

НАУКИ О ЗЕМЛЕ В УЗБЕКИСТАНЕ

GEOSCIENCES IN UZBEKISTAN

TASHKENT 2012

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Академии наук Республики Узбекистан**

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National Committee of geologists of Republic of Uzbekistan

**State Committee of Republic of Uzbekistan
on geology and mineral resources**

**State Enterprise
«Scientific-research Institute of Mineral Resources»**

**Institute of Geology and Geophysics
of the Academy of Science of Republic Uzbekistan
named after Kh.M.Abdullaev**

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Книга рассчитана на широкий круг читателей – ученых и специалистов минерально-сырьевого комплекса, научных учреждений и учебных заведений.

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Collected works of Uzbekistan geologists to 34th International Geological Congress (Brisbane, Australia, 2012) include articles covering a wide range of problems of Earth Sciences reflecting the current state of geological studies in Uzbekistan. The book consists of three sections: ore deposits and metallogeny, oil and gas geology, geophysics, seismology and geocology. The articles reflect the views of the authors on geological development, deep structure and metallogeny of Uzbekistan interior, analyze the state of mineral base of the country and discuss the problem of deposits search, etc.

The book is designed for a wide range of readers – scholars and specialists of mineral complex, scientific and educational institutions.

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HEAVY METALS IN SOILS: DISTRIBUTION, MINERAL FORMS, BIOAVAILABILITY AND IMPACTS ON SOIL BIOGEOCHEMICAL PROPERTIES

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Abstract

The present study examined air pollution effects on soil health applying microbiological parameters. It was carried out near the Angren-Almalyk mining and industrial area in a semiarid region of Uzbekistan. This area was selected in order to establish a national monitoring program for assessing environmental condition of areas remote but downwind from greater emission sources. Moreover, little information exists about how air pollution affects microbiological functioning of soils in semiarid and arid regions of the world, and especially those of Central Asia. Soil samples were collected in May 2005 along a 20-km NE–SW river valley transect down-wind from the industrial complex. Soil chemical analyses included electrical conductivity, pH, water soluble Na, Ca, and K, total soluble nitrogen, and mineralizable nitrogen content upon a 1:2 digestion by deionized water. Major elements and heavy metal inventory in solids was measured by X-ray fluorescence and atomic absorption spectrometry. Microbiological ecosystem properties were assessed by biological indicators such as basal respiration (RB), microbial biomass related C and N contents, and microbial community functioning coefficients like the metabolic quotient $q\text{CO}_2$. There was a significant spatial dependence and differences for all soil chemical and microbiological parameters tested. The highest contents were found for the relatively volatile metals Zn ($\leq 1,136$ mg/kg) and Pb (≤ 373 mg/kg) in upper soil layers near the power station suggesting that the metal pollutants are derived from local stack emissions. Soil microflora was obviously affected by heavy metals. Significant positive correlations ($p \leq 0.001$) were found between the metal content, RB, and $q\text{CO}_2$, while a negative one was found for the mineralizable N and C_{mic}/C_{org} ratio. A high total number of nematodes was found only most distant from the industrial emission sources. The results disclosed remarkable spatial dependence not only of the heavy metal impact onto the soil but also of microbiological soil properties in the study area. The latter suggests bioavailability of the anthropogenic metals in the soil affecting the soil microbial community. This is suggested by less biomass formation and higher $q\text{CO}_2$ values in heavy metal-contaminated compared to less-polluted soil plots. Knowledge of these spatial ecosystem functioning patterns and dependence could be very useful in determining and delineating specific land use and management programs that would be suited and feasible for the highly polluted area. Results of this study can be utilized to develop a monitoring program that may quantify harmful effects on the soil health and impact of any future remediation activities. Studies on the relationship between soil biota and pollution levels have raised the question regarding the status of natural soil microbial health, stressing the importance of background data of environmental conditions, and elucidating the importance of this environmental monitoring approach even in semiarid and arid regions. Soil microbiological parameters, in particular the metabolic quotient $q\text{CO}_2$ as one of the most sensitive bioindicators identified for that region, should clearly become part of the national environmental monitoring program.

Key words: Environmental geochemistry, heavy metals, soil pollution, bioindicators, soil microorganisms, Angren-Almalyk industrial area.

Introduction

The soil samples, which are heavily impacted by aerial emissions from the coal burning and metal-processing industry of the Angren-Almalyk industrial area, were the main subject of our intensive investigation. Accumulation and persistence of heavy metals in surface soil layers, their bioavailability and potential toxicity were the main reasons for conducted research. Knowledge of chemical mobility of heavy metals is fundamental to understanding their toxicity, bioavailability, and geochemical behavior. In first part of our studies we have used two different methods, i.e. mineralogical means and sequential extractions, were employed to analyze the total contents, existing states, and chemical forms of heavy metals in soil. Also, we have used the stable metal isotope signature for separating anthropogenic (airborne) metals from geogenic (lithogenic) ones in bulk soil, soil size fractions. This approach is well established for Pb and tested for Cu, Zn, Cr. The obtained data demonstrate that the Angren Power Plant (APP) and Almalyk Mining Metallurgical Combine (AMMC) are major sources for Pb, Zn, Cu, Cd, and As enrichment in soils of study area. Highest contents above mentioned metals were determined in the upper soil layer (0-10 cm) near the sources of pollution. The concentrations and forms of Cr, Ni, and Co in contrast suggest that the concentrations of these metals in soil were derived from the lithogenic background. Microprobe observations have shown that the studied heavy metals (Cu, Zn, Pb, Cd and As) are associated with two major forms in the contaminated soils. Metals bound to fine grains of sulphide ore minerals, occasionally covered with weathering rims of secondary ore minerals (sulphates or carbonates) can be related to contamination by mining activities. Spherical metallyferrous particles can be found in smelter-impacted areas. Therefore, we have divided enrichments of all studied heavy metals into two groups: 1) anthropogenic (airborne) source: Pb, Zn, Cu, Cd and As U; 2) lithogenic (geochemical) source: Cr, Ni and Co.

The influences of heavy metals (Cu, Pb, Zn, Cd and AS) on soil microbial and nematode population characteristics along the pollution gradient in Angren-Almalyk industrial area was investigated in detail. Obtained data has shown intensive effect of mining industry to soil ecosystem. Results on the influences of heavy metals (Cu, Pb, Zn, Cd and As) on soil microbial and nematode characteristics in Angren-Almalyk mining industrial area along the two deposition transects illustrate their negative response to heavy metal pollution. Studies on the relationship between soil biota and pollution levels have raised the question regarding the importance of natural soil abiotic properties, stressing the importance of background data of environmental conditions, and elucidating the importance of further studies on this subject. Obtained data on nematode population and community structure, ecological indices and other indicators including microbial biomass, metabolic quotient in soils of Angren-Almalyk mining industrial area show bright future of this techniques as a tools for environmental biomonitoring and bioremediation measures.

The study sites are situated in 65km (Almalyk) and 114km (Angren) distances from the Tashkent along the Akhangaran river valley (the right tributary of the Sir-Darya river), in foothills, between Chatkal and Kurama mountain ranges (Western Tien-Shan), extending into the southeast part of the Tashkent region of the Republic of Uzbekistan. The industrial complexes includes a metal mining and processing enterprise, metallurgical and chemical plants located in flat bottom of the Akhangaran river valley near the city

of Almalyk (40°50' N - 69°34' E), and, also, a coal mining, power plant and resin industries located in upper side of the Akhangaran river valley near the city of Angren (41°01' N -70°09' E). The Almalyk mining and smelting complex (AMSC), and the Angren coal burning power plants, are the major sources of air and soil pollution in this area. Almalyk city was founded in 1951 from several settlements exploiting the rich nonferrous-metal resources of the Qurama Mountains. The city has become an important centre of nonferrous metallurgy and most important industrial center of Uzbekistan. As one of the largest mining companies in Uzbekistan, AMSC produces refined copper, gold, silver, lead, metallic zinc, and other products. It has the capacity to mine and process about 25 Million ton of ore per year, with annual metal-producing capacity of Cu 130,000, Zn 40,000, and Pb 80,000 ton per year (Levine, 1999). Due to a lack of efficient air-treatment facilities for copper smelting, the complex is also a major source of air pollution. According to an environmental report that was prepared in 2000, the Almalyk mining and smelting complex was found to emit about 100,000 tons of toxic substances (sulphur dioxide, carbons, nitrogen oxides, sulphuric acid, heavy metals, arsenic, etc.) per year. This is approximately 13% of all of Uzbekistan's air emissions from stationary sources (The State Committee for Nature Protection, 1998). Angren is the largest coal mining and power producing center in Uzbekistan, which was developed during and after World War II. The Angren power plant (250 MW capacity) works on basis of brown coal from the nearby Angren coal field (production capacity of 2,5 million tons per year, ash content 11-35%). The coal mining and power generation industry in Angren uses old equipment that has not been upgraded since the 1990s. Air pollution control technology is in poor condition and several units need to be modernized. Emissions in the range of about 100000 tons/year over the last few decades have caused severe damage to natural ecosystems of the area, and dramatically increased impact of heavy metal rich dust and fly ash particles in this area. (UNECE, 2000).

The research area represents a mountain-valley area with a large variability of seasonal and daily air temperature and wind direction. Thermal inversions provide cyclic circulation of air masses and cause pendulum distributions of dust and gaz-smoke emissions from the industrial enterprises. The prevalent wind at the study area is in a western and south-western (in Almalyk), and north-eastern (in Angren) directions. An additional feature of the study area location, is that it is surrounded by a chain of mountains, which create poor conditions for air circulation. This further worsens the air pollution and the secondary pollution of soils and vegetation in the valley. Since 1994, the State committee for Geology and Mineral Resources has carried out environmental monitoring in the Angren and Almalyk mining areas. The results showed that surface and groundwater, soil and vegetation are highly contaminated with heavy metals, such as lead, copper, zinc, selenium, wolfram, cobalt, cadmium and arsenic (UNECE, 2000).

The climate is sharply continental for the hull area (the absolute maximum +40°C in summer and -25°C in winter (average annual temperature varying +14° to 15°C). Average annual precipitation 339mm-511mm (96%of this falls for the autumn, winter and spring seasons) (Information Agency Jahon of the Ministry of Foreign Affairs of the Republic of Uzbekistan, 2003).

The vegetation cover along the study sites is dominated by annual and perennial plants, where the most common are: *Astragalus*, *Stipa*, *Medicago* and *Artemisia* genera. The soils at the study area belong to the lithosols (FAO, 2003), with high levels of CaCO₃ contributing to a stable accumulation of heavy metals on top of the soil layer.

Methods

On framework of our studies we have carried out research investigations on heavy metals forms and concentrations in soils along the two downwind transects of Angren-Almalyk mining industrial area and their interactions and impact to soil microorganisms (microbial biomass and soil free living nematodes). First part of our investigations were focused on "Concentrations and forms of heavy metals in soil samples along two downwind transects in Angren-Almalyk mining area" (fig. 1). There were carried out also studies on integrative mineralogy and geochemistry for understanding heavy metal transport in these soils. The aims of our first year studies were (i) to assess the relationship between distance from the emission sources and heavy metal concentrations in soil and to analyze the depth distribution of the metals as indication of their mobility along the downwind transect; (ii) to evaluate the mobile and immobile metal forms, and (iii) to analyze the composition of heavy metals mixtures by means of statistical methods to get hints of common sources of different metals. Soil samples were collected in April 2005 along the two 20-km down-wind transects in Akhangaran river valley, nearby the industrial complexes. In the laboratory, all samples were sieved through a 2 mm mesh sieve and separated into two parts. The first part was stored in a refrigerator at 4°C for the biological studies and was transferred to Bar-Ilan University for further investigations. The second part of soil samples (n=176) were used for chemical and mineralogical studies.

These samples were air-dried, grounded in agate mortar, and homogenized for sequential chemical extraction (AAS, ICP-MS) and preparation of powder pellets for XRF and XRD analyses. QA/QC was performed on basis of certified reference materials (CRM). We have completed the analyses of the total metal concentrations (Cu, Pb, Zn, Cd, Ni, Cr and Mn) and metal forms (mineralogy and chemistry) in all soil samples. Were quantified the mineralogical characteristics, morphology, and chemistry of air-borne spheroids to characterize the mineralogical sources of the soil contamination by heavy metals using JEOL scanning electron microscope (SEM). Sub-samples for mineralogical studies were separated by gravity and size. Particle-size fractionation was carried out by sieving and by sedimentation in aqueous media. The fine grained fraction was subjected to gravity separation and fractionated into (A) the heavy mineral fraction (ore minerals and spherical airborne metal-rich particles) and (B) the light mineral fraction (parent rock minerals). The heavy mineral fraction separated from the fines is embedded in epoxy resin. Element mappings were performed with the polished and carbon-coated thin sections using a JEOL microprobe.

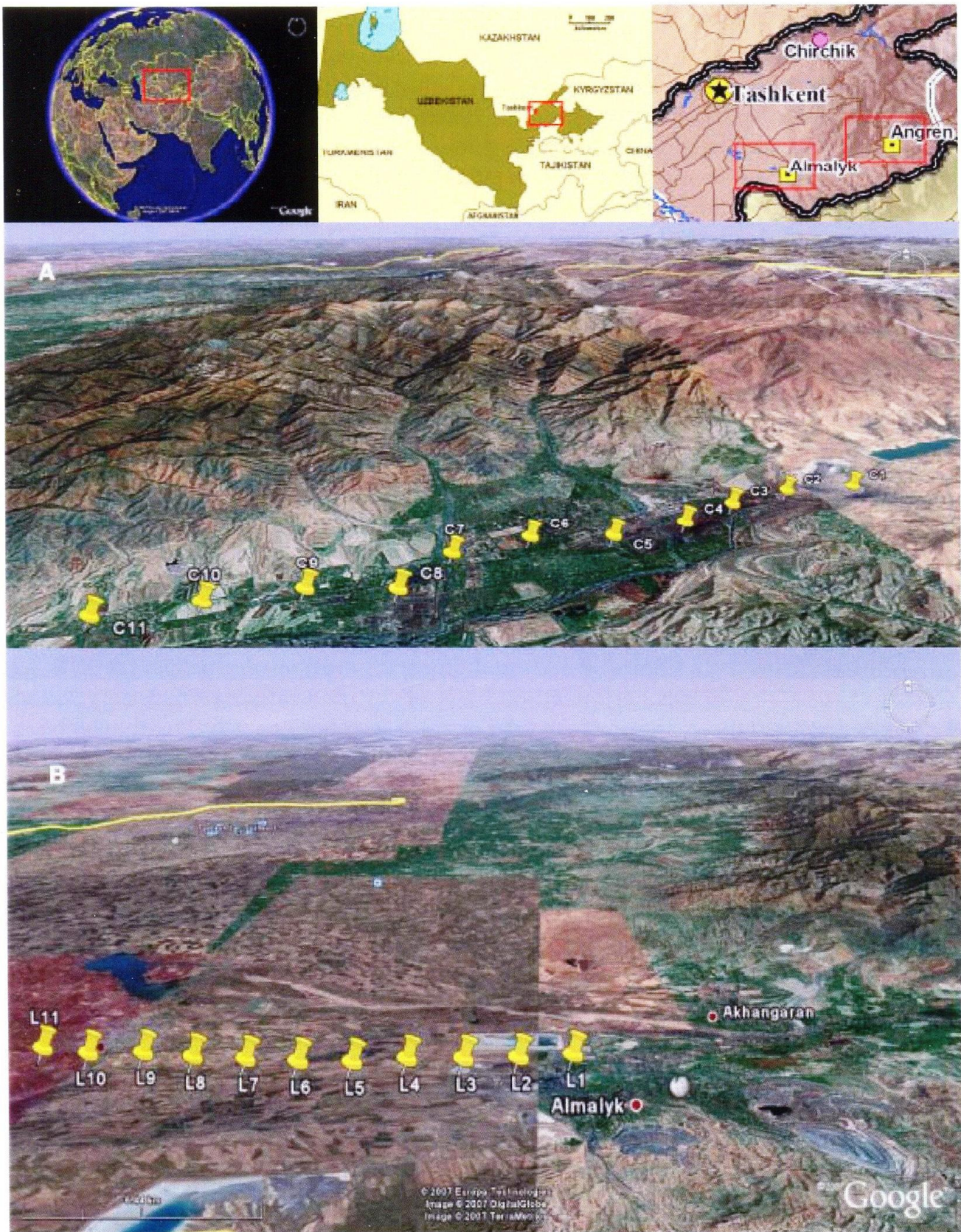


Figure 1. Soil sampling locations in Angren-Almalyk mining industrial area (A – Angren; B – Almalyk).

Heavy metals were also fractionated by the sequential extraction procedure, in which the metal fractions were defined as exchangeable, carbonate-, Fe–Mn oxide-, organic matter-bound and residual fractions. The sequential extraction scheme was developed from that of Tessier *et al.* (1979) and the same terminology is retained. Since, this is the method that has given results about the possible bioavailability of the elements and their

extractability or leachability, it is described in detail. Extraction was carried out progressively on an initial weight of 1.000 gram of soil sample.

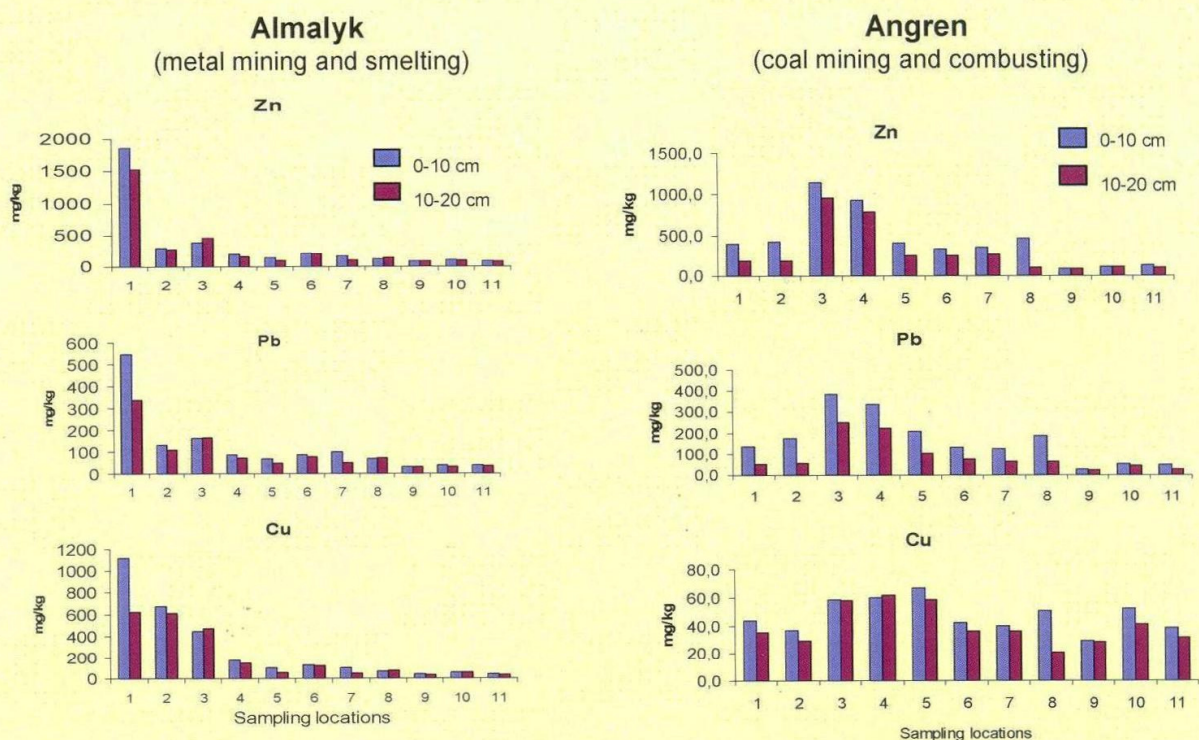
To measure the bioavailable heavy metal fraction we used diffusive gradients in thin films (DGT) at the maximum water holding capacity (MWHC) of the sub-samples. In particular, we separated the concentrations and effects of geogenic and smelter metals with the help of the stable isotopic signature.

Second part of our intensive studies were focused to determine the influences of heavy metals (Cu, Pb, Zn, Cd and As) on soil microbial and nematode characteristics in Angren-Almalyk mining industrial area along the two deposition transects. Nematode population, community structure, ecological indices, pollution tolerance and other indicators including microbial biomass, metabolic quotient were studied in soil samples. Soil microbial biomass, nematode population and community ecological indices response to heavy metal pollution in Angren-Almalyk mining industrial area were evaluated. Metal pollution indicators and tolerance species are established and recommended for developing biomonitoring and bioremediation measures which planned in future for this region. Microbiological ecosystem properties were assessed by biological indicators such as basal respiration (RB), microbial biomass related C and N contents, and microbial community functioning coefficients like the metabolic quotient qCO_2 .

We have also worked on stable isotope signature of Pb and interactions between plants and microorganisms, and their response to heavy metal contamination. Also some preliminary studies on "Organic pollutants behaviour and concentrations in polluted soils of Angren-Almalyk area" were investigated. "The impact of polycyclic aromatic hydrocarbons (PAHs) and their oxygenated derivatives (OPAHs) on microorganisms in soils of Angren industrial area". There was a significant spatial dependence and differences for all soil chemical and microbiological parameters tested. PAHs and OPAHs concentration in upper soil layers (0-10 cm) were highest near the power station (C and B) followed by A (near coal mine pit) suggesting that these pollutants are derived from local stack emissions. Areas far from these emission sources had progressively lower PAH/OPAH concentration except near rubber factory (F). Deeper soil layer (10-20 cm) had lower concentration than upper soil layer except in location (A). OPAHs followed similar trend as PAHs indicating that they are emitted together with PAHs. Soil micro-flora was obviously affected by PAHs and OPAHs near the pollution source. Highest total number of nematodes and number of plant parasite nematodes trophy group were found most distant from the industrial emission sources with comparatively lower PAH and OPAH concentrations. Positive correlations were found between PAHs and OPAHs concentrations, soil respiration, and metabolic quotient qCO_2 , while a negative one for the mineralizable N, C_{mic}/C_{org} and C_{mic}/N_{mic} ratios.

Results

Heavy metals Geochemistry and mineralogy in contaminated solis. The concentrations of a number of potentially toxic metals varied along the sampling transects, revealing a gradual decrease at increasing distance from the emission sources. The highest levels were found for the relatively volatile heavy metals (Cu: 457-871, Zn: 424-1695, and Pb: 163-441) in soils near the Almalyk metal smelter, and Zn (850-1051 ppm) and Pb (270-320 ppm) in soils near the Angren power plant suggesting that the metal pollutants probably derive from local stack emissions (figure 2).



Note: Sampling location No. 1 is the nearest point to Smelter factory in Almalyk transect. Sampling points No 3 and No 4 are within the Angren power station, No 8 Resin factory along the Angren transect. Length of transects 20 km each, distance between locations 2 km

Figure 2. Distribution of heavy metal (total) concentrations in soil along the deposition gradients in Angren-Almalyk mining industrial area (mg/kg).

Significant differences ($p < 0.05$) were observed between the upper (0-10 cm) and deeper (10-20 cm) soil layers and among the particle size fractions for Cu, Zn, Pb, U, and Th at most sampling locations of both study areas. The concentrations of a number of metals (Zn, Cu, Pb, Cd and As) were higher in the upper soil layer and in the clay fractions at all sampling sites. The distribution and concentrations of some other metals (Co, Cr, Ni, V, and Sc) showed an opposite trend, probably due to the native geochemical background.

Furthermore, heavy metals were fractionated by a sequential extraction procedure, in which the metal fractions were defined as exchangeable, carbonate-, Fe-Mn oxide-, organic matter-bound and residual fractions. Most of Cd was extracted with the exchangeable and carbonate-bound fractions. The highest concentrations of Ni and Cr were found in residual fraction. The speciation of Cu, Pb, Zn, and Cd was related to distance from the emission source and soil depth. The mean contributions of Cu, Pb, Zn and Cd in the exchangeable fraction to the total concentrations were higher at the locations (nearest to the pollution sources) and decreased with increasing distance. Metals bound to carbonates and Fe-Mn oxides increased consistently with depth in first sampling location. There were almost no significant changes for the metal contents in all fractions after 12 km. (figure 3).

Several metal phases such as galena, chalcopyrite, sphalerite, pyromorphite, cerrusite, hydrocerrusite, leadhillite, and anglesite were identified by XRD. Of the crystalline metal phases, the presence of cerrusite, malachite, azurite, hydrocerrusite, and pyromorphite were identified as weathered forms of primary ore minerals at the first sampling location in Almalyk.

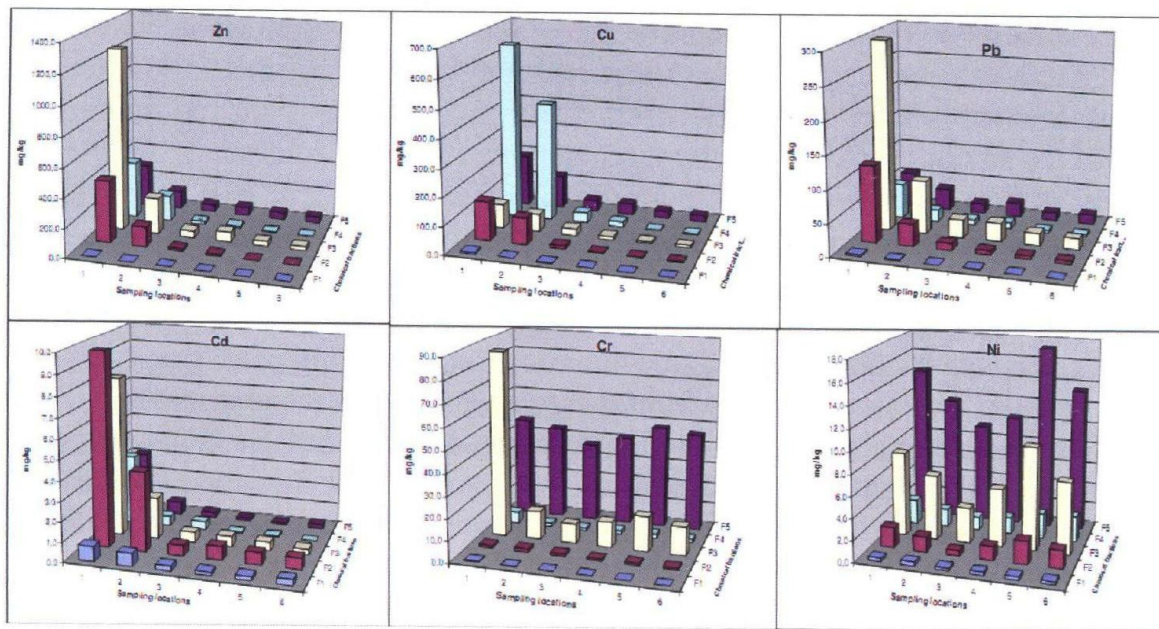


Figure 3. Results from sequential metal extraction procedure, mg/kg (Almalyk).

A lot of grains and spherical particles with bright contrast appeared in the microprobe scans indicating a metal-rich composition. Spherical particles dominated in the soil samples collected near the metal smelters, whereas angular sulfide minerals (pyrite, galena, chalcopyrite, sphalerite) dominated in samples collected near the mine spoils and tailings depository (figure 4).

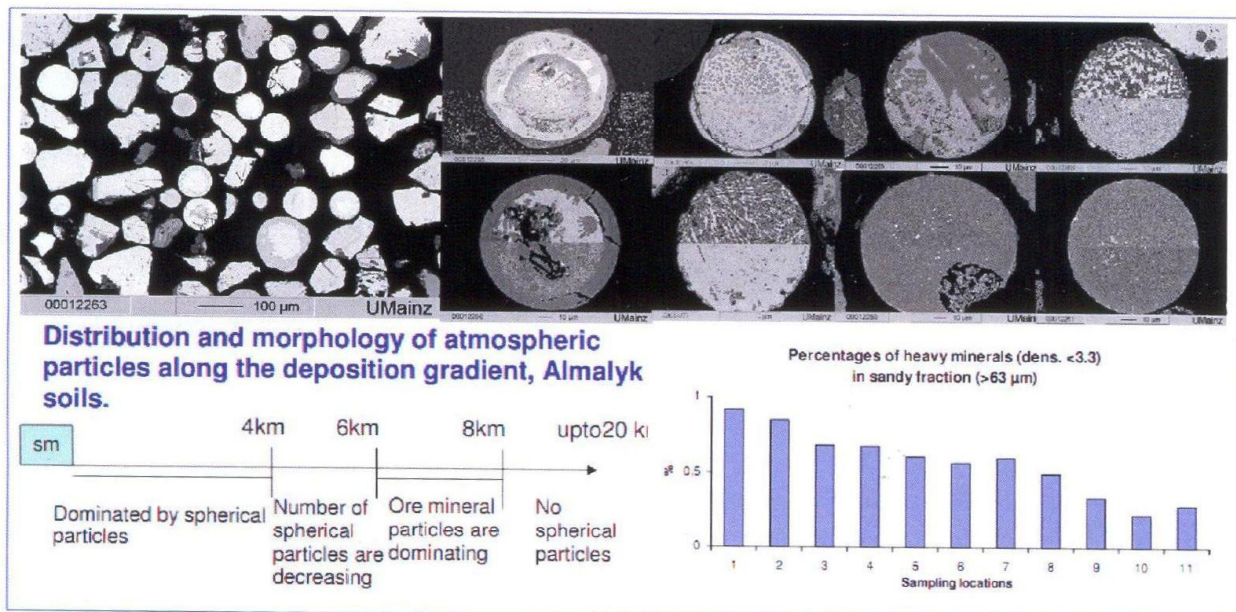


Figure 4. The morphology and internal microstructure of the spherical particles in heavy mineral fraction of the soil samples from the Almalyk, and their distribution along the deposition gradient.

Sampling locations: 1- L1 (100 m distance from the Smelter); 2- L3 (4km distance from the Smelter; 3- L5 (8 km); 4- L7 (12 km); 5- L9 (16 km); 6- L11 (20 km). Chemical fractions: F1- Exchangeable; F2- Bound to Carbonates; F3- Bound to Fe and Mn oxides; F4- Bound to Organic Matter; F5- Residual.

The morphology and internal microstructure of the spherical particles in heavy mineral fraction of the soil samples from the Angren-Almalyk area indicates formation from a pre-existing molten phase, probably emitted by an inefficient air pollution control technique of the smelter in Almalyk and thermal power plant in Angren.

Spherical particles can be divided into: pure metal particles (Cu, Zn, Al), metal-rich cores with silicate rims, small spherical metal sulfide or oxide particles within larger heterogeneous glassy particles, well organized particles with dendrite structure. Fine grains of sulfide ore minerals, covered with weathering rims of secondary ore minerals (sulfates or carbonates), can be related to contamination by mining activities.

Fe was abundant in most particles from the Angren soils, associated with Ti, S or Pb, and Zn. A few bright grains showed a rim dominated by Si and K. Other associations found were Fe+S+Pb+Mn, or S+Pb+Zn+Mn+Fe, or pure Cu or Zn only. The absence of an O peak indicates that Cu, Zn, and Pb are largely associated with sulfides rather than sulfates. Numerous spherical particles showed distinct dendrite-like structures within a dark grey matrix. The matrix is composed largely of Si, Mn, Zn, Pb, Al and Fe or Fe alone, Ti, Cu, Si, and Ca, whereas the bright areas were formed mainly by Fe and Al, presumably in an oxide (spinel) form considering the strong O signal. Most of the spherical particles showed holes inside, or had metallyferrous rims around alumina cores, which indicates formation from a pre-existing molten phase. Occasionally, these technogenic spheres contained angular grains of primary ore minerals.

Percentage of heavy minerals in Angren soils is more than in Almalyk's (1>4). Elemental composition of spherical particles very poor than Almalyk's (almost all of them have elevated content of Fe oxide with less Si, Al, Mn, Ca content). In some particles less contents of Pb, Cu and S were observed. Spherical particles in Angren soils can be divided into 3 groups: a) well organized massive dendritic particles containing Fe oxide in light growing crystals and matrix containing Si, Al, Ca, Fe; b) homogenous spherical particles without any structures containing Fe oxide; c) small microscopic particles coated Fe oxide. Ore mineral particles mainly Hematite, titan-magnetite, magnetite and very less pyrite particles. Many of them covered with secondary ore minerals as Fe hydroxide and carbonates. Calcareous condition of soils in studied areas can stabilize atmospheric particles and prolong their weathering process by covering them with carbonate material (figure 5).

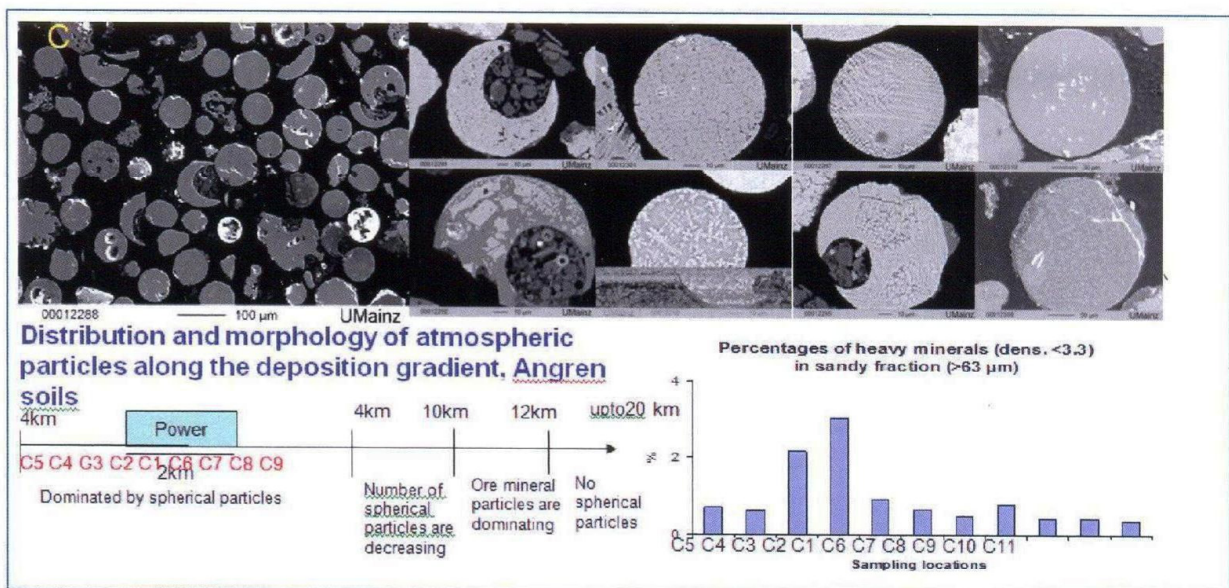


Figure 5. The morphology and internal microstructure of the spherical particles in heavy mineral fraction of the soil samples from the Angren, and their distribution along the deposition gradient.

The Angren industrial coal mining and combustion and the Almalyk metal mining and smelting complexes are major sources for Pb, Zn, Cu, Cd, Zr, Th, U and As enrichment in soils of studied area. Highest contents were determined in the upper soil layer (0-10 cm). The concentrations and forms of Co, Cr, Ni, V, and Sc, in contrast suggest that the concentrations of these metals in soil were derived from the lithogenic background. Heavy metal accumulated particularly in the fine heavy mineral fraction of the Almalyk soils. Microprobe observations have shown that the studied heavy metals (Cu, Zn and Pb) are associated with two major forms in the contaminated soils. Metals bound to fine grains of sulphide ore minerals, occasionally covered with weathering rims of secondary ore minerals (sulphates or carbonates) can be related to contamination by mining activities. Spherical metalliferous particles can be found in smelter-impacted areas. Other particles related to the latter include (i) essentially pure metal particles (Cu, Zn, Al), (ii) metal-rich cores with silicate rims, and (iii) small spherical metal sulphide particles within larger, heterogeneous glassy particles. Morphology and internal microstructure of the spherical particles in the heavy mineral fractions of the soil samples indicates formation from a pre-existing molten phase, probably emitted by an inefficient air pollution control technique of the smelters. Abundance of such particles is critical in that they are known to easily release their metal inventory into the environment.

Biogeochemical Processes in Metal-Contaminated Soils/ Within the biogeochemical part of our investigation we have studied the effect of heavy metal total concentrations and their different forms onto soil free-living nematodes, microbial biomass (C_{mic}) and basal respiration (BR) along the downwind deposition gradients, originating at the Angren-Almalyk mining-industrial area.

Soil moisture was found to be significantly ($p < 0.001$) higher in the deeper (10-20 cm) soil layer than in the upper (0-10 cm) soil layer, with no significant differences between sampling sites. Unlike soil moisture content, organic matter content was found to be significantly ($p < 0.001$) higher in the upper (0-10 cm) soil layer than in the deeper (10-20 cm) soil layer along the sampling sites of both transects. As noticed above the heavy metals (Pb, Cu, Zn, Cd and As) contents along the both transects in Angren-Almalyk mining industrial area were highest near the pollution sources, decreasing along the downwind direction. Contents of As, Zn, Cu, and Pb were found to be maximal at sampling location L1 (in Almalyk) and at locations C3-C4 and C8 (in Angren), decreasing significantly toward by increasing distance from the sources, with no significant difference between the other downwind sampling locations (fig. 6).

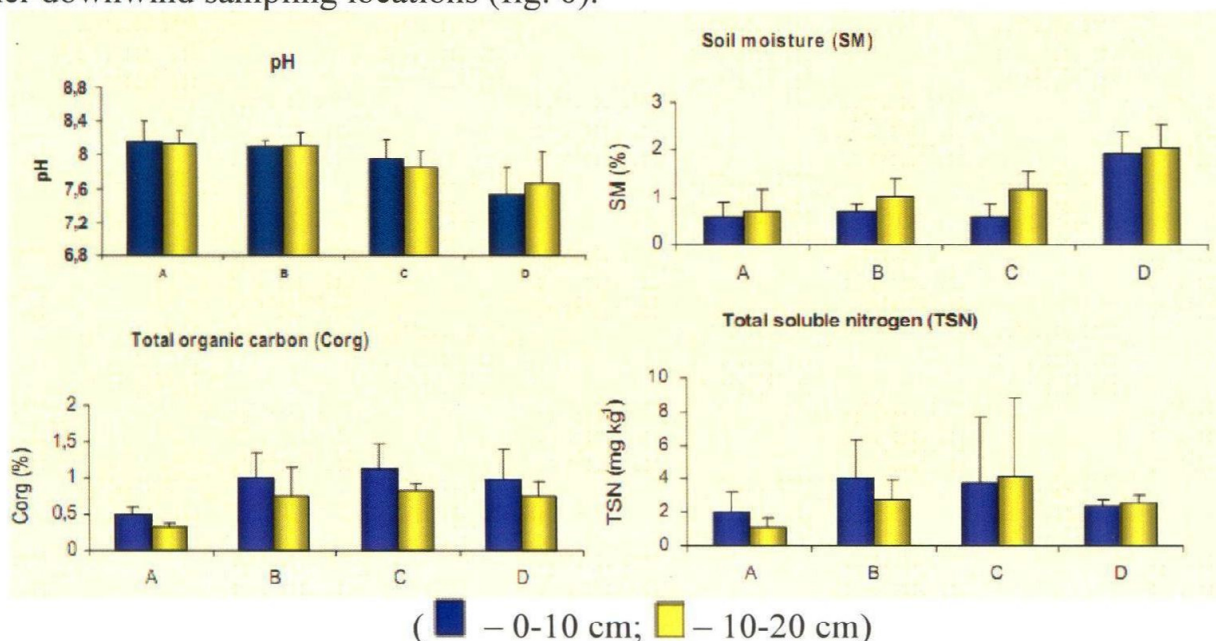
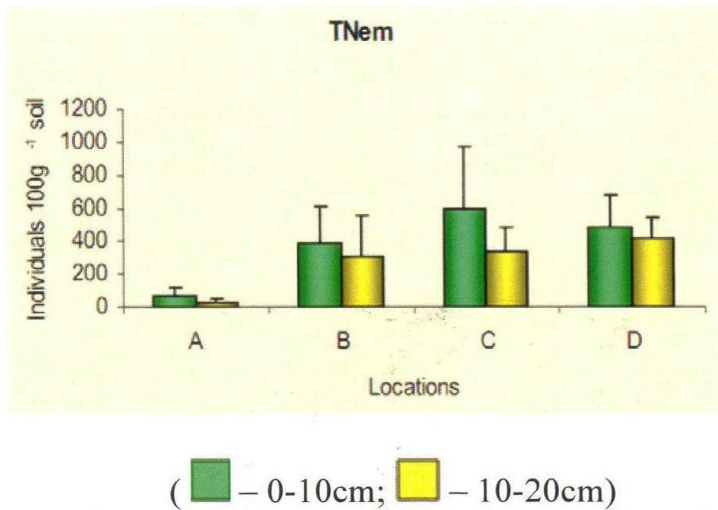


Figure 6. Soil pH, soil moisture, total organic carbon and total soluble nitrogen contents along the deposition gradient at two soil layers in Almalyk.

Twenty nine nematode taxa were identified in the present investigation for Almalyk downwind transect: twelve taxa belonged to the bacterivore trophic group, 5 were fungivores, 7 were plant parasites and 5 were omnivores-predators. The mean density of the soil free-living nematodes increased with distance from pollution source in both soil layers (fig. 7).



(■ – 0-10cm; ■ – 10-20cm)
Figure 7. Distribution of total number of nematodes along the deposition gradient at two soil layers.

Moreover, nematode density in the upper (0-10) cm soil layer showed a gradual increase in TNEM population between sampling locations, yielding significantly high $R^2 = 0.98$ values, a value which decreased to a level of $R^2 = 0.69$ with the increase in distance (20km). In the deeper (10-20 cm) soil layer, the R^2 values obtained for a similar distance were found to be similar ($R^2 = 0.8$). The nematode density was higher in the upper soil layer (from 1.2 to 2.0 times at the different sampling locations) than in the deeper soil layer ($p < 0.02$). The percentage of each trophic group out

of the whole population was found to be affected by the distance from the pollution sources. Trophic group density increased with distance; however, these increases were not always similar along the both transects. Bacterivores (BF) and plant-parasites (PP) in the deeper soil layer and omnivore-predators (OP) in both soil layers, increased consistently from the pollution source to the edge of the study area, whereas BF, PP and fungivores (FF) in the upper soil layer increased to first three locations and then decreased to other locations in Almalyk.

The calculated indices, such as the dominant index (λ), Shannon Index (H), and Richness (SR), were not found to reflect changes in distance from the pollution source in Almalyk. However, the MI and MMI indices exhibited a gradual ($p < 0.05$) increase, with increasing distance from the pollution source. The mean total numbers of nematode males, females, and juveniles per 100 g dry soil at the observed sites were 57.3, 161.9, and 113.9 individuals, respectively. The total number of all three sex groups of nematodes extracted at ST I (pollution source) in Almalyk was significantly ($p < 0.01$) lower than at the other three locations for both soil layers. The mean values of males and juveniles out of the total population were found to remain unchanged in both soil layers, whereas the female population exhibited a significant depth ($p < 0.003$) effect. Moreover, regressions of male and juvenile density in the two soil layers with distance were higher between first three stations than between all four observed stations. Regression of female density was higher between all four observed stations than between the first three sampling locations. The proportion of the three sex groups next to the pollution source was 9:23:1 (male:female:juvenile, respectively). At location C, this proportion was 1:2:3 and at K it was 1:3:1.5. The changes in the total biomass of the soil free-living nematodes (Annex 1, slide 20, Fig. 6A, A') exhibited a gradual and significant ($p < 0.01$) increase from 18.3 to 274.9 μg per 100 g soil in the upper (0-10 cm) soil layer, with increase in distance from the pollution source. A similar trend was found in the deeper (10-20 cm) soil layer, with a minimum value of 14.2 and a maximal value of 133.3 μg per 100 g soil. This was significantly ($p < 0.01$) lower than in the upper soil layer. The contribution of the

different trophic groups to the total biomass was found to be significantly affected by the sampling site and not by the soil layer, except for the omnivore-predator feeding group. Based on the obtained values, the total female biomass (TBf) in the upper (0–10 cm) soil layer was found to be significantly higher ($p < 0.008$) in comparison to the deeper layer and significantly ($p < 0.002$) affected by location. The male as well as juvenile biomasses were found to be significantly affected only by sampling sites along the gradient.

Nematode data obtained from the Angren sampling site also show almost similar responses to pollution with small varieties because of the complicate nature of disturbance (coal ash, resin and metallo-organic) and also nonlinear distribution of pollutions sources along the transect. The effect of pollution on the density and biomass of soil free-living nematodes was found to be highest at the pollution source in Almalyk, with fungivores and plant parasites dominating at the upper and deeper soil layers next to the pollution source. These groups decreased along the transect, yielding domination to bacteria and fungi feeders. The females of the total soil free-living nematodes were found to be the most resistant to heavy metal pollution levels, while the juveniles were found to be sensitive to changes in heavy metals. The Maturity and modified maturity indices, that reflect the degree of disturbance of the soil ecosystem, were found to be the most sensitive indices. The soil microbial biomass, basal respiration and derived microbial indices for soil samples from the Angren-Almalyk industrial area were analysed. The lowest soil microbial biomass and total number of free-living nematodes were found in soil samples near the industrial complexes, with a high heavy metal and weak total organic carbon (C_{org}) content in Almalyk and with elevated contents of heavy metals and organic carbon (C_{org}) in Angren. The highest C_{mic} was found in the soil samples collected 20 km from the pollution source in Almalyk and near the pollution sources in Angren. BR displayed similar results. The derived indices, metabolic quotient (qCO_2) and microbial ratio (C_{mic}/C_{org}), revealed significant differences with distance, confirming environmental stress in the first and second locations (fig. 8).

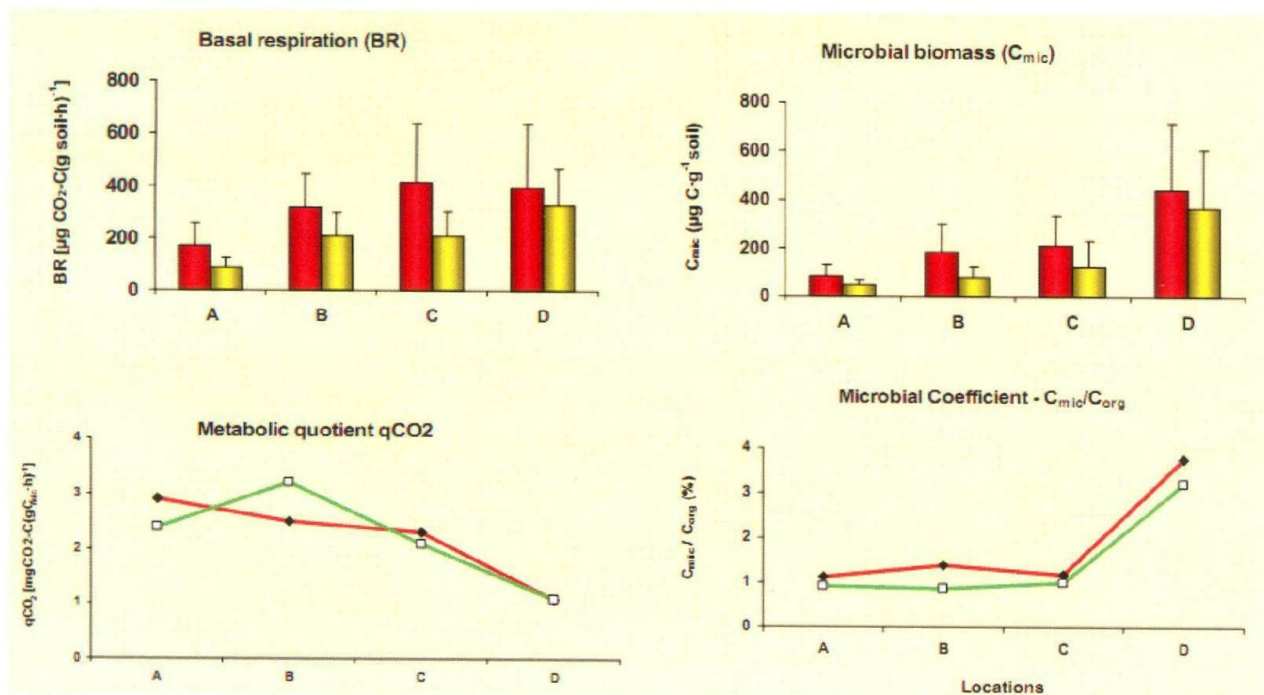


Figure 8. Basal respiration, microbial biomass, metabolic quotient and microbial coefficients values obtained in soil samples contents along the deposition transect at 0 to 10 (■) and 10 to 20 cm (■) soil layers in Almalyk mining industrial area.

Discussion of the research in the context of similar or earlier studies

The locations of the study sites in relation to the main wind direction at Almalyk and Angren industrial areas determined the degree of heavy metal accumulation in the soil to a large extent, as did the distance to the emission source. Change in total heavy metal concentration in the two soil layers along a downwind transect of 20 km was found to decrease the total concentration levels of Cu, Zn and Pb by 3 to 20 times. The complementary use of a sequential extraction and JEOL microprobe observations demonstrates that in the contaminated soils, most of the Zn, Pb, Cu, Cd concentration associated with inorganic and organic forms from which they can be relatively easily removed. Ni, Co and Cr mainly associated with inorganic residual forms presumably of a sulphide and oxide nature. Therefore, there is a much greater risk of Zn, Pb, Cu, Cd mobilization from these soils than there for Ni, Co and Cr. The first two steps of the sequential extraction, which represent the more mobile metal forms, removed an appreciably larger average proportion of Cd (44%), Pb (24%), Zn (19%), and Cu (17%) than that of Cr (2%) and Ni (8%). The third and fourth fractions, which are under oxidizing conditions may be released into solution, contains twice the average amount of Cu (68%), Cd (44%), Pb (64%) and Zn (70%) than that of Ni (31%). In the residual, i.e. chemically inert form, the total average proportion of Ni (56%) and Cr (76%) is more than twice that of other metals (Cd (12%), Pb (10%), Zn (13%), and Cu (19%)) (Annex 1, slide 9-10) The distribution of heavy metals in this study was found to be strongly correlated to heavy metal physico-chemical properties, as reported in other studies (Alloway, 1990; Talipov et al., 1997). This distribution yielded two different distribution patterns: (1) a gradual, continuous decrease from the pollution source along the downwind transect; and (2) a relatively low level of contamination at the source with a gradual increase moving away from the source (in some cases a gradual decrease occurs at a certain distance from the source). Both distribution patterns are defined by both the abiotic components and the physico-chemical properties of the pollutant. Because of the alkaline nature of our study site, pH and soil organic carbon values were found to exhibit a different pattern than those reported for the Camacary region of Brasilia (Klumpp et al., 2003). A high level of carbonate (CO_3^{2-}) content in the studied soils might reduce heavy metal mobility in soils by increasing chemisorptions (Pinsky, 1997), causing a significant difference in heavy metal contamination between soil layers, as obtained in our study. However, only Cd exhibited a different pattern, perhaps due to its different chemo-physical properties. With the exception of Cd in the first location, all the heavy metals studied, As, Cu, Pb, Zn, accumulated in the upper (0-10 cm) soil layer at values twice as high as in the deeper (10-20 cm) soil layer. These values were found to decrease to values more than 10 times lower along the 20 km transect, similar to results reported by Klumpp et al. (2003) for Cu. Earlier studies by Hutchinson and Whitby (1974); Kandeler et al. (1999) support our findings that Cu, Pb, Zn and Cd contamination accumulates mainly in the top soil layer and that a rapid and gradual decrease occurs with increasing distance from the smelter. The decrease in heavy metal concentration along the transects was found to be an opposite trend to the changes in soil moisture availability, organic carbon and total soluble nitrogen, which may act as a source and sink of nutrients in the soil, triggering the soil biota community. In our study, the studied biotic components and their indices showed a clear relationship with the pollutant concentration in soil. Such detrimental effects of heavy metals and other soil contaminants have repeatedly been reported in the literature (Hattori, 1992; Bardgett et al., 1994; Kandeler et al., 1996; Wilcke et al., 1999; Klumpp et al., 2003). Because of its biological function as a well-known bioindicator, the soil free-living nematode population

(Wardle et al., 1995; Yeates and Williams, 2001) exhibited a significant positive response to a decrease in soil heavy metal contamination.

Our results, which used food consumption characteristics in order to understand the soil free-living nematode function in the system as a response to heavy metal pollution, indicate that plant parasites, followed by fungi-feeding nematodes, were the most dominant trophic groups at the pollution source, while, with distance, the dominance was replaced by bacteria-feeding and omnivore-predators nematodes. The contribution of BF, FF, PP, and OP trophic groups to the trophic composition of the nematode population near the pollution source accounted for 4, 53, 42, 1 and 17, 5, 72, 6 percent in the upper and deeper soil layers, respectively. The contribution of the observed trophic groups at the end of the study area accounted for 23, 6, 9, 62 and 32, 3, 40, 26 percent in the upper and deeper soil layers, respectively. However, the OP, BF, FF, and PP were found to correlate with the changes in heavy metal content in the soil along the emission transect. Our data are in agreement with other studies (Parmelee et al., 1993; Korthals et al., 1996), that showed that the addition of Cu, Ni, and Zn up to 1600 mg kg⁻¹ significantly affected many parameters of the nematode community structure, such as the populations of certain omnivorous and predatory nematodes with a K-strategist type of life history (Bongers and Bongers, 1998; Bakonyi et al., 2003). Moreover, the populations of several nematode taxa were significantly affected by the concentration of Cu, Ni, and Zn (Korthals et al., 1996). In our study, the Maturity and modified maturity indices were found to be a useful tool for assessing metallurgical-industry pollution in soil systems along an air pollution emission gradient. The use of trophic diversity was found to show a mild response to changes along the pollution transect while the Shannon, Richness and Evenness indices were not found to respond numerically to any changes in the study sites.

The females of the total soil free-living nematodes were found to be the most resistant to heavy metal pollution levels and were present along the sites, while the juveniles were found to be sensitive to changes in heavy metals. The low number of juvenile nematodes in the present research could be explained using the view of researchers who believe that juvenile nematodes are more sensitive to pollution than are the adults (Kammenga et al., 1996; Camargo et al., 1998), and that the sex proportion changes under the effects of pollution (Anderson et al., 2001); or using the view of researchers (Vranken and Heip, 1986) who believe that nematode reproduction is more sensitive to heavy metal pollution than survival and development. The present studies elucidates the possibility as well as the importance of using nematode density, biomass, activity, and community structure as indicators of ecosystem health. The biomass of soil organisms, together with their number dynamics, has been found to be a useful indicator of environmental pollution by Ingham et al. (Ingham et al., 1986a, 1986b) Paul and Clark (1989), Nannipieri et al. (1990), and Yeates et al. (2003), in their studies of different systems. Moreover, in monitoring soil organism dynamics, we can detect detrimental ecosystem changes and possibly prevent further degradation (Lal and Stewart, 1992).

In this study microbial biomass as well as CO₂ evolution patterns were found to increase gradually in response to the decrease in heavy metal concentration at the two soil layers, yielding a clear inverse significant relationship with the pollutant concentration in the soil. These results are supported by similar studies (Kandeler et al., 1996; Kandeler et al., 1999). Moreover, the ecophysiological quotient qCO₂, which represents a specific physiological status evaluating the environmental effect on the soil microbial community, revealed a small difference between the two soil layers, with a significant change toward the end of the transect. Such differences were mainly affected by heavy metal distribution. Results from our study on microbial coefficients were found to be within the range

of values reported by Hofman et al. (2003), working on a monitoring microbial biomass and respiration program aimed to determine the relationship between soil properties and contamination. The microbial coefficient was found to be an important aid for gaining more complete understanding of the heavy metal, abiotic, soil chemical composition and soil microbial interaction. Since the microbial coefficient interpreted as substrate availability exhibited a significant increase at station D, which is complementary to the significant decrease in ecophysiological status, it seems to elucidate the sensitivity of the microbial community as one of the most sensitive indicators of soil contamination.

All obtained data within these studies were presented in international scientific conferences and published in international peer-reviewed journals (Shukurov et al. (2005); Shukurov et al. (2009); Pen-Mouratov et al.(2008); Kodirov, Shukurov (2009); Pen-Mouratov et al.(2009); Bandowe et al. (2010)).

Conclusions

Studies on the relationship between soil biota and pollution levels have raised the question regarding the importance of natural soil abiotic properties, stressing the importance of background data of environmental conditions, and elucidating the importance of further studies on this subject. This preliminary monitoring studies applied a wide range of geochemical and microbiological parameters to characterize the air pollution impact on the soil health in the Angren-Almalyk industrial area. Variability of individual parameters differed widely and significantly among sampling sites and transect directions. Although detrimental effects and cross-correlation by persistent organic pollutants (POP's) cannot be ruled out on the basis of this study, similar effects of heavy metals on the soil microbiological parameters have repeatedly been reported in the literature as discussed in the introduction. Because of its well-known function as a bioindicator, the soil free-living nematode population exhibited a most sensitive indicator of unpolluted soil, and hence appear to be useful for an evaluation of any future efforts for the recovery of the polluted soil sites. The metabolic quotient qCO_2 revealed a small difference between the two soil layers, but a significant effect in the most air polluted sampling sites. Since the microbial coefficient, C_{mic}/C_{org} , exhibited a significant increase at stations E and H in an inverse manner to the significant decrease in the ecophysiological status, both parameters seem to elucidate complementarily the sensitivity of the microbial community to soil contamination. We conclude that both coefficients (and even a ratio thereof) are the most useful bioindicator of soil pollution in that region. Based on the results of the present study and on previous investigations in the same and similar regions, an extensive environmental monitoring of the industrial regions of Angren and Almalyk is proposed. Such a program should include not only the determination of the soil inorganic and POP inventory, but also the aforementioned (minimum) three biological soil health parameters.

There was a significant spatial dependence and differences for all soil chemical and microbiological parameters tested. The highest contents were found for the relatively volatile metals Zn (≤ 1136 mg/kg) and Pb (≤ 373 mg/kg) in upper soil layers near the power station suggesting that the metal pollutants are derived from local stack emissions (Angren industrial area). Soil microflora was obviously affected by heavy metals. Significant positive correlations ($p \leq 0.001$) were found between the metal content, RB, and qCO_2 , while a negative one was found for the mineralizable N and C_{mic}/C_{org} ratio. A high total number of nematodes were found only most distant from the industrial emission sources. The results disclosed remarkable spatial dependence not only of the heavy metal impact onto the soil but also of microbiological soil properties in the study area. The lat-

est data suggests bioavailability of the anthropogenic metals in the soil affecting the soil microbial community. This is suggested by less biomass formation and higher qCO_2 values in heavy metal-contaminated compared to less-polluted soil plots. Knowledge of these spatial ecosystem functioning patterns and dependence could be very useful in determining and delineating specific land use and management programs that would be suited and feasible for the highly polluted area. Results of this study can be utilized to develop a monitoring program that may quantify harmful effects on the soil health and impact of any future remediation activities. Studies on the relationship between soil biota and pollution levels have raised the question regarding the status of natural soil microbial health, stressing the importance of background data of environmental conditions, and elucidating the importance of this environmental monitoring approach even in semiarid and arid regions. Soil microbiological parameters, in particular the metabolic quotient qCO_2 as one of the most sensitive bioindicators identified for that region, should clearly become part of the national environmental monitoring program.

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Коллектив

Науки о Земле в Узбекистане

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