UNIVERSITY ”ALEXANDRU IOAN CUZA” OF IAȘI

FACULTY OF GEOGRAPHY AND GEOLOGY

DOCTORAL SCHOOL OF CHEMISTRY AND EARTH SCIENCES

The Geochemistry of soils evolved on Neogene volcanites from Eastern Carpathians

ABSTRACT

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Contents

Introduction

There is now a rich and impressive literature on geochemical abundance of chemical elements in soils on Earth. Among the many contributions of first order pedogeochimic mention valuable monograph published by Adriano D. C., Alina Kabata Pendias and Henryk Pendias. This is also supported by the Geochemical Atlas of Europe, European countries (Slovakia, Croatia, Romania, Moldova, etc.) and several European cities, including cities within Romania (Bucharest, Iasi, Baia Mare, Ploiesti).

In our country, the geochemical abundance of chemical elements in soil and especially trace elements was initiated and subsequently addressed systematically by specialists from the Agrochemical Research Institute (ICPA) in Bucharest as well as those of Biogeochemical Department in Iaşi, Branch of the Romanian Academy. First, researches approached quantitative distribution of trace elements in different soils in plains, mountains and the Danube Delta. Later, researches had grown and diversified given the need for more detailed knowledge of the quantitative distribution of chemical elements in each of the components of the environment.

The main objective of the thesis is to approach complex geochemical soil evolved on Neogene volcanites of the Eastern Carpathians, on the basis of macro quantitative distribution analysis characterizing classical silicate (Si, Ti, Al, Fe, Ca, Mg, Na, K, P, S), and some trace elements which exhibit affinity for the alkaline soda lime silicate composition of volcanoes parental heavy metals: Cu, Pb, Zn, Ni, Co, Mn, Cr, Cd, Mo, rare earth metals: Rb, Sc, Nb , La, Ce, Sm, Nd and metalloids: As. Worth mentioning are the first analytical data on pedogeochemical abundance of rare metals (Rb, Nb, Sc, TR) in volcanic soils in the Eastern Carpathians.

I. The geographical characteristics of the region

Positioned in the Western Carpathians, the Neogene chain Oaş-Hargita is considered to be the longest of its type in Europe.

Neogene volcanic arch of the Carpathians Geotectonics falls in the overall development of the region during the Tertiary Carpatho-Pannonian.

II. The geotectonic framework of the Neogene volcanic arc of the Eastern Carpathians

On the basis of petrographic, geochemical and structural analysis, the Neogene volcanic arc of the Carpathians was divided into three sectors (Fig.1):

- Oaș-Gutâi = volcanic;
- Țibleș-Rodna-Bârgău = subvolcanic;
- Călimani-Gurghiu- Harghita = volcanic.

The Neogene volcanic arch of the Carpathians was always an object of interest for mineralogical, petrographic, geochemical and metallogenic research, given the presence in the mass of hydrothermal polymetallic mineralization of gold and silver with great economic value.

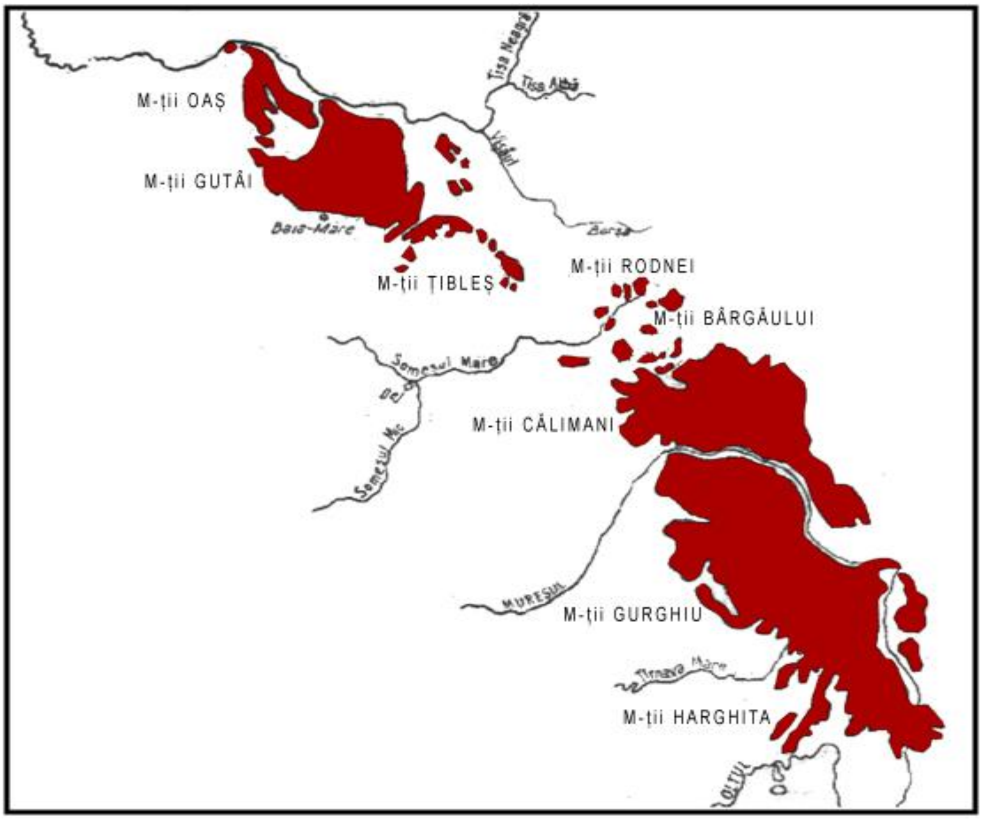

Fig.1. The Neogene volcanic arch of the Carpathians

(Rădulescu and Dimitrescu, 1982)

III. Petrography and mineralogy of the Neogene volcanic arc magmatic rocks in the Eastern Carpathians

Structural characteristics, mineralogy and petrography of the Neogene magmatic reservoir and age of the Eastern Carpathians are shown graphically in Table 1 (in thesis).

IV. Geochemistry of the Neogene magmatic Eastern Carpathians

The soils studied belong to the northern (Fig.2) and southern (Fig.3) sectors and have evolved on rhyolite volcanic rocks belonging to the series dacite-andesite-basalt-(TAS diagram) having a calc-alkaline character (AFM diagram) (Fig. 4). The diagram K2O: SiO2 (Fig.5) lies mostly in the "medium K".

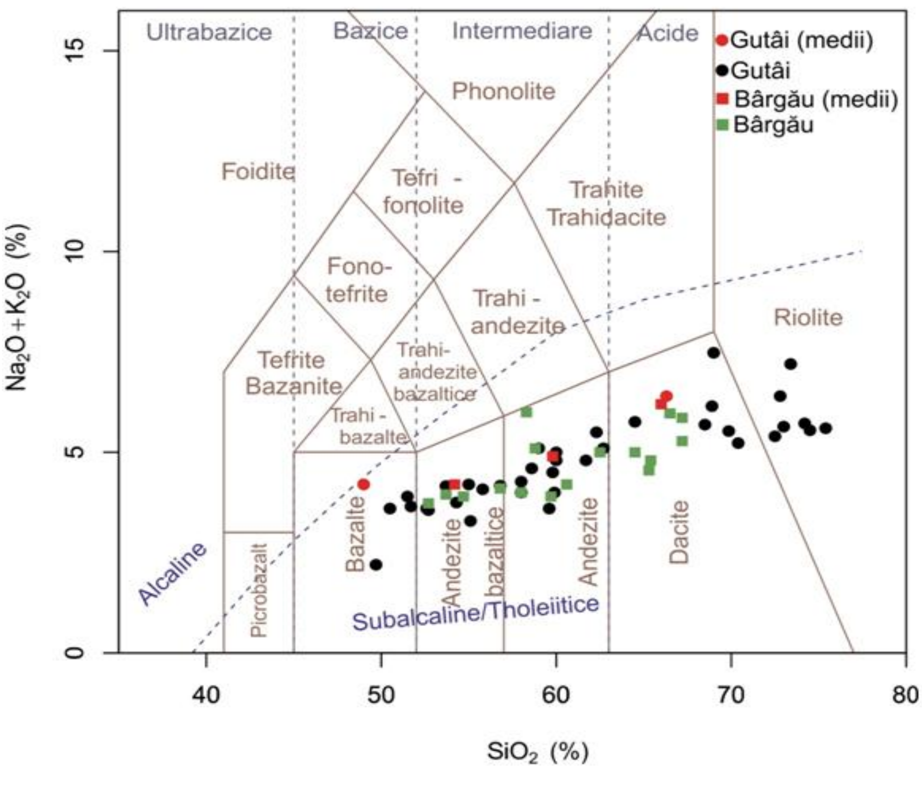

Fig.2. TAS diagram (Lebas et al., 1986) for Gutâi and Bârgău volcanites (analytical data from Kovacs, 2002 and Delia Cristina Papp et al, 2005)

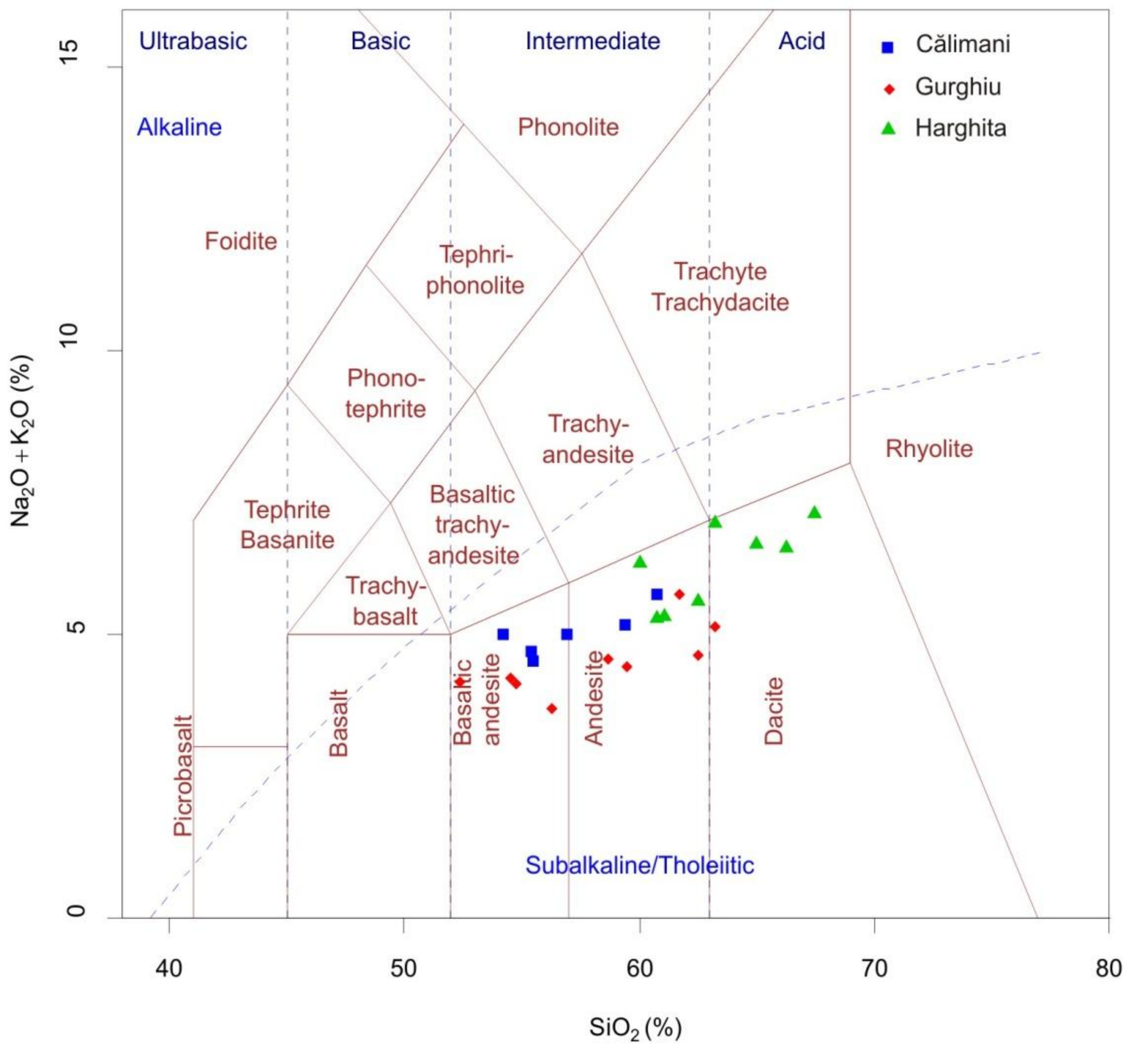

Fig.3. TAS diagram (Lebas et al., 1986) for magmatic Călimani-Gurghiu-Harghita sector (analytical data of Mason et al., 1995)

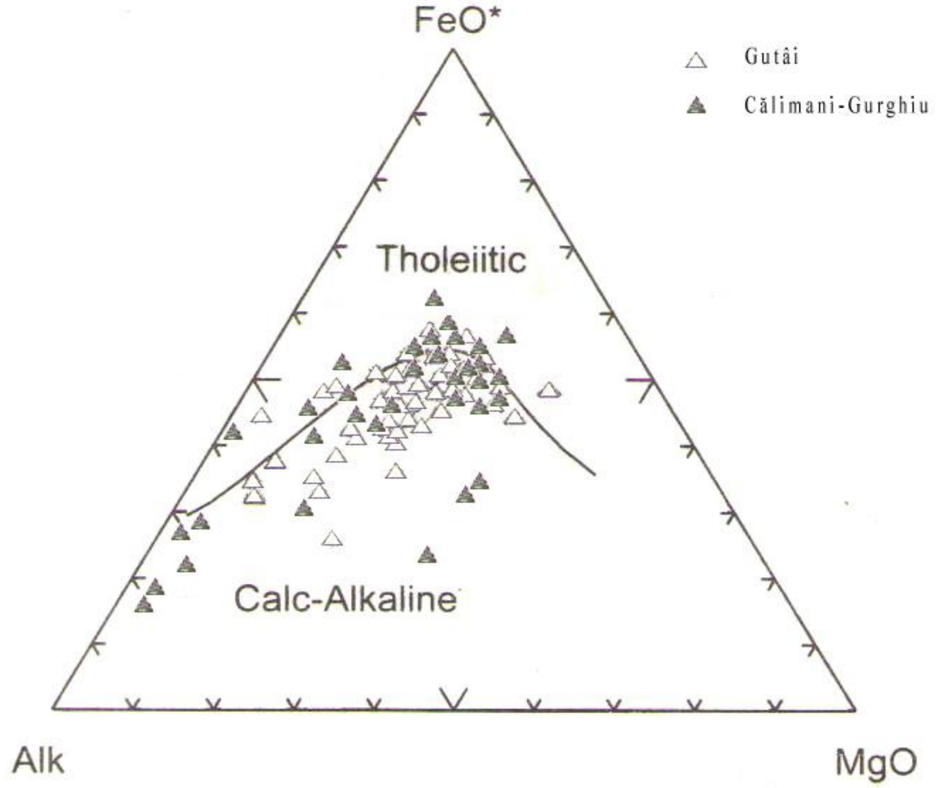

Fig.4. Magmatic distribution of Gutai, Gurghiu and Călimani Mountains in AFM diagram

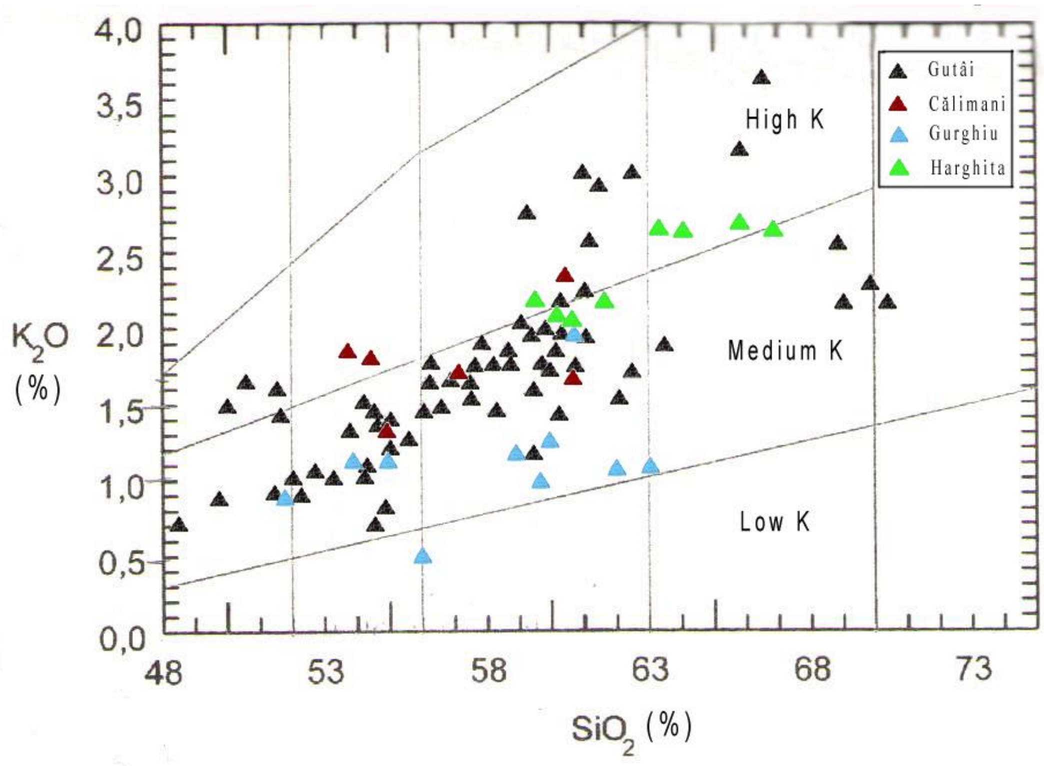

Fig.5. Magmatic distribution of Gutai, Calimani, Gurghiu and Harghita Mountains in K2O: SiO2 diagram

-In all three sectors, the andesites prevalent.

- Volcanic rock minerals consist of primary (main plagioclase, pyroxene, amphibole, biotite, quartz and accessories: apatite, garnet, tourmaline, ilmenite, magnetite) and secondary minerals (epidote, chlorite, calcite, limonite, kaolinite, illite , montmorillonite). Noteworthy is the presence of garnet in magmatic origin andesite (Table ...).
- The age of Neogene magmatic volcanic arc in the Eastern Carpathians is between 13 M.y. (the Oas-Gutai) and 0.15 M.y. (sector-Gurghiu- Călimani Harghita).

V. Method and the database

The thesis is based on a complex study of 17 soil profiles including 98 samples from Oaş, Gutai, Gurghiu, Harghita and Giurgeu Mountains. Physico-chemical and geochemical analyzes were performed for the following types of soils formed on volcanites Neogene of the Eastern Carpathians: eutricambosoil, districambosoil, andosoil, prepodzol and stagnosoil in the mountains above mentioned areas.

VI. Typology and physico-chemical properties of soils evolved on the Neogene volcanites of the Eastern Carpathians

Typology: The Neogene volcanic arc paedeological Fund of the Eastern Carpathians is constituted in accordance with Romanian System of Soil Taxonomy (SRTS-2012, Florea Munteanu) of the following four types of soil: Cambisoils (CAM); Spodisoils (SPO); Andisoils (AN);

Hydrisoils (HID).

Cambisoils are the dominant and include two subtypes:

- Eutricambosoils (EC);
- Districambosoils (DC) and hydrisoils lowest deals surfaces being represented by stagnosoils (SG).

Physico-chemical properties of soils formed on the Neogene volcanites of the Eastern Carpathians were established and interpreted according to the following determinations: pH (H_2O), organic C, humus, the amount of exchange bases (SB), exchange acidity (SH), the degree of base saturation (V), cation exchange capacity (T). The results show the following key features:

- Frequency of strong acid reaction, with an increasing trend from surface to depth;
- Humus content falls within large limits, with the highest values in stagnosoils and a decrease in concentrations below the upper horizons;
- Exchangeable bases (SB) have higher values in soils rich in humus and soil low in strong leachates;
- The degree of base saturation (V) are studied soils: oligobasic, eubasic and mesobasic;
- High humus content determines the presence of a cation exchange capacity (T) quite large.

Soil Texture: size composition of soils formed on volcanites convincing highlights of the Eastern Carpathians Neogene rocks parental footprint and intensity of alteration classic manifestation biogeochemical processes in which coarse textures (G), textures predominate medium (M) and fine (F) (Fig .6).

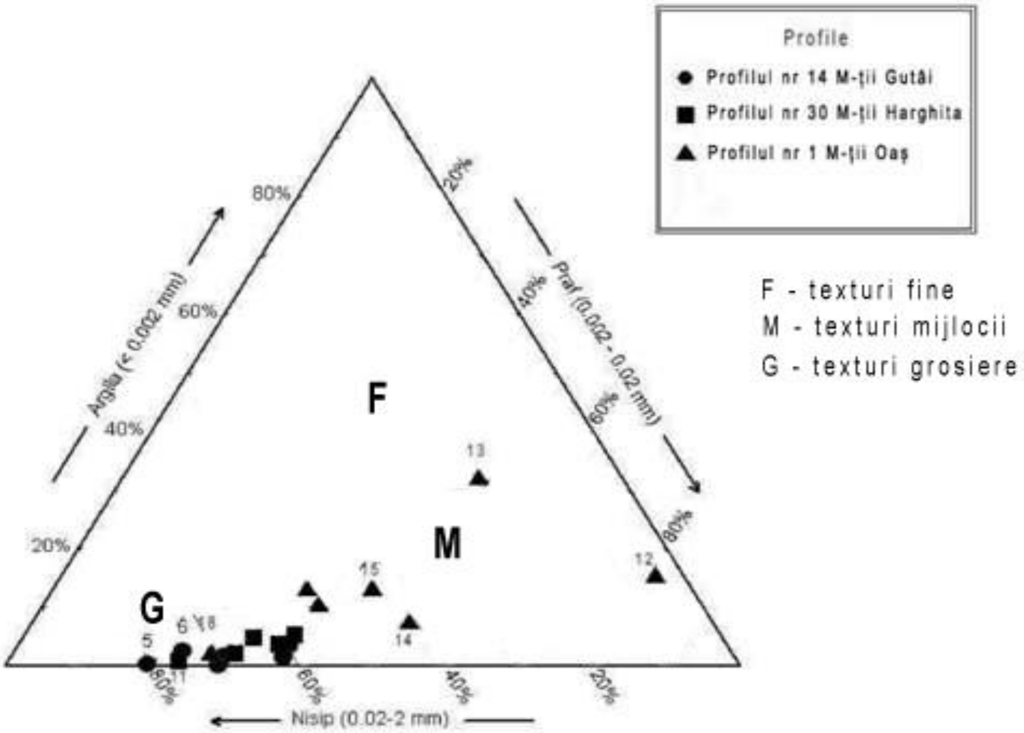

Fig.6. Ternary diagram of evolved soil texture on Neogene volcanites of the Eastern Carpathians

Organic matter:

Components analysis of organic matter or humus fractionation was performed for all types of soils formed on volcanites Neogene of the Eastern Carpathians in the areas of Oaş, Gurghiu, Harghita and Giurgeu. The research focused on identifying quantitative and qualitative differences in the organic component of soils studied.

Analyses on the amount of soil organic matter and humus reveals the main components of the quantitative distribution of the various fractions of humic profile closely with complex pedogenetic factors (parent material, coating plant, biological activity, hydrothermal conditions, physico-mechanical and chemical).

In terms of qualitative analysis of humus fractionation indicates a variable content of humic acids (humic acids and fulvic acids). This composition is the result intake nitrogen rich vegetation cover which favors the formation of humic acids. Rich in pine litter remains poor in nitrogen favors the production of fulvic acids, compounds with low polymerization degree on easy livigabili, which are accumulated in some horizons.

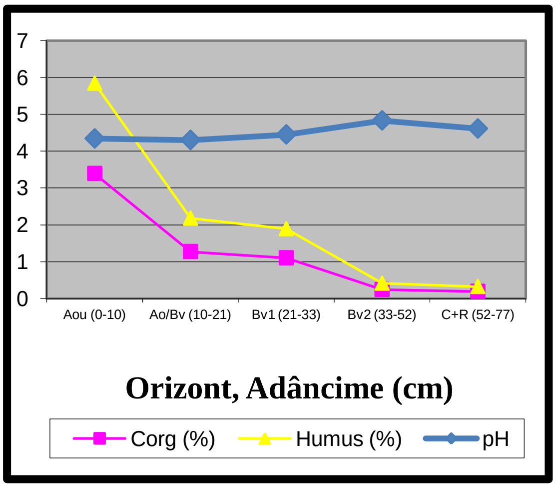

Fig.7. Humic fractions from Andic Eutricambosoil andic (400 m altitude)

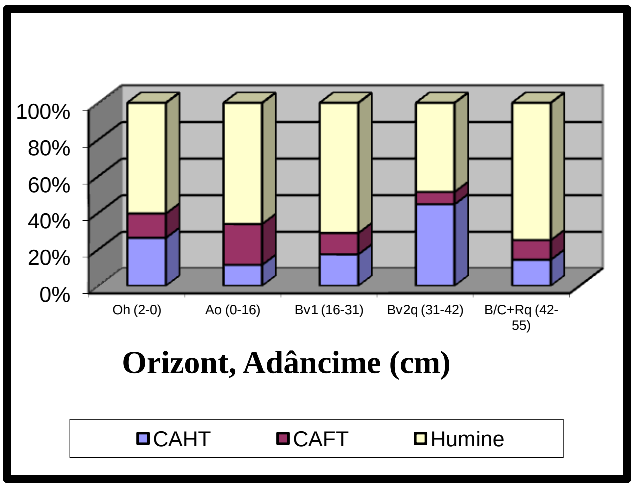

Fig.8. pH, Corg(%), humus(%) distribution from Andic Eutricambosoil (400 m altitude)

Andic Eutricambosoil (400 m altitude) was taken from the Berii valley (Tarna Mare) under vegetation cover, having 77 cm thickness profile. The chemical reaction of the soil is acid throughout the profile (4.30 to 4.83) but slightly lower in the horizon BV2. Organic accumulation is reduced and placed on a curve which shows a decrease with depth; C org. has values of 3.39% in the AOU, 1.1% by BV1 and decreased to 0.19% C + R. The degree of humification can be represented on a sinuous curve with values of 36.87 in the AOU and 75 in BV2. Humifiable material is composed mainly of fulvic acids; the profile and BV1 which accumulate in excess of 4.7 times the humic acid. Humic acids tend to accumulate towards the base profile where they form stable bonds with the mineral.

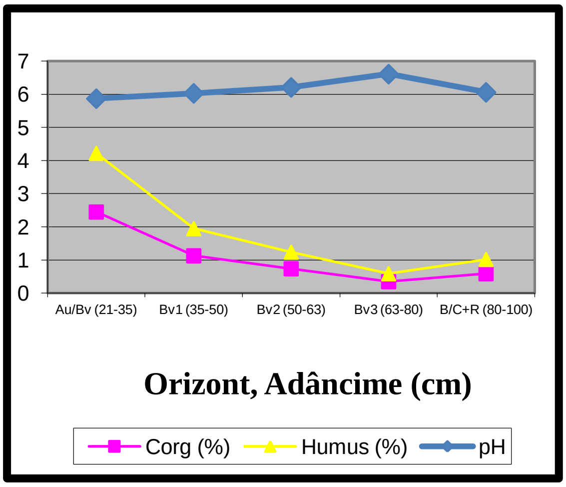

Fig.9. Humic fractions from cambic Andosoil (1197 m altitude)

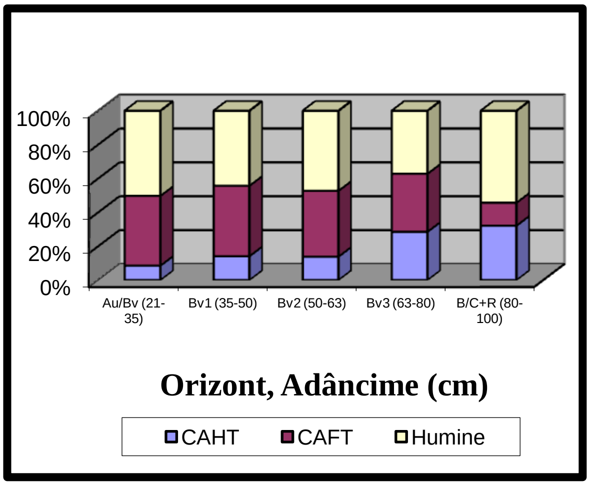

Fig.10. pH, Corg(%), humus(%) distribution from cambic Andosoil (1197 m altitude)

The Cambic Andosoil (1197 m altitude) taken from atop an andesitic plateau. The thickness profile is 110 cm.

The chemical reaction is slightly acid soil throughout the profile (pH = 5.87 to 6.61). Accumulation moderate organic humus in the upper horizons and arranged on a curve which shows a decrease with depth (2.45 to 0.59% C org.total); data show a good mineralization. The degree of humification may represent slightly sinuous curve with values between 49.79 in Au / BV and 62.85 in BV3. Humifiable material consists mainly of fulvic acids exceeding (by 4.8 to 1.2 times) throughout the humic profile (except the last horizon).

VII. Soil Geochemistry of the Eastern Carpathian Neogene evolved volcanites

The main objective of the thesis and deals abundance in volcanic soils of the Eastern Carpathians macro characterizing the classical analysis of silicates (Si, Ti, Al, Fe, Ca, Mg, Na, K, P, S) and trace elements characteristic of volcanoes parental calc-alkaline basalt-rhyolite series of heavy metals (Cu, Pb, Zn, Cd, Mn, Co, Ni, Cr, Mo); rare metals (Rb, Sc, Nb, La, Ce, Nd, Sm); metalloids (As). Of these trace elements, analytical data for Rb, Nb, Sc, La, Ce, Sm, Nd is the first time.

For each chemical element geochemical analysis is the following considerations:
- Serial number Z in the periodic table;
- Electronic configuration;
- Oxidation state (valence);
- Number and abundance of natural isotopes;
- Crustal abundance;
- Geochemical behavior in magmatic and sedimentary processes mineralogenetice;
- Their main minerals of the element and mineral concentrators;
- The average content in the soils of the world by different authors;
- Overall abundance coefficient pedogeochemical values (CGAPdg) and the coefficient of pedogeochemical concentration (CCPdg);
- The order of accumulation in the four soil types according to the average content;
- Values geochemical correlation coefficients (positive, negative) and graphical binary correlation diagrams of the major:minor elements.
- Statistical processing of macro and microelements analyzed concentrations.

VII.1.Major elements geochemistry

- Abundance of major elements in volcanic soils of the Eastern Carpathians content highlights some variations depending on the chemical composition of parental volcanoes, soil type and organic matter content and clay minerals;
- In general, the data content of major elements in soils studied are comparable to those present in calc-alkaline volcanites parenting;
- The analytical results show that acidic soils formed on volcanites contain more Si, K, Na and less Fe, Mg, Ca than soils evolved intermediate and basic rocks;
- Pedogeochemical data obtained by us for the contents of the macro volcanic soils in the Eastern Carpathians are fully consistent with those determined by domain experts for similar volcanic soils in some regions of the world (Italy, Portugal, Slovakia, Colombia, etc.);
- Correlations between macro are characteristic geochemical soil type and composition correspond to calc-alkaline volcanoes parenting;

Geochemical positive correlations:
- Andosoil P-S correlation ($r = 0.9$) in districambosoils ($r = 0.86$), spodisoils ($r = 0.84$) and andosoils ($r = 0.9$):

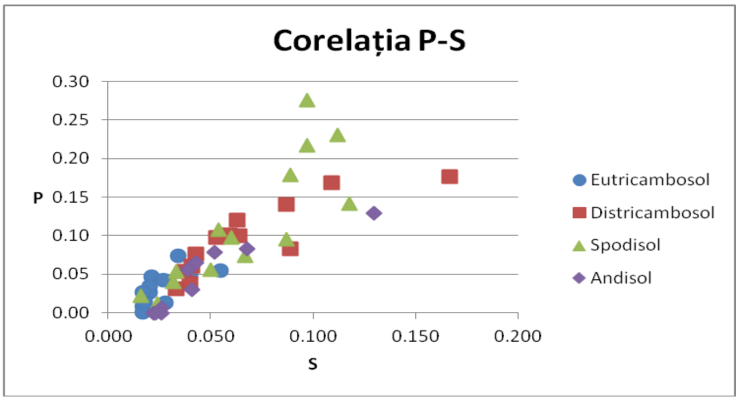

Fig.11. P-S Correlation

- Ti-Fe correlation in districambosoils (r = 0.957), andosoils (r = 0.954), spodosoils (r = 0.932) and eutricambosoils (r = 0.863):

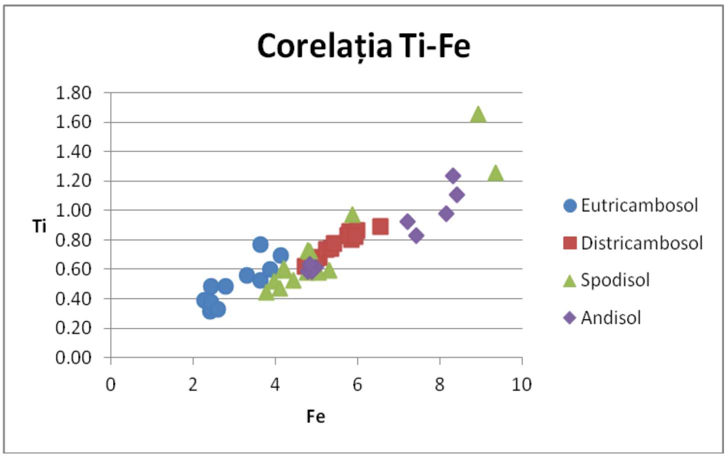

Fig.12. Ti-Fe correlation

Negative correlations geochemical most important are:

- Si-Al correlation in andosoil (r = -0.939) in eutricambosoils (r = -0.826) and spodisoils (r = -0.72):

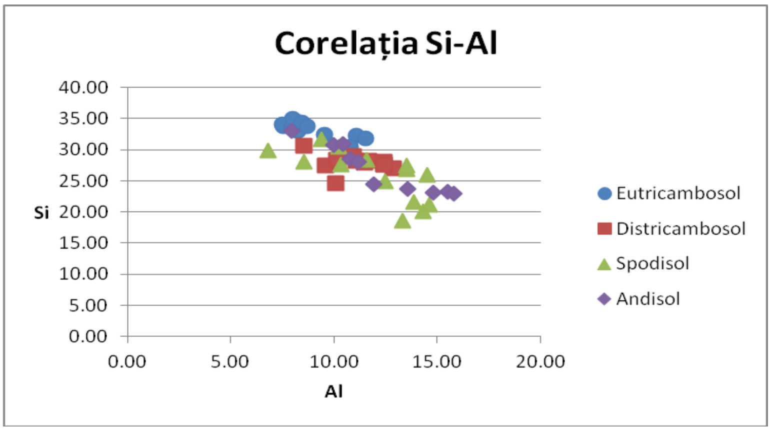

Fig.13. Si-Al correlation

Among the highlights of macro geochemical correlations with trace elements analyzed mention:

a) positive geochemical correlations:

- Mg-Cr correlation in districambosoils (r = 0.762):

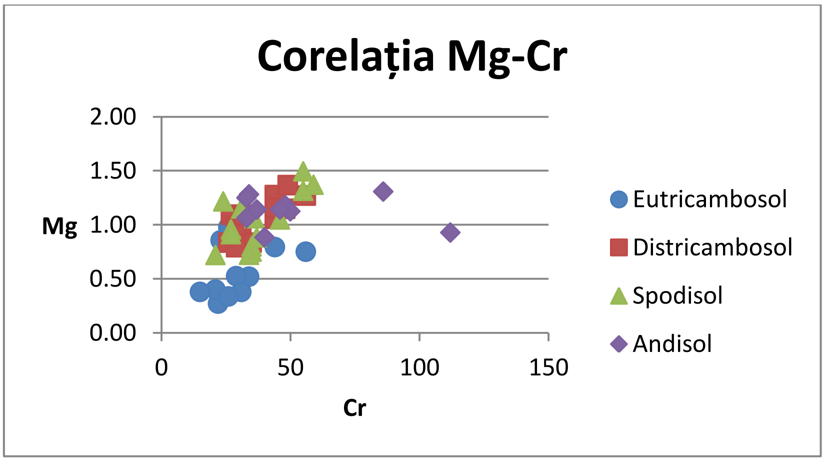

Fig.14. Mg-Cr correlation

b) negative geochemical correlations:

- Sc-Rb correlation in districambosoils(r = -0.910), andosoils (r = -0.963) and spodisoils (r = -0.859):

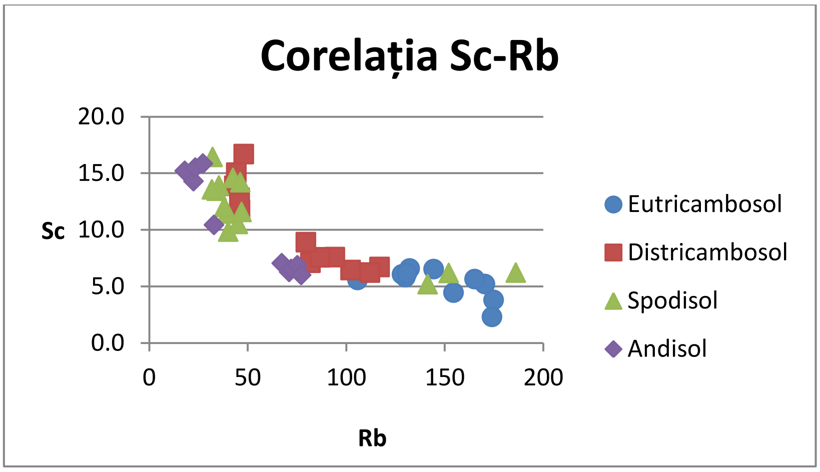

Fig.15. Sc-Rb correlation

- • Abundance profile of macro elements shown in the following diagrams is subject to soil type, size variation, clay mineralogy, the amount of humus, etc. • In general, the soils studied, the macro elements abundance profile is quite uniform. • There is still some concentration of Si, Ca, Na and Al in the lower horizons in intermediate horizons rich in clay.

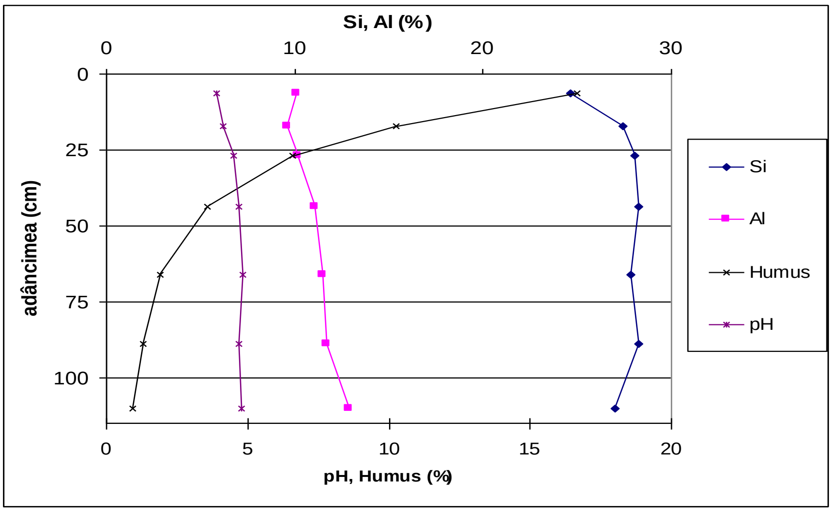

Fig.16. Distribution profile of Si and Al in districambosoil (DC) of Gutâi

(Profile no. 16 Săpânța)

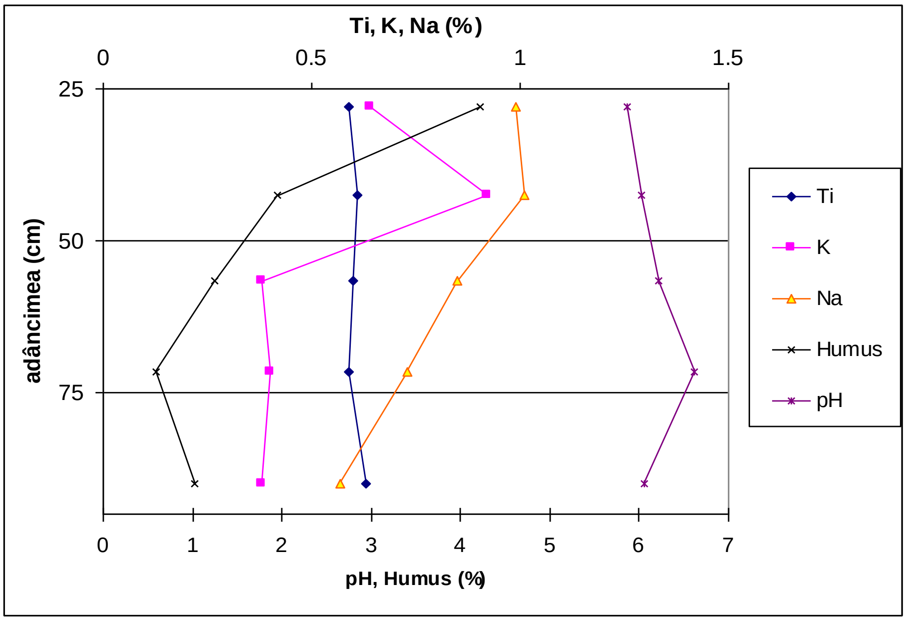

Fig.17. Distribution profile of Ti, K, Na in andosoil (AN) Giurgeu Mountains (Nr.46 Profile Sărmaş)

VII.2. Geochemistry of trace elements:

Pedogeochemical distribution of trace elements in the four soils was strongly influenced by parental volcanoes composition of organic matter and processes that have shaped pedogeochemical soils.

An important role is the abundance of trace elements and expression of biogeochemical cycles: absorption by plant roots organizations → → release assimilation by them in the ground.

The data contained in the soils studied trace elements are fully consistent with those obtained for volcanic soils in other countries: Italy, Portugal, Slovakia, Colombia.

The range of concentrations of trace elements of volcanic soils in the Eastern Carpathians is within the limits set by various researchers for Earth soils.

The abundance of trace elements in volcanic soils in the Eastern Carpathians compared to overall abundance in the Earth's crust and soils of the world is reflected in the overall pedogeochemical abundance coefficient values (CGAPdg) and the coefficient of pedogeochemical concentration (CCPdg).

The analytical data obtained confirm that the new soil rocks evolved acid (dacite) concentrated than trace amounts "granitofphile" (Rb, Nb, Mo, TR) and the land formed on the intermediate and base rock (andesite, basalt) are enriched microelements "basitophile" (Co, Ni, Cr, Sc).

In most cases, the soil concentration is comparable to that of trace elements present in the parent volcanites.

Our results for the contents of soil trace elements analyzed were compared with normal values, alert and response threshold of sensitive soils (used in agriculture), or less sensitive (as unused agricultural land) covered by the Emergency Ordinance no. 756/1997 issued by the Ministry of Waters, Forests and Environmental Protection.

In general soil samples analyzed alert threshold for sensitive soils. Some content higher metal trace elements (Cu, Pb, Zn, Cd) in the soils analyzed in andesites are due to the presence of parental polymetallic mineralization.

Geochemical affinity of trace metal for organic matter is confirmed by their high content present in humus and some positive correlations of microelements with organic matter.

An important role in the abundance of trace elements (Mn, Cu, Pb, Zn, Ni) is the adsorption in studied soils.

Geochemical affinity of trace elemnts is exemplified using geochemical correlation coefficient values and graphic representations of binary correlations between micronutrients. The obtained analytical results confirm that some trace elements (Pb, Mn, Cu, Zn) has higher content in the upper layers rich in humus and (Mo) is preferred intermediate horizons.

The pedogeochemical behavior presents a remarkable abundance of light lanthanides (LREE) evidencing the presence of concentrations comparable to those of volcanoes paternal and the same order of concentration: What> La> Nd> Sm suggestive confirming their common origin and hence the chemical distribution.

The abundance of the trace elements in the profile is dependent on the type of soil, the amount of humus, clay mineralogy, pH values, their capacity for adsorbing the organic and inorganic soil constituents, and size composition.

In the following we present several significant correlations between trace elements analyzed:

Lead forming strong positive correlation with geochemical As in districambosoils (r = 0.869), spodisoils (r = 0.835) and andosoils (r = 0.957).

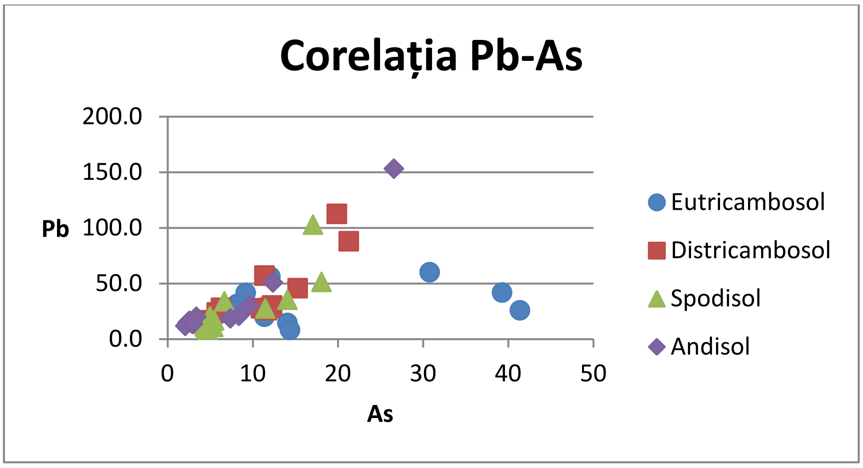

Fig.18. Pb-As correlation

The geochemical correlations between strong negative correlation with stands in districambosoils Sc (r = -0.956) and andosoils (r = -0.943).

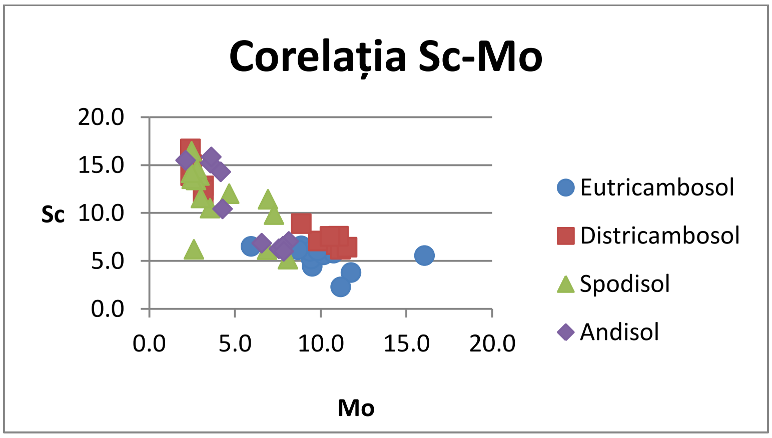

Fig.19. Sc-Mo correlation

Among the analyzed trace elements Zn, Pb, Cd, Cr generally in high concentrations in the horizon surface; Cu, Pb, Mn, Ni, Co tend to accumulate in the horizon C / R.

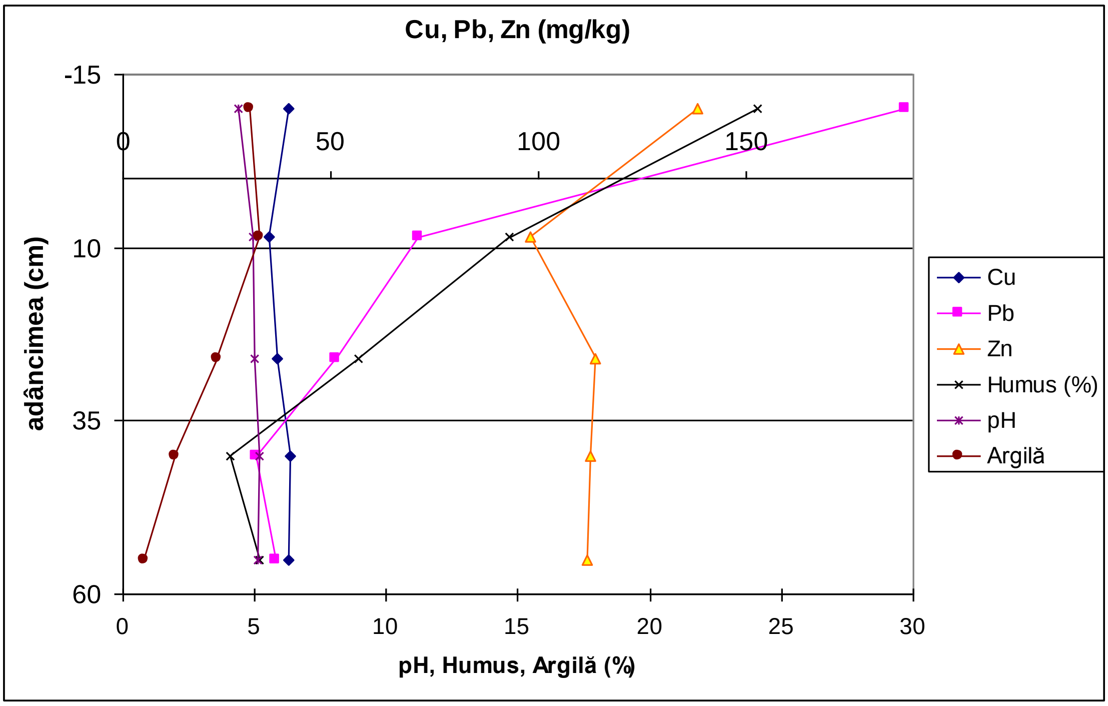

Fig.20. Profile Distribution of Cu, Pb, Zn in eutricambosoil (EC)
Oas Mountains (Profile No. 1 Tarna Mare)

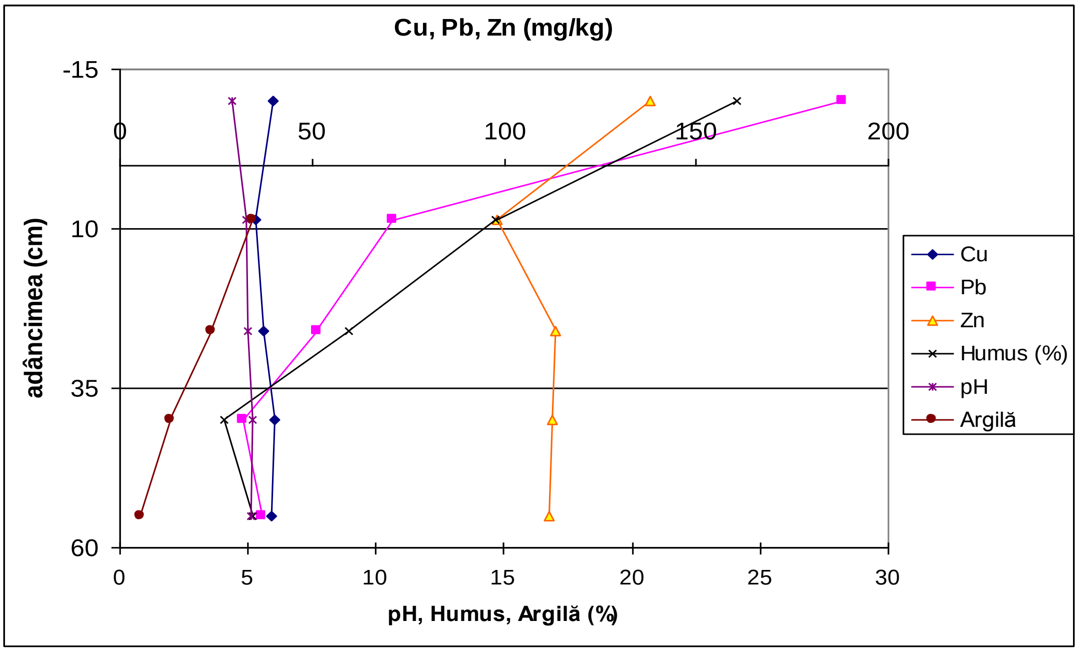

Fig.21. Profile Distribution of Cu, Pb, Zn in andosoil (AN)
from Gutai Mountains (Profile No. 14 Vama)

VII.3. Rare earth elements (REE)

In the literature are cited about 150 rare earth minerals and own over 250 minerals containing REE. Of these only 20 are important economic minerals.

International Association of Mineralogy (IMA) that the rare earth minerals name be accompanied by the chemical symbol of the element predominant. The silicate minerals belong to the TR (24%), carbonates (16%) and oxide (12%).

Among minerals concentrators REE mention those belonging to groups: oxides, carbonates, silicates, phosphates, etc .: sfen, epidote, spessartine, zircon, pyrochlore, Branner, perovskite, Colombia, strontianite, Anker, apatite, wolframite, Powel etc. Noteworthy is that the elements present name "rare" is an outdated term, given that the crustal abundance of lanthanides is higher than that of other metals: Pb, Sn, Au, Ag, Hg, Pt, Bi.

The relative mobility of the lanthanide in the pedogenesis is controlled adsorption capacity of cationic exchange of the soil and it varies in relation to the reaction. Typically, the mobility of lanthanides increases with atomic number, indicating that heavy-REE are more mobile than light-REE. Lanthanides adsorption capacity is higher in soils containing more organic substance and clay.

In soil, the fractionation of lanthanides is controlled largely by the presence of minerals and REE concentrators own and in a lesser extent the dynamics of organic matter, soil reaction and the amount of clay minerals.

Chemical analyzes of new soils studied, show concentrations of lanthanides light close to those present in calc-alkaline volcanites paternal (Table nr.IV.9.)

The strong positive geochemical correlations representative of other rare earth lanthanum are light in all four types of soil, especially with Ce and Nd. The geochemical positive correlations at the Ce (Fig.22.) have values ($r = 0.94$) in districambosoils ($r = 0.938$) in eutricambosoils ($r = 0.84$) in andosoils and ($r = 0.917$) in spodisoils.

The geochemical positive correlations with Nd (Fig.23.) are also strong ($r = 0.954$) in eutricambosoils ($r = 0.938$) in districambosoils ($r = 0.891$) in andosoils and ($r = 0.857$) in spodisoils.

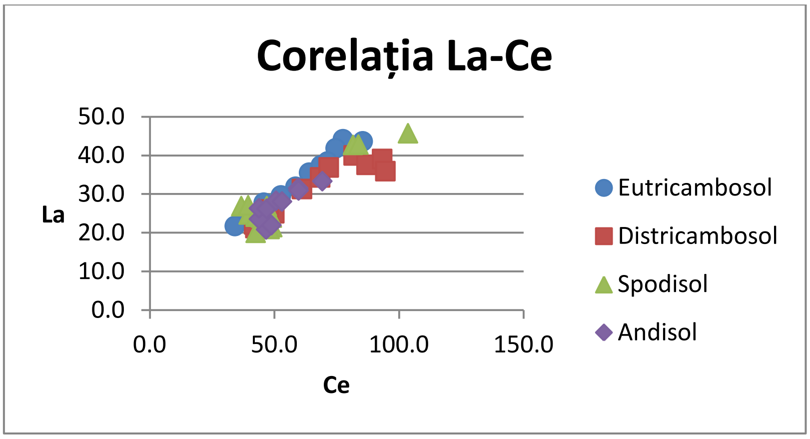

Fig.22. La-Ce correlation

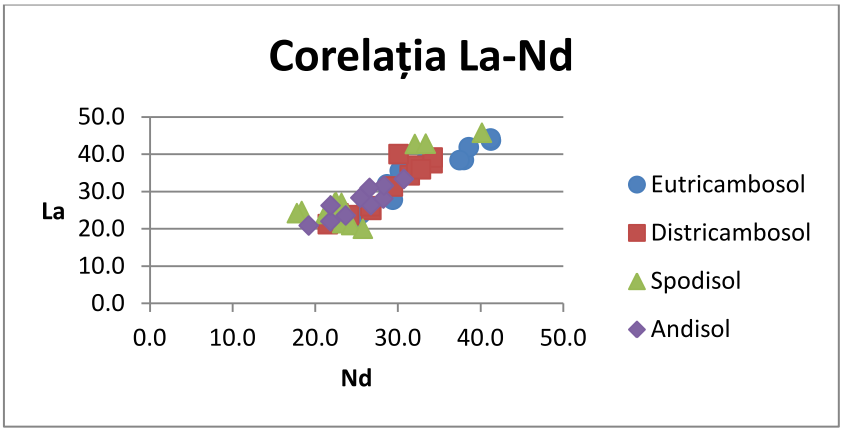

Fig.23. La-Nd correlation

Quantitative distribution of lanthanum on higher profile reveals content of the upper horizon and spodisoil eutricambosoil and lower horizon and andosoil districambosoil. It is noted lanthanum accumulation of eutricambosoils Bv horizon (from 43.6 to 44.2 mg / kg) of Oaş and Oh1 horizon of spodisoils (45.7 mg / kg) of Harghita Mountains.

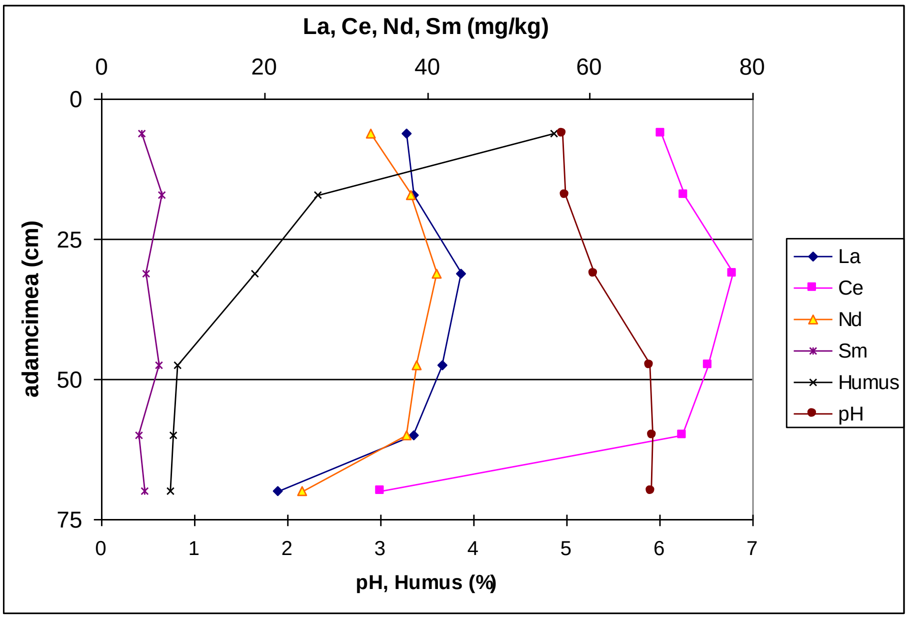

Fig.24. Distribution profile of La, Ce, Nd, Sm to eutricambosoil (EC) of Oaş (No.9 Profile Cămârzana)

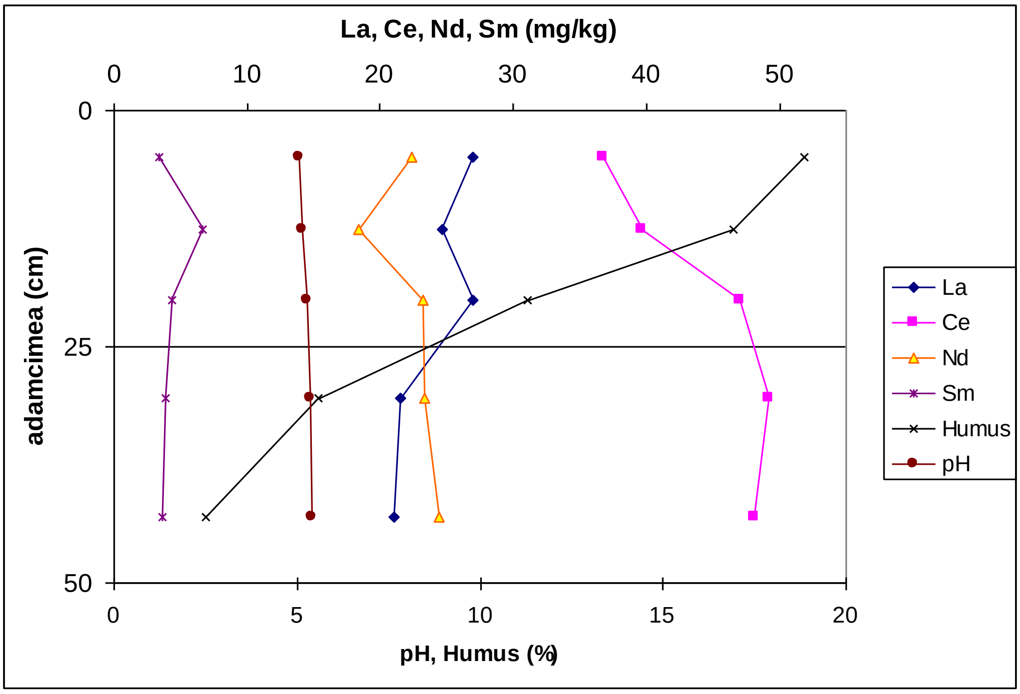

Fig.25. Distribution profile of La, Ce, Nd, Sm to spodisoil (SPO) Gurghiu Mountains (Profile No. 13 peak Saca)

Conclusions

a

Pedological component volcanic arc in the Eastern Carpathians is characterized by the presence of four types of soil (SRTS-2012): Cambisoils (CAM), spodisoils (SPO), andisoil (AND) hidrisoil. (HID).

The main factor that led to the accumulation of volcanic soil macro- and microelements in the Eastern Carpathians is the chemical composition of volcanoes plus parental and convincing manifestation suggestive of classical processes of pedogenesis.

Macro and micronutrients contents in the soils studied are of the same order of magnitude as the values present in calc-alkaline volcanites parenting.

Geochemistry macro is based on quantitative determinations (%) silicate characteristic of classical analysis: Si, Ti, Al, Fe, Ca, Mg, Na, K, S, P. Trace elements are considered characteristic of calc-alkaline volcanoes parental basalt-rhyolite series of heavy metals (Cu, Pb, Zn, Cd, Mn, Co, Ni, Cr, Mo); rare metals (Rb, Sc, Nb, La, Ce, Nd, Sm); metalloids (As). Of these trace elements, analytical data for Rb, Nb, Sc, La, Ce, Sm, Nd is the first time.

Geochemical mobility macro and microelements in soils studied was influenced by pH, organic matter content and clay minerals and volcanic soil structure and texture.

An important role in the abundance of trace elements represents their adsorption by humic acids, oxides of Fe, Mg, Al and clay minerals in soils.

The range of concentrations of macro- and microelements in soils of volcanic Carpathians, within the limits set by various researchers for Earth soils. The abundance of the soil profile of macro and microelements is conditioned by the type of soil, amount of organic matter, clay mineralogy, pH values, texture and adsorption capacity.

Calculation geochemical correlation coefficients showed both macroelements and microelements studied for some positive and negative correlations determined by the affinity between them.

The study contributes substantially developed and convincing volcanic soil geochemical characterization of the Eastern Carpathians based on performance analysis of macro and micronutrients and modern interpretations.

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