 hectares, in which the least one or two main pollutants exceeding the maximum admissible limits. After two decades, research conducted by Vrinceanu in the year 2005 and published in 2009, outlined a polluted area of 22,565 hectares, in which at least one heavy metal was at a higher content of the alert threshold for a sensitive land use. Therefore, it can be said that there was a slight tendency to increase the areas with polluted soil. Moreover, the latest data published by Damian et al. (2008) on the levels of heavy metals in soils near the emission source Copşa Mică SC Sometra SA show the heavy metals concentration values higher than those reported previously by other authors. However, we can speak about a reduction in the levels of heavy metals in the soils outside the area of pollution itself, the content of heavy metals are below the maximum admissible limits, but are higher than those normally recorded in unpolluted soils. The reduction may be due to a "self-purification" of the soil by the absorption of chemical elements by plants and by leaching. Moreover, reducing industrial production at SC Sometra SA (source of emissions) and increasing plant performance in the purifying emissions sent into the atmosphere contributed to reduce the amplitude of the pollution phenomenon.

INTRODUCTION

The Copṣa Mică area and its surrounding became worldwide known because of the pollution phenomenon generated by emissions from two industrial units that are produce mainly carbon black and metallic chemical elements. The pollution intensity was so great that it was known, in the years that passed, as the most polluted town in Europe.

Industrialization of this area started in the 1930's, when the facilities for the manufacture of carbon black (1935) and zinc production (1939) were built, due to the presence of methane gas, water, and relatively cheap labour. Over the coming

years, industrial activity developed separately in two units, first with chemical profile, and other with non-ferrous metallurgy profile. Between the sixth and eight decades of the last century, production activity recorded maximum intensity without consider, seriously considering environment protection.

In the late 90's, the two industrial units from Copşa Mică renamed SC CARBOSIN SA and SC SOMETRA SA, after the year 1990 currently producing 10 chemical and 15 metallurgical products. Among them, carbon black, methyl methacrylate and methyl polymethacrylate, zinc, lead, gold-silver alloys and others.

Because of the inadequate production to environmental protection, there were sent into the atmosphere and surface water network large quantities of: carbon black, carbon oxides, sulphur oxides, particulates loaded with heavy metals. Pollutants released into the atmosphere, returned to the ground, polluting vegetation, soil and surface water. Oxides of sulphur, carbon and even nitrogen resulting from the combustion processes, and released into atmosphere were subjected to oxidation, hydration, then return to the ground as acid precipitation, causing burns to the vegetation, emphasizing the acid reaction of soils and hence triggering a chain of soil degradation, with serious depreciation of organic matter, alteration of mineral part, the loss of nutrients, surface and deep erosion, and others.

Black carbon released into the atmosphere and then returned to the soil created "sinister landscapes" throughout the area. Initially, black smoke was the trigger factor of attention on the pollution phenomenon at Copşa Mică. It could be "seen", unlike the heavy metals that getting into the soil, plants, animals, and human bodies, and, because they couldn't be "seen", had no warnings in the early stages. Only when animals and people started to get sick, heavy metals came to the attention of specialists in environmental and related disciplines.

The major sources of pollution recorded in the early 90's were black carbon plants, agglomeration plants and sulphuric acid installations [9].

In 1993, the closing of carbon black factory solved the problems of pollution by this chemical. Over time, the atmosphere and houses were clean. It should be noted that, for soil, carbon black was not a real source of pollution, knowing that, normally, carbon is an important element in soil composition.

In 1998, a Greek company, "Mytilines Holdings", acquired plant SOMETRA SA, and made some improvements to facilities for the remediation of gaseous and liquid emissions, significantly reducing production in order to lower pollution intensity from the previous period of 1990. However, there were many periods of time when they reported excedents of the maximum admissible limits for the pollutants constituents of air (sulphur oxides, heavy metals). As a result, the company decided to build a new sulphuric acid plant, with modern technology and a very small pollutant impact on soil. In fact, since February 1, 2009, plant production was temporarily stopped.

If other components of the environment (air, water) have their own dynamic of "self-purification" capacity, soil is the only component that accumulates pollutants, not having the degradation capacity of inorganic pollutants, and, moreover, being a transfer factor of pollutants in plants, water, animal and human bodies. If no special measures are taken, heavy metals can remain in soil for a long time such as decades or centuries [5]. This paper represents a synthesis of the most important research on soil pollution in the Copşa Mică area.

MATERIAL AND METHODS

To achieve this synthesis work, we studied the most significant papers published over the years concerning soil pollution in the area under the influence of the SC SOMETRA SA. Major elements were outlined that helped to present the evolution of the pollution phenomenon. It was interpreted from new points of view, how air pollution has manifested, at Copşa Mică lately, and its effects on vegetation and the soil.

RESULTS AND DISCUSSION

The first data concerning heavy metals and sulphur soil pollution at Copşa Mică were published by Rauţă et al. [7], and were related to forest soils.

The first measurements of areas polluted with heavy metals, performed in the 1990s and reported by Vadineanu et al. [9], Toti et al. [8] showed that the entire area of land affected by pollution was 180,750 ha, of which 149,465 ha of agricultural land and 31,285 of forest land. The polluted territory covers an area eastward to Dumbrăveni and even more, in the south up to Şeica Mare, in the west to Blaj, and in the north up to Cetatea de Baltă. Toti et al. [8] states that the highly polluted area, where at least one from entire heavy metals had content above the maximum admissible limit (MAL) was 21,875 ha, of which 18,630 ha of agricultural land and 3,245 ha of forest land (Figure 1).

In a paper published in 2005, Dumitru [1] presented data determined in three different years (1990, 1993 and 1995) regarding the total content of heavy metals in A horizon (0-20 cm) of arable soils from the Copşa Mică area. The author noted an obvious decrease in the total content of cadmium and copper, mainly, at the maximum and average values. Compared to 1990, in 1993 and 1995 content values have decreased by 13 and 50% for cadmium and by 32, and 51%, respectively for copper (Table 1). For lead and zinc, in 1993 compared to 1990, increases in contents, by 7% and 10% respectively, were recorded. But, in 1995 compared to 1990, decreases in contents, by 28% and 12% respectively, were observed. These changes in the content of heavy metals, especially decreases, must be attributed to the agricultural operations, especially deep ploughing, which allowed the dillution of polluted upper horizon, in a greater mass of soil.

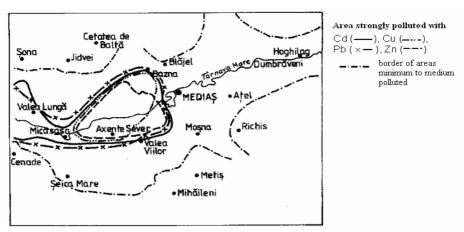


Fig. 1. Distribution of heavy metals polluted areas from the Copşa Mică territory in the year 1985 (adapted from Raută et al., 1987, Toti et al., 1993)

Table 1

Dynamics of heavy metals content (mg·kg⁻¹) in arable horizon (0-20 cm) from

Copşa Mică strongly polluted area (after Dumitru, 2005)

Element	1990			1993			1995		
	min.	max.	med.	min.	max.	med.	min.	max.	med.
Cd	1.5	31.5	8.7	2.5	28.0	7.7	1.8	15.1	4.3
Cu	20	370	75	30	110	51	25	92	37
Pb	25	805	228	85	735	243	35	584	165
Zn	110	765	465	190	1640	514	134	1350	407

If we make a comparative analysis of the heavy metal content ranges, especially the mean values of the upper soil horizon, the result, for forest soils, is an increase of the content in these elements, by 1.1 times for cadmium and lead, 3 times for copper and 2 times for zinc, in 1993 compared to 1987 (Table 2). It should, however, be noted that, in fact, the reported values were significant for earlier periods, when samples were collected. For agricultural soils, in the same period, Toti [8] reported values close to those of forest soils (Table 2). Lăcătuşu et al. [3] calculated close mean values, also for soils from pasture, although, except for zinc, content ranges were narrower than in agricultural soils (Table 2). In all cases, the heavy metal content decreased with soil depth profile, so that at depths varying from 20 to 60 cm, the heavy metal content reached normal values, characteristic to the pedogeochemical fund.

Table 2

Intervals and average values of total heavy metals in the A horizon of soils from Copsa Mica area, according to land use

		Forest	soils	Agricultural	Soils from	
Chemical element	Statistical parameter	Răuță et. al. (1987)	Toti et al. (1993)	soils Toti et al. (1993)	pastures Lăcătușu et al. (1995)	
Cd	X_{min}	0.8	1.0	0.5	4.4	
	X_{max}	22.6	26.0	31.5	11.9	
	X_{med}	5.4	6.2	6.0	6.9	
Cu	X_{min}	6.0	20.0	10.0	15.0	
	X_{max}	77.3	174.0	370.0	127.0	
	X_{med}	22.2	67.0	59.0	39.0	
Pb	X_{min}	20.0	22.0	25.0	37.0	
	X_{max}	552.0	335.0	835.0	476.0	
	X_{med}	116.0	124.0	195.0	171.0	
Zn	X_{min}	51.1	91.0	50.0	178.0	
	X_{max}	791.0	1,646.0	675.0	1819.0	
	X_{med}	219.0	442.0	349.0	539.0	

If we consider measurements of Vrinceanu [10], we find that, in the year 2005, the total polluted area, where at least one heavy metal concentration exceeded the alert threshold for a sensitive use, was 22,565 ha, characteristic of lead pollution in the horizon 0-20 cm.

Other polluted surfaces, characteristic to cadmium (10,320 ha), zinc (7,040 ha) and copper pollution (1,600 ha), overlaped in the area outlined by lead pollution, proportionally to the size of each of the areas, as suggested in Figure 2.

The above data lead us to conclude that reducing the amount of land affected by pollution occurred only when the levels of heavy metals were below the maximum admissible limits. Specifically, if we compare the size of areas contaminated with content levels above the maximum admissible limit or above the alert threshold value, we find that the amount reported in 1991 and 1993 (21,875 ha) by Vadineanu et al., and, respectively Toti et al., was close to that reported by Vrinceanu in 2009 (22,565 ha) for year 2005. This finding shows that, after 1993, the rate of pollution has become lower and lower.

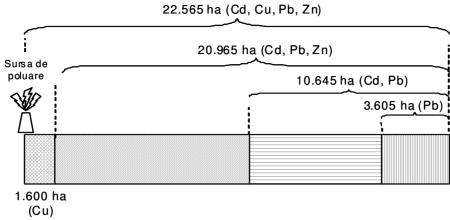


Fig. 2. Areas of land polluted by a single chemical element (lead), two (cadmium, lead), three (cadmium, lead, zinc), four (lead, cadmium, zinc and copper)

Otherwise, if the pollution intensity of SC Sometra SA did not decrease, the polluted area would have increased much more. In support of this assertion, the data obtained by Ozunu et al. [6], which, by modelling the dispersion powders containing heavy metals emitted into the atmosphere by industrial activity of SC Sometra SA and deposited on the soil, showed a significant reduction in deposits and consequently on their heavy metal content. Thus, over 9 years, from 1999 to 2008, the maximum annual deposits decreased in intensity, and their heavy metals content (g/m²) also decreased from 315.02 to 5.90 for zinc, from 191, 56 to 3.69 for lead, and from 2.79 to 0.008 for cadmium. Furthermore, the above named authors calculated the time required for these deposits to achieve the levels of alert and intervention thresholds for sensitive land use if, hypothetically, would not naturally contain such chemical elements. The results showed that the periods of time would be from 250 to 1,000 years for zinc, from 42 to 333 years for lead and from 25 to 167 years for cadmium, depending on the land type (arable or natural, forest cover or pasture) and the nature of the threshold (alert or intervention). Since the last time, the amounts of heavy metals that were deposited on the ground were quite low, allowed the polluted land area not to increase exponentially, and the overall content of heavy metals from soil to maintain, as magnitude order, around levels determined in the years 1991-1993.

Therefore, once again, laboratory experiments [4] verified and confirmed the earlier assertion that soil is an accumulator of pollutants, mostly inorganic, and, by natural means, the disappearance rate of heavy metals in soil is carried on in historical times, of tens, hundreds, thousands of years.

While surface soils contaminated with heavy metals has increased slightly, the area covered with forest vegetation, destroyed due to air pollution has decreased

dramatically over the past 22 years (Figure 3). Satellite images published by Ianculescu et al. [2] highlighted the polluted area restriction in 2001 to only 20% of that found in 1986. The surface of contaminated vegetation decreased continuously over the coming years, so that, degraded forest area in 2008 represented only 2% of the original degraded area.

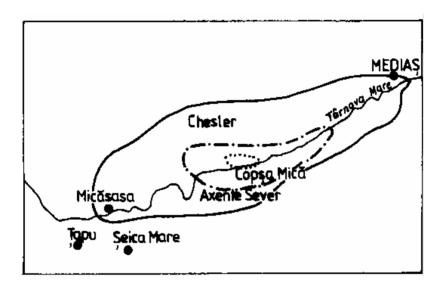


Fig. 3. Degradation of forest vegetation dynamics in the years: 1986 (_____), 2001 (_ _ _ _) și 2008 (_ _ _ _) (according to Ianculescu et al. 2009, using satellite images)

Therefore, the forest has responded very well to reducing air pollution by SC Sometra SA, following the improving of environmental protection facilities and production reducing. However, the authors showed an increased concentration of pollutants in soil, especially cadmium and lead.

CONCLUSIONS

- 1. As a result of the environmental pollution exerted by the two industrial units over the decades, the area of polluted land, assessed in the 1990s, was 180,750 ha, of which 31,285 ha forest fund and 149,465 ha of agricultural land. The heavily polluted area, where at least one or two of the main pollutants exceeded the maximum admissible limits, was 21,875 ha, of which 3245 ha forest fund and 18,630 ha of agricultural land.
- 2. The polluted land area, assessed during 1991-1993, to 21,875 ha, was very close to that reported for 2005, equal to 22,565 ha. The difference of only 690 ha, showed a slight increase in the polluted area in the time since the

- year 1991 to 2005, which demonstrates a systematic reduction in the pollution intensity over time. Otherwise, the polluted area expanded greatly.
- 3. The dynamics of forest vegetation pollution in 22 years (1986-2008) showed a very significant reduction, while the surface covered with contaminated soil remained almost constant, with even a slight increasing trend.

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