GEOLOGIJA. 2006. Vol. 56. P. 13–26

Geochemija • Geochemistry • Геохимия

Siliceous rocks as a raw material of prehistoric artefacts in Lithuania

Valentinas Baltrūnas,

Bronislavas Karmaza,

Dainius Kulbickas,

Tomas Ostrauskas

Baltrūnas V., Karmaza B., Kulbickas T., Ostrauskas T. Siliceous rocks as a raw material of prehistoric artefacts in Lithuania. *Geologija*. Vilnius. 2006. No. 56. P. 13–26. ISSN 1392-110X

The goal of the project and all the work was to establish the distribution, composition, origin and possible directions of transportation of prehistoric siliceous (flint) artefacts in Lithuania. Siliceous concretions and nodules from outcrops and quarries, artefacts from well-investigated Stone Age ancient settlements (Margionys, Kretuonas, Jara, Biržulis, etc.) stored at the funds of Lithuanian museums were surveyed and petrographically described. For revealing the distinctions in the composition of siliceous rocks and distribution of major and trace elements, the samples were analysed by Direct Current Arc Emission Spectrophotometry. The data processing by statistical methods (correlation and cluster analysis) allowed a conclusion that siliceous artefacts could belong to three genetic types of raw material. The artefacts of Margionys ancient settlement can be identified as made of local flint concretions. However, the raw material of Biržulis artefacts is presumably of two types: flint concretions imported from the south and the local stratified flint (silicified gaize). The raw material of Kretuonas and Jara artefacts is also of two genetic types: imported Upper Cretaceous flint from the south south east and the relatively local Upper Devonian siliceous rocks.

Key words: siliceous rock, flint artefact, geochemistry, archaeology, Stone Age, Lithuania

Received 26 January 2006, accepted 12 April 2006

Valentinas Baltrūnas, Bronislavas Karmaza, Institute of Geology and Geography, T. Ševčenkos 13, LT-03223 Vilnius, Lithuania. E-mail: baltrunas@geo.lt; karmaza@geo.lt
Dainius Kulbickas, Vilnius Pedagogical University, Studentų 39, LT-08106 Vilnius, Lithuania. E-mail: geogr.kat@vpu.lt
Tomas Ostrauskas, Institute of Lithuanian History, Kražių 5, LT-01108 Vilnius, Lithuania. E-mail: tomasos67@yahoo.com

INTRODUCTION

Siliceous rocks are sedimentary rocks of chemical and biochemical origin, which are mostly composed of opal and chalcedony or, in rarer cases, by quartz (tripoli, diatomite, gaize, flint, jasper and others). Articles made of solid, shelly and razor-edge siliceous rocks usually referred to as "flint" (hardness according to Moso scale 6.5–7) abound among archaeological artefacts. The first Lithuanian postglacial (Late Palaeolite) inhabitants used flint for the production of arrowheads, knives, burins, scrapers, borers, axes, etc. The natural bedding form of siliceous rocks varies. Cretaceous flint concretions (with a concentric structure) and Devonian siliceous nodules (silicified rounded rock concretions) are sized 2–30 and even 70 cm. The Upper Cretaceous gaize and thin interlayers (lenses) of its silicified varieties lying in the carbonaceous rock mass are known in Lithuania. Siliceous rocks were scattered over a large territory by advancing glaciers in south-western, southern and south-eastern directions. It was found that the network of finding sites of Palaeolithic and Mesolithic flint artefacts almost coincided with the distribution of flint concretions in the Upper Cretaceous carbonaceous sediments (Skuodienė, Katinas, 1981). This was a shrewd observation, yet no special geological research was undertaken.

Based on trace element Direct Current Arc Emission Spectrophotometric analysis, the studied sites and archaeological monuments of South Lithuania were hierarchically classified (Karmaza, Juodagalvis, Ostrauskas, 2001).

A preliminary survey of flint artefacts in the Lithuanian National Museum showed that a rather wide spectrum of rocks with similar properties is referred to as "flint". Their colour (blackish, bluish, grey, white, and brown) and natural contacts with other kinds of rocks show that rocks of different age and genesis are often classified into one group of "flint". According to the available data of visual observations, the artefacts uncovered in the Žemaičiai Upland (e.g., in the Daktariškės, Pabiržulis, Dreniai, Šernelė and other settlements) are made of the local raw material (white silicified gaize) which is not found in other localities (Baltrūnas, Karmaza, Kulbickas, Pukelytė, 2004).

The issues of the use of siliceous rocks as a raw material are relevant in many countries. E. Kalechits (Калечиц, 1984) has studied the use of resources of siliceous rocks in prehistoric times and their distribution in the territory of Belarus. A. Makhnach and L. Gulis (Махнач, Гулис, 1993a, b, c; Махнач, Гулис и др., 2004) have carried out particular investigations of Devonian and Cretaceous flint concretions. V. Petrun et al. (Петрунь, 1971; Бруяко, Видейко, Сапожников, 2005) have thoroughly studied flint artefacts in Ukraine. He managed to distinguish two types of flint (from the Prut and Dnestr basins) spread in the territory of Ukraine.

Sixteen finding sites of flint artefacts were identified in the Upper Volga Valdaj Uplands (Селиванова, 1984; Синицына, 2005). V. Galibin and V. Timofejev carried out a comparative analysis of archaeological flint artefacts found in the East Baltic (St. Petersburg Region, Estonia and Lithuania) Stone Age settlements (Галибин, Тимофеев, 1993). The geochemical properties of flint artefacts and the sources of raw material are also investigated in the north-eastern European part of Russia (the Republic of Komi) (Майорова, Волокитин, 2005).

The issue of using siliceous rocks in the prehistoric times is also relevant in other neighbouring countries. M. Kaczanowska (1986), B. Balcer (1988) and others described the main centres of flint industry and directions of flint artefact distribution in Poland. M. Kobusiewiez (1997) investigated the spread of raw material of flint artefacts in Poland. J. Kozlowski et al. (1981) carried out mineralogical and geochemical investigations of siliceous rocks. Similar research has been carried out in other European countries: J. Konda (1986), E. Bácskay (1984), K. Biró (1997) in Hungary, J. Hoika (1986) and A. Binsteiner (2004) in Germany and C. Becker (1951) and L. Nielsen (1997) in Denmark. Flints are also investigated in Czech, Bulgaria, Slovakia and other countries (http://www.flintsource.net).

Archaeologists of Lithuania have identified three prehistoric complexes of flint mining and manufactories in Ežerynai (Alytus District) (Jablonskytė-Rimantienė, 1966; Rimantienė, 1984), Margionys village and Lake Titnas environs (Varėna District) (Ostrauskas, 2000; Šatavičius, 2002). There are assumptions about a possible different origin of flint and its use in different times, yet exhaustive investigations are lacking. Therefore, the combination of geological (sedimentological) research results with archaeological data would be of service in solving regional archaeological questions and evaluating the character of economic, social and cultural processes in the Stone Age in particular.

The main **aim** of this publication was to determine the spread, composition and genesis of the raw material of prehistoric flint artefacts and the possible directions of its transportation in Lithuania.

The main tasks were:

- to evaluate the dependence of the spread of flint artefacts uncovered in Stone Age settlements on natural bedding areas of siliceous rocks (flint concretions, silicified gaize layers, etc.) in the region;
- to use the chemical composition of flint artefacts for identifying the raw materials and their natural sources *in situ*.

Twelve objects were chosen for the present study, including the archaeologically well-investigated ancient settlements of West, South and East Lithuania and natural sources of some siliceous rocks (Fig. 1).

METHODS

The study was accomplished in a few stages: 1 – familiarization with the archival and literary material on the spread, composition and genesis of siliceous rocks in Lithuania and neighbouring countries and its systematization; 2 – analysis of characteristic flint artefacts of the Stone Age stored at the Lithuanian museums and their statistical evaluation and sampling for laboratory examination (Fig. 2); 3 – field works in the Lithuanian regions known for flint artefacts and sampling for laboratory examination; 4 – laboratory work and statistical and graphical generalization of obtained data.

Direct Current Arc Emission Spectrophotometric analysis was used for determining the distribution of some macro- and microelements in flint varieties (flint concretions, siliceous nodules, stratified gaize) and in archaeological flint artefacts for highlighting the differences. Point sampling was applied; for some archaeological artefacts, samples were taken at the limit of minimal content. A DFS-13 spectrograph from a laboratory of the Institute of Geology and Geography was used for examination of samples. The spectrum lines were deciphered by a DM-100 microdensitometer. The international standard samples were used for control of spectral analysis results. Twenty eight chemical elements were determined (Ag, Al, B, Ba, Ca, Co, Cr, Cu, Fe, Ga, La, Li, Mg, Mn, Mo, Nb, Ni, P, Pb, Sc, Sn, Sr, Ti, V, Y, Yb, Zn, and Zr). The measuring units were mg/kg for most of them or percentage for Al, Ca, Fe and Mg. The limit of tracing chemical elements in rocks

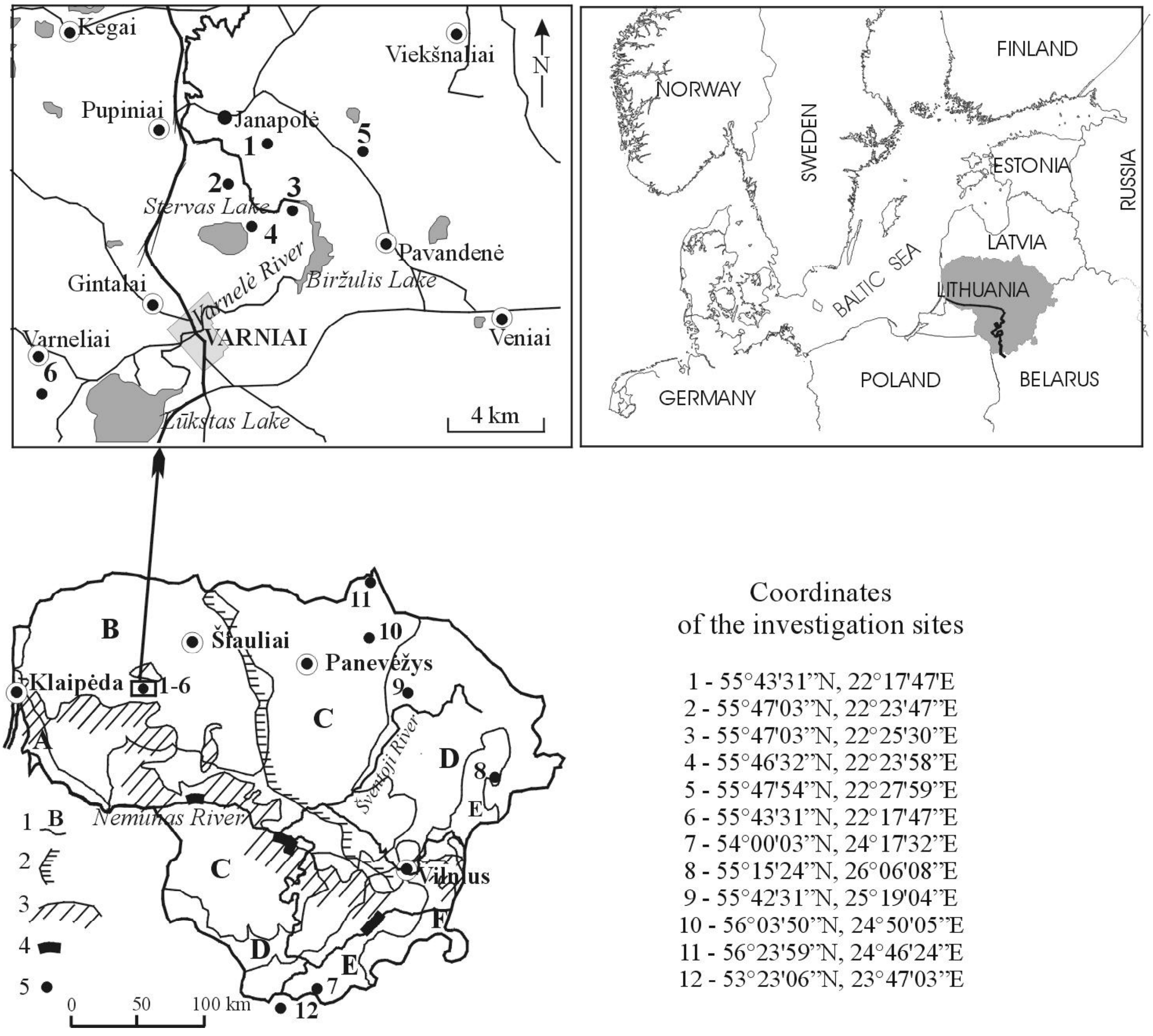

Fig. 1. Investigation sites of silicites and their artefacts.
1 – geomorphological regions according to the Atlas of the Lithuanian SSR (1981): Baltic Depression (A), Žemaičiai–Kuršas (B), Baltija Plain (C), marginal morainic uplands of the last glaciation (D), glaciofluvial plains of the last glaciation (E), uplands of the penultimate glaciation (F); *2* – the spread boundary of Devonian deposits in the Lithuanian sub-Quaternary surface (Lietuvos geologija, 1994); *3* – the spread boundary of Cretaceous deposits in Lithuania (Lietuvos geologija, 1994); *4* – outcrops of Cretaceous carbonaceous flint deposits; *5* – investigation sites of siliceous concretions, nodules and archaeological artefacts: *1* – Kalniškiai, *2* – Dreniai, *3* – former isthmus of Biržulis Lake, *4* – Ožnugariai, *5* – Šūkainiai, *6* – Jonikaičiai, *7* – Margionys, *8* – Kretuonas, *9* – Jara, *10* – Sviliai, *11* – Nemunėlio Radviliškis, *12* – Grodno (Grandichi) chalk quarry (Belarus)

1 pav. Silicitų ir jų dirbinių tyrimo vietos.
1 – geomorfologinės sritys pagal Lietuvos TSR atlasą (1981): Baltijos duburio (A), Žemaičių-Kuršo (B), Pabaltijo žemumos (C), paskutiniojo apledėjimo pakraštinių moreninių aukštumų (D), paskutiniojo apledėjimo fliuvioglacialinių lygumų (E), priešpaskutiniojo apledėjimo aukštumų (F); *2* – devono sistemos nuogulų paplitimo riba Lietuvos pokvarteriniame paviršiuje (Lietuvos geologija, 1994); *3* – kreidos sistemos nuogulų paplitimo riba Lietuvoje (Lietuvos geologija, 1994); *4* – kreidos sistemos karbonatinių titnagingų nuogulų išeigos; *5* – silicitinių konkrecijų, gniutulų ir archeologinių dirbinių tyrimo vietos: *1* – Kalniškiai, *2* – Dreniai, *3* – Biržulio ežero buvusi sąsmauka, *4* – Ožnugariai, *5* – Šūkainiai, *6* – Jonikaičiai, *7* – Margionys, *8* – Kretuonas, *9* – Jara, *10* – Sviliai, *11* – Nemunėlio Radviliškis, *12* – Grodno (Grandiči) kreidos karjeras (Baltarusija)

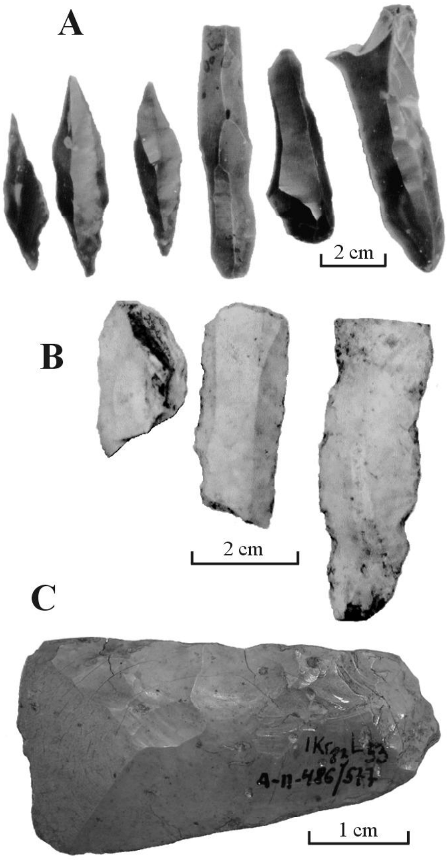

Fig. 2. Artefacts of siliceous rocks from ancient settlements (*A* – Kabeliai, *B* – Duonkalnis, *C* – Kretuonas). Photo of the artefact from Kretuonas by Gytis Piličiauskas

2 pav. Senovės gyvenviečių (*A* – Kabelių, *B* – Duonkalnio, *C* – Kretuono) dirbiniai iš silicitinių uolienų. Kretuono dirbinio nuotraukos autorius Gytis Piličiauskas

(percentage) was Al – 0.3, Ca – 0.05, Fe – 0.1, Mg – 0.01. The limit of identifying trace elements (mg/kg) was Ag – 0.03, B – 5, Ba – 30, Co – 1, Cr – 2, Cu – 2, Ga – 1, La – 3, Li – 10, Mn – 10, Mo – 0.5, Nb – 3, Ni – 2, P – 300, Pb – 3, Sc – 1, Sn – 1, Ti – 10, V – 1, Y – 0.3, Yb – 0.3, and Zn – 10. Only traces of some chemical elements were detected (Ba, La, Sc, Y, Yb). The content of other chemical elements was at the tracing limit (Al, Co, Ga, Mo, Nb, Pb, Sn, Sr, Zn, and Zr). Thirteen elements found in all samples (Ag, B, Ca, Cr, Cu, Fe, Li, Mg, Mn, Ni, P, Ti, V) were chosen for a more detailed geochemical analysis. The statistical parameters of element distribution were calculated using Excel program. Their interrelations (correlation coefficients) and associations (clusters) were determined by the method of cluster analysis (*Statistika* program Ward's method). The average concentration values of comparatively poorly variable chemical elements were used for statistical generalization. The correspondence of their distribution pattern to the normal and lognormal pattern was checked. Three samples with anomalous values of some elements from Šūkainiai and Dreniai ancient settlements were eliminated from some calculations. The anomalous values of some elements were up to 12 times as large by their average contents (Ca – 1256%, Mg – 1121%, P – 407%, Mn – 449%, Ti – 181%).

GEOLOGICAL SETTINGS OF THE STUDY OBJECTS

The distribution of the objects studied depended on the available information about the ancient settlements and abundance of archaeological material, peculiarities of geomorphological domains and regions and the geological structure of sub-Quaternary surface and Quaternary sediments (Lietuvos TSR atlasas, 1981; Lietuvos geologija, 1994; Lietuvos Žemės gelmių..., 2004; Guobytė, 2001) (Fig. 1).

Biržulis area. It is situated in the Middle Žemaičiai Upland and includes the strand of Lake Biržulis, Dreniai, Kalniškiai and Ožnugariai archaeological settlements whose flint artefacts were geochemically analysed. The area also includes Šūkainiai and Jonikaičiai localities from which samples of gaize and its silicified varieties were taken. The sub-Quaternary surface is lying at a depth of 150–314 m. Terrigenous Upper Jurassic and Lower Cretaceous sediments (sand, silt and clay) predominate. The present northern boundary of the spread area of Upper Cretaceous carbonaceous sediments (chalk and chalk marl) runs south of Lake Biržulis (Lietuvos geologija, 1994; Lietuvos Žemės gelmių..., 2004). At the time of the first glaciation, this boundary might have been north of the study area. This is proved by rare glacial erosion relicts of these sediments further north. The Upper Cretaceous Campanian rocks (chalk and marl with gaize patches and interlayers) 10–15 km north of Lake Biržulis represent one of such "islands". Advancing glaciers eroded these sediments, reworking gaize and its silicified varieties (whitish flint).

Jara area. It is situated in the West Aukštaičiai Plateau, on the right bank of the Jara rivulet, about 4 km north-west of Svėdasai, near the Visėtiškės village. It is a plain formed by a glacier lobe and later covered by glaciolacustrine, glaciofluvial and alluvial sediments. The complex of Quaternary sediments is mainly composed of till wedging in the south-eastern direction (toward the Jara valley). The sub-Quaternary surface lying at a depth of 10–25 m is composed of the Upper Devonian sediments of the Šventoji Formation (sand, sandstone, aleurolite, aleurite, and clay). The Upper Neogenic sand is sporadically spread further south. Sediments of the Jara, Suosa and Kupiškis formations (dolomite, marl, clay, sandstone, and aleurite) are spread further north and north-west of the locality. The dolomite complex of Įstras Formation with solid silicate nodules dispersed by advancing northern glaciers over the area is found even further north.

Lake Kretuonas area. It is situated in the glaciolacustrine "bay" of the glaciofluvial North-Eastern Plain in the eastern periphery of Lithuania. The sandy surface of the area is extending at a height of 160 m above the present sea level. The environs of Lake Kretuonas used to be in the glacier divide zone between the eastern (Dysna) lobe of the glacier and its northern tongue. The glacier lobe advancing from the north-east (the northern part of Belarus) transported till material with the local Devonian rock fragments. The Upper Devonian flint concretions and other kinds of siliceous rocks occur among them. The sub-Quaternary surface, seamed with paleoincisions lying at a depth of 100–160 m, is composed of Devonian domerite, clay, dolomite and gypsum of Ledai Formation, and aleurolite, clay and sandstone of Kernavė Formation.

Margionys area. It is in the southern periphery of Lithuania in the sandy South-Eastern (Dainava) Plain, which is the spread area of glaciofluvial sediments (Baltrūnas, 2001). The Margionys sandur left by the last glaciation is composed of badly sorted gravel with coarse pebble and scanty boulders up to 0.6 m in diameter. It contains many flint concretions and debris. Flint fragments also abound on the surface. According to the data of Margionys borehole 357, the thickness of Quaternary sediments in this locality reaches 183.5 m. The thickness is smaller (120–100 m) east and west of the locality. This sediment complex is overlying the Campanian carbonaceous rocks (chalk, chalk marl, etc.) of the Upper Cretaceous. It is composed of till layers left by the Middle Pleistocene glaciations and interstratified interglacial lacustrine sediments of Butėnai (Holsteinian) Formation. The *in situ* carbonaceous Cretaceous sediments are exposed only further south of Margionys (Grodno District of Belarus). The flint concretions lying close to the surface or exposed are related with the large blocks (up to several million m^3) of Cretaceous carbonaceous rocks carried by glaciers or with their erosion sites (residua). These blocks of old rocks and their relicts are characteristic of South Lithuania (Baltrūnas, 1995, 2002).

Other objects. These are objects not directly related with archaeological finds, but analysis of siliceous contained in them is of comparative value. The outcrop of the Upper Devonian layers of Įstras Formation in North Lithuania, on the left bank of the Nemunėlis near Nemunėlio Radviliškis Township, is one of these objects. This outcrop and the Muoriškės outcrop not far away are known as a finding site of siliceous nodules in dolomite layers (Водзинскас, 1966). Siliceous nodules up to 20–30 cm in diameter occur in the dolomite powder interlayers which are a product of the washout of the former gypsiferous dolomite in North Lithuania.

An irregular flint concretion 15 cm in diameter was found among field boulders not far from Sviliai village 10 km north-north-east of Vabalninkas Township in North Lithuania. It was geologically examined. The internal section of this concretion has a concentric structure. The internal (grey) part is enveloped by a thin white stripe and the latter by an external pink stripe. Similar flint concretions are found in the outcrops and quarries of Devonian sediments in the central and eastern parts of Latvia and in the northern part of Belarus (Гравитис, 1963; Сорокин, 1963; Махнач, Гулис, 1993a; Махнач, Гулис и др., 2004).

One of the Upper Cretaceous flint concretions 10–15 cm in diameter from the Grodno (Grandichi) chalk quarry (Belarus) south of Lithuania was taken for comparison. The Neolithic and Bronze Age flint mines are known in the western part of Grodno Region. Flint artefacts might have been transported from this locality to the neighbouring countries (Археалогія..., 1993; Quaternary..., 1997).

DATA OF GEOCHEMICAL STUDIES

Geochemical properties of silicite concretions and nodules

Generalization of geochemical data of archaeological flint artefacts was based on a preliminary analysis of chemical element distribution patterns in flint concretions and siliceous nodules. Rock fragments where one kind of siliceous rocks merge into rocks of another kind were also analysed. Concretions of diagenetic flint composed of amorphous SiO_2 (0.53–6.44%) and crystalline (83.6–91.8) quartz abound in the Cretaceous carbonaceous rocks of Lithuania (Radzevičius, 2004). Based on the analysis of these trace elements, it was expected to differentiate the stratigraphic genetic diversity of siliceous rocks. The flint concretions found in the Grodno (Grandichi) chalk quarry (Belarus) are characterized by a very irregular form. Their size ranges between a few cm to 1 m (Fig. 3). Belarusian researchers studied the petrographic, mineralogical and geochemical composition of these concretions (Махнач, Гулис, 1993 a, b, and c) which are also predominated by SiO_2 (95.4% on the average) and contain small concentrations of Al_2O_3 (1.6%), Fe_2O_3 (0.6%) and CaO (0.6%) (Махнач, Гулис, 1993b). We conclude that the determined concentrations of oxides and some trace elements (Ni, Cu, Ti, Mn, Ba, Zr, B, and Y) in different spots of concretions are comparable and hardly depend on formation of patina and recrystallization processes.

Five samples of a blackish flint concretion from the Grodno quarry were taken for spectral analysis (Table 1). The samples strongly differ in B, Cu, Mg, Mn, Ti, and V concentrations. The correlation coefficients of the five elements showed a strong positive correlation of most of the elements except B, Mn, P, and partly Li. The strongest links are characteristic of Cu, Ca, V, Mg, Fe, and Ag.

Samples for the same analysis were taken from one flint concretion found in the ancient Margionys settlement complex, manufactories and flint quarries. A white patina (weathering film), flint with patina and blackish and grey flint from the inner part of the concretion were analysed separately. The samples with patina differ from the flint of the inner part by smaller concentrations of Cr, Fe, Li, Mn, P, and V.

Three samples from the Devonian flint concretion with a concentric structure found in the environs of Sviliai were analysed for comparison. The external pinkish (up to 15 mm in thickness), intermediate white (up

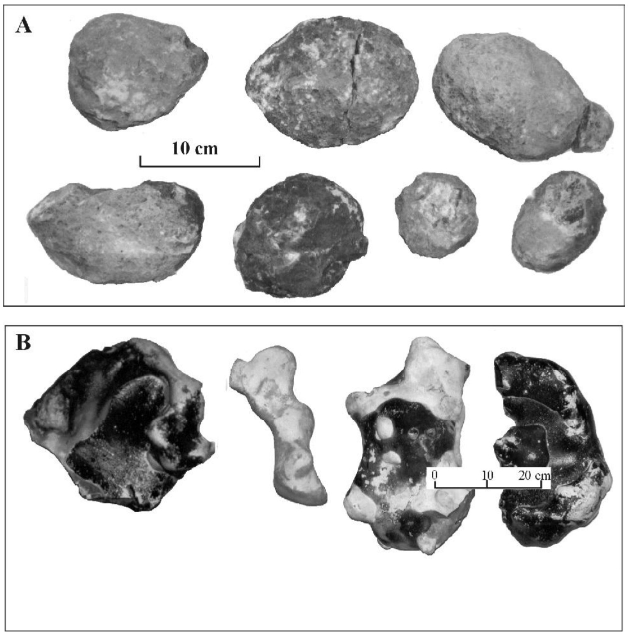

Fig. 3. Siliceous nodules of the Upper Devonian Įstras Formation from the Nemunėlio Radviliškis outcrop (*A*) and Upper Cretaceous flint concretions from the Grodno (Grandichi) quarry (*B*, exhibits from the Geological Museum of the Institute of Geology and Geography)
3 pav. Viršutinio devono Įstro svitos silicitų gniutulai iš Nemunėlio Radviliškio atodangos (*A*) ir viršutinės kreidos titnago konkrecijos iš Gardino karjero (*B*, pavyzdžiai iš Geologijos ir geografijos instituto muziejaus)

to 7 mm) and the largest internal (grey) parts were examined. The internal part stands out for smaller concentrations of B, Ca, Fe, Mg, Mn, Ni, Ti, and V.

An archaeological flake from the Kalniškiai ancient settlement marked by blending from light grey gaize to brownish grey flint was examined. The pure flint contains smaller concentrations of Ca, Cr, Cu, Fe, Li, P, and Ti than flint with gaize and pure gaize.

A nodule of Devonian siliceous from the Nemunėlio Radviliškis outcrop was tested in three samples. It was determined that the cementing light grey part (including the external patina) slightly differs from the internal striped part (dark brown flint) by higher concentrations of Ca, Mg, Mn, Ti, and V and slightly smaller concentrations of Ag, B, P, Li (Fig. 4).

The obtained data show that differences among separate parts of concretions and nodules in concentrations of chemical elements are rather pronounced. These differences of geochemical composition became especially evident after cluster analysis of the obtained data (see below).

Distribution patterns of the chemical composition of siliceous artefacts from ancient settlements

The composition of macro- and trace elements in siliceous concretions and nodules is assumed to reflect the composition of the "insoluble residue" of the primary carbonaceous silt (allothigenous and authigenous part).

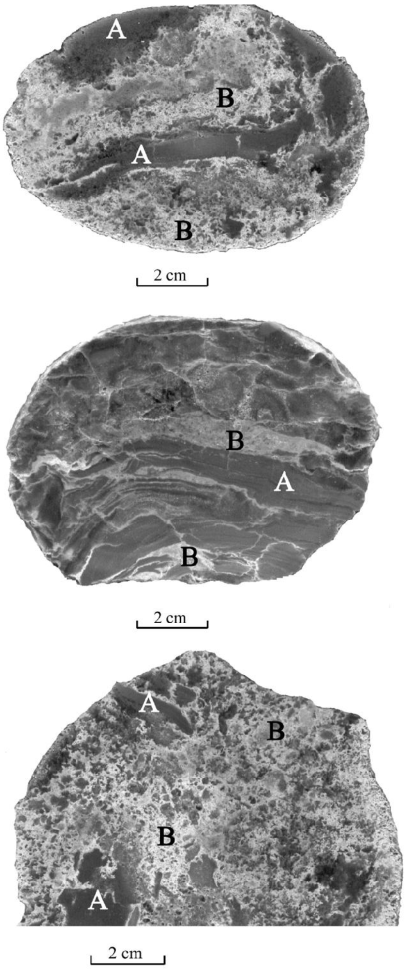

Fig. 4. Siliceous nodules of the Upper Devonian Įstras Formation from the polished section of the Radviliškio Nemunėlis outcrop. *A* – irregular concretion of dark brown flint; *B* – light grey mass of chalcedone and quartz grains (0.01–0.1 mm) cementing a banded and spotted siliceous nodule
4 pav. Viršutinio devono Įstro svitos silicito gniutulų iš Nemunėlio Radviliškio atodangos poliruotos nuopjovos. *A* – tamsiai rudo titnago netaisyklingos formos sutankėjimas; *B* – juostuoto ir dėmėto silicito gniutulą cementuojanti šviesiai pilka chalcedono ir kvarco grūdelių (0,01–0,1 mm) masė

The latter is a reflection of sedimentation conditions (salinity of the basin, composition of specified silt, etc.). The average concentrations of chemical elements for different objects were calculated taking into consideration the high geochemical resemblance of siliceous rocks and the variation of geochemical composition in their concretions and nodules

Sixteen groups of samples, which are relatively homogeneous in their genesis, composition and visual description, were analysed. The archaeological artefacts from each of the ancient settlements (e.g., Kretuonas, Dreniai and Kalniškės) were additionally classified into a few groups. For example, the Kalniškiai and Jara flints and Nemunėlio Radviliškis siliceous rocks stand out for elevated concentrations of Ag, Grodno and Margionys flints for higher concentrations of B, Jonikaičiai, Šūkai-

Table 1. **Distribution of chemical element composition in flint concretions, siliceous nodules and archaeological artefacts. % for Ca, Fe, Mg and mg/kg for other trace elements**
1 lentelė. **Atskirų titnago konkrecijų, silicito gniutulų ir archeologinių dirbinių cheminių elementų sudėties pasiskirstymas (Ca, Fe, Mg nustatyti %, kiti mikroelementai – mg/kg)**

Locality	Rock	Note	Nr.	Ag	B	Ca	Cr	Cu	Fe	Li	Mg	Mn	Ni	P	Ti	V
Grodno	Concretion of black flint	Unpatina	1	0.040	84	0.03	14	3.6	0.07	5	0.014	20	9	500	70	1.2
Grodno		Unpatina	2	0.045	84	0.06	13	5.8	0.10	8	0.033	200	9	560	120	2.3
Grodno		With thin patina	3	0.080	105	0.12	19	9.0	0.20	9	0.090	60	12	560	120	4.0
Grodno			4	0.064	125	0.07	17	6.0	0.12	5	0.027	30	14	400	120	2.0
Grodno			5	0.040	110	0.03	10	3.9	0.07	5	0.013	20	7	500	55	1.0
		Average		**0.054**	**102**	**0.06**	**15**	**5.7**	**0.11**	**6**	**0.035**	**66**	**10**	**504**	**97**	**2.1**
Margionys	Concretion flint	White patina	1	0.036	98	0.06	8	4.0	0.07	8	0.014	40	8	850	56	1.6
Margionys		Grey flint	2	0.043	94	0.05	20	4.8	0.07	12	0.013	130	11	600	40	2.3
Margionys		Black flint	3	0.470	100	0.07	36	7.6	0.30	10	0.016	190	25	1200	56	2.4
Margionys		Flint with patina	4	0.066	84	0.04	10	5.2	0.07	9	0.011	70	11	850	33	1.8
		Average		**0.154**	**94**	**0.06**	**19**	**5.4**	**0.13**	**10**	**0.014**	**108**	**14**	**875**	**46**	**2.0**
Sviliai	Concretion flint	External pinkish part	1	0.090	86	0.09	6	11.5	0.23	5	0.038	45	16	400	210	3.3
Sviliai		Intermediate white part	2	0.064	36	0.03	12	4.8	0.07	5	0.018	20	5	400	40	1.0
Sviliai		Largest internal grey part	3	0.060	66	0.10	35	5.0	0.21	7	0.062	50	10	500	330	5.0
		Average		**0.071**	**63**	**0.07**	**18**	**7.1**	**0.17**	**6**	**0.039**	**38**	**10**	**433**	**193**	**3.1**
Kalniškiai	Flake of rock and artefact	Flint wiht gaize	1	0.040	58	0.06	11	3.0	0.12	8	0.019	40	7	800	90	2.5
Kalniškiai		Flint	2	0.030	37	0.03	3	1.6	0.07	5	0.017	20	3	500	80	1.6
Kalniškiai		Gaize	3	0.060	58	0.06	8	2.4	0.12	10	0.02	20	3	800	88	2.0
		Average		**0.043**	**51**	**0.05**	**7**	**2.3**	**0.10**	**8**	**0.019**	**27**	**4**	**700**	**86**	**2.0**
Nemunėlio Radviliškis	Siliceous nodule	Wiht internal striped part	1	0.250	23	1.2	29	11.0	0.22	49	1.2	155	22	500	105	29.0
Nemunėlio Radviliškis			2	0.100	19	2.7	22	2.0	0.11	30	1.5	140	9	500	86	6.4
Nemunėlio Radviliškis		Wiht patina unstriped part	3	0.100	16	5.2	35	4.3	0.19	28	2.4	200	16	350	125	11.0
		Average		**0.150**	**19**	**3.0**	**29**	**5.8**	**0.17**	**36**	**1.7**	**165**	**16**	**450**	**105**	**15.5**

niai and Nemunėlio Radviliškis siliceous for higher concentrations of Ca and Jonikaičiai, Šūkainiai, Nemunėlio Radviliškis silicites and Kretuonas and Margionys flints for higher concentrations of Cr. The concentrations of other elements also differ. Comparison of flint and silicified gaize artefacts from the ancient settlements of Kretuonas, Dreniai and Kalniškiai has shown that gaize contains smaller concentrations of B or Ag and Mg and, sometimes, higher concentrations of Cu, Fe, Mn, P and Ti. The Kalniškiai and Dreniai artefacts made of flint and silicified gaize are most comparable. Cluster analysis was applied to the mentioned 16 groups of samples (see below).

In order to classify the archaeological artefacts according to their belonging to concrete ancient settlements and maximally increase the number of samples for statistical evaluation, generalization of geochemical data and correlation analysis, the number of groups was reduced to 11 (Table 2). In some cases (Kretuonas, Dreniai and Kalniškiai) the generalized geochemical data levelled down the petrographic diversity of artefacts. According to the data of spectral analysis, the Nemunėlio Radviliškis, Šūkainiai and Jonikaičiai siliceous samples are most comparable. The samples from Dreniai (B, Ca, Mg, and Ti), Jara (Ag, Cr, Mn, Ni) and Kalniškiai (Ag, Ti) resemble these siliceous samples in some groups of elements.

Table 2. **Arithmetic mean (X), standard deviation (S) and variation coefficient (v, %) of chemical elements in siliceous rock artefacts from ancient settlements, siliceous concretions and nodules. % for Ca, Fe, Mg and mg/kg for other trace elements**

2 lentelė. **Senųjų gyvenviečių silicito dirbiniuose, silicitų konkrecijose ir gniutuluose nustatytų cheminių elementų aritmetinis vidurkis (X), standartinis nukrypimas (S), variacijos koeficientas (v, %) (Ca, Fe, Mg nustatyti %, kiti mikroelementai – mg/kg)**

Locality	Number of samples	Ag			B			Ca			Cr			Cu			Fe			Li		
		X	S	v	X	S	v	X	S	v	X	S	v	X	S	v	X	S	v	X	S	v
Kalniškiai	10	0.110	0.050	45	50	4.1	8	0.081	0.010	12	10	1.0	10	3.9	1.0	25	0.12	0.01	9	10	0.8	8
Dreniai	13	0.065	0.010	15	39	6.7	17	1.985	1.918	97	11	1.1	10	3.9	0.5	12	0.18	0.06	33	9	0.9	10
Biržulis	12	0.053	0.004	8	53	2.9	6	0.039	0.003	8	13	1.7	13	3.8	0.4	11	0.10	0.00	1	9	1.0	10
Ožnugariai	5	0.051	0.002	5	68	4.0	6	0.080	0.010	13	17	3.0	18	3.8	0.5	13	0.12	0.02	15	10	0.9	9
Šūkainiai ir Jonikaičiai	6	0.086	0.021	24	39	4.0	10	1.800	1.221	68	33	6.5	20	6.4	0.8	12	0.44	0.08	17	9	0.9	10
Margionys	27	0.050	0.016	32	82	2.7	3	0.050	0.003	6	17	1.3	7	4.5	0.2	4	0.21	0.08	38	9	0.2	2
Kretuonas	32	0.044	0.003	6	51	3.5	7	0.104	0.030	29	16	2.9	18	4.5	0.2	4	0.20	0.06	30	9	0.5	5
Jara	21	0.129	0.052	40	57	2.7	5	0.069	0.020	29	17	1.3	7	5.2	0.5	10	0.21	0.07	33	12	0.6	5
Sviliai	3	0.071	0.009	13	63	14.5	23	0.073	0.020	27	18	8.8	50	7.1	2.2	31	0.17	0.05	29	6	0.7	12
Nemunėlio Radviliškis	9	0.157	0.045	29	21	1.3	6	2.620	0.631	24	26	1.9	7	4.8	0.9	18	0.15	0.01	9	27	3.2	12
Grodno	5	0.054	0.008	15	102	7.9	8	0.062	0.016	26	15	1.6	11	5.7	1.0	17	0.11	0.02	21	6	0.9	14

Locality	Number of samples	Mg			Mn			Ni			P			Ti			V		
		X	S	v	X	S	v	X	S	v	X	S	v	X	S	v	X	S	v
Kalniškiai	10	0,099	0,0076	8	36	8,8	25	6,7	0,72	11	588	52	9	145	29	20	3,3	0,44	14
Dreniai	13	0,226	0,1900	84	73	15,1	21	7,1	0,67	9	596	40	7	129	31	24	2,7	0,51	19
Biržulis	12	0,020	0,0050	25	42	7,4	17	7,9	0,93	12	598	42	7	100	12	12	2,1	0,15	7
Ožnugariai	5	0,035	0,0100	29	62	16,3	27	11,4	3,38	30	684	54	8	106	7	7	2,5	0,02	1
Šūkainiai ir Jonikaičiai	6	0,135	0,0272	20	323	226	70	11,4	2,39	21	2210	1368	62	717	186	26	21,7	7,69	35
Margionys	27	0,010	0,0003	3	110	8,3	8	12,5	0,83	7	552	38	7	45	3	7	1,9	0,06	3
Kretuonas	32	0,033	0,0070	21	53	4,8	9	8,4	0,57	7	562	52	9	98	15	16	2,5	0,27	11
Jara	21	0,013	0,0003	2	108	9,7	9	9,5	0,62	7	529	43	8	77	7	9	2,2	0,14	6
Sviliai	3	0,039	0,0127	32	38	9,3	24	10,3	3,18	31	433	33	8	193	84	44	3,1	1,16	37
Nemunėlio Radviliškis	9	1,422	0,1993	14	144	9,7	7	14,7	1,57	11	490	45	9	94	15	16	11,4	3,49	31
Grodno	5	0,035	0,0100	28	66	34,3	52	10,1	1,18	12	504	29	6	97	14	15	2,1	0,53	25

Table 3. **Ratio between the average value of chemical elements in Margionys ancient settlement artefacts and artefacts of other ancient settlements**

3 lentelė. **Margionių senovės gyvenvietės dirbiniuose nustatytų cheminių elementų vidurkio santykis su kitų senovės gyvenviečių dirbinių, taip pat silicitinių uolienų cheminių elementų vidurkiu**

Locality	Number of samples	Elements												
		Ag	B	Ca	Cr	Cu	Fe	Li	Mg	Mn	Ni	P	Ti	V
Kalniškiai	10	0.45	1.64	0.62	1.67	1.16	1.81	0.89	0.10	3.06	1.86	0.94	0.31	0.58
Dreniai	12	0.74	2.00	0.75	1.41	1.20	1.14	0.99	0.27	1.45	1.69	0.94	0.35	0.67
Biržulis	12	0.94	1.54	1.28	1.35	1.19	2.17	0.97	0.49	2.59	1.58	0.92	0.45	0.92
Ožnugariai	5	0.98	1.21	0.63	1.01	1.20	1.72	0.92	0.29	1.79	1.10	0.81	0.43	0.77
Šūkainiai ir Jonikaičiai	6	0.58	2.10	0.03	0.51	0.70	0.47	1.00	0.07	0.34	1.10	0.25	0.06	0.09
Kretuonas	26	1.19	1.45	0.78	1.04	0.99	1.16	1.04	0.30	2.42	1.47	1.00	0.65	0.94
Jara	21	0.39	1.44	0.73	0.98	0.86	0.98	0.76	0.77	1.02	1.31	1.04	0.58	0.88
Nemunėlio Radviliškis	9	0.32	3.86	0.02	0.67	0.94	1.41	0.34	0.01	0.76	0.85	1.13	0.48	0.17

Table 4. **Associations of chemical elements in artefacts from ancient settlements and siliceous rocks determined by correlation analysis. In bracket precarious CR**
4 lentelė. **Tyrinėtų senovės gyvenviečių ir silicitinių uolienų cheminių elementų asociacijos, nustatytos koreliacinės analizės pagrindu (skliausteliuose abejotinas KR)**

Locality	Associations
Kalniškiai	V–Ti–Fe–Mg–(Cu)–(Ni); Cr–Ca–B–Ag–(Cu)–(Ni)
Dreniai	Li–Cr–Ti–V–Mn–Fe–(Ni); B–Ca–P–Mg–(Cu)
Biržulis	V–Fe–Ti; Li–Mn–Ni–Cu–Cr–(V)–(Ag); B–Ca–Mg–(P)
Margionys	Ag–Cr–Cu–Ni–Mn; Ti–V–Fe–Mg–Ca
Kretuonas (flint artefact)	Mn–Ni–Ti–Cu–Fe–Li; Ca–Ag–Mg–(Ti)–(Li)
Kretuonas (silicified gaize artefact)	Ca–Mg–Fe–Mn–Cr–Cu–Ni–Ti; Ag–P–B–Li
Jara	Cu–Fe–Mn–(Cr); Li–Ti–V; Cr–Ni– B
Nemunėlio Radviliškis	Mg–Ca–Mn–(Ti); Ni–Fe–Cu–Li–V–Ag– (P)–(B)
Margionys, Kretuonas and Biržulis together	Ca–Mg–Fe–V–Ti–(Cu); Mn–Ni–B–Li–(Cu)–(Cr)

The average values of the geochemical composition of siliceous artefacts from the ancient Dreniai, Kretuonas, Jara, Biržulis, Ožnugariai, and Kalniškiai settlements were compared with the average value of flint artefacts from Margionys settlement (Table 3). This allowed determining the resemblance of the chemical composition of the archaeological finds studied with the finds from Margionys settlement. According to most chemical elements, the Margionys samples are most comparable with the finds from Kretuonas, Jara, Ožnugariai and Biržulis (?). The siliceous rocks from Nemunėlio Radviliškis, Jonikaičiai and Šūkainiai showed the greatest difference of chemical composition from the Margionys flint artefacts. The differences of Kalniškiai and Dreniai samples were less conspicuous.

Interesting and informative results were obtained by correlation analysis of chemical elements often revealing the causality of correlative relations (CR) of components (Table 4). CR matrixes and their analysis showed a high variety of correlations from a well-marked existence of two "antagonistic" associations to 3–4 small and inconspicuous groups correlated by one or two elements (Fig. 5).

From this point of view, it is expedient to analyse data from the ancient Margionys settlement, because its archaeological artefacts are most likely made of the local Cretaceous flint concretions. The distinguished [Ti–V–Fe–Mg–Ca] and [Ag–Cr–Cu–Ni–Mn] associations presumably reflect the chemical composition of the insoluble residue and thindispersed part of the Upper Cretaceous sediments. The CR analysis of the samples from the combined group of ancient settlements (Margionys, Kretuonas and Biržulis) revealed two associations [Ca–Mg–Fe–V–Ti–(Cu)] and [Mn–Ni–B–Li–(Cu)–(Cr)] (in bracket precarious CR). The association [Ca–Mg–Fe–Ti–V] is characteristic of Margionys alone. The second association most probably implies sediments of higher salinity basins.

The Kretuonas artefacts were visually classified into two types of siliceous rocks for correlation analysis. The flint artefacts from Kretuonas are marked by two associations, [Mn–Ni–Ti–Cu–Fe–Li] and [Ca–Ag–Mg–(Ti)–(Li)], which resemble the element associations in the artefacts of Margionys settlement. Other associations mark artefacts made of silicified gaize: [Ca–Mg–Fe–Mn–Cr–Cu–Ni–Ti] and [Ag–P–B–Li]. The correlative relations of chemical

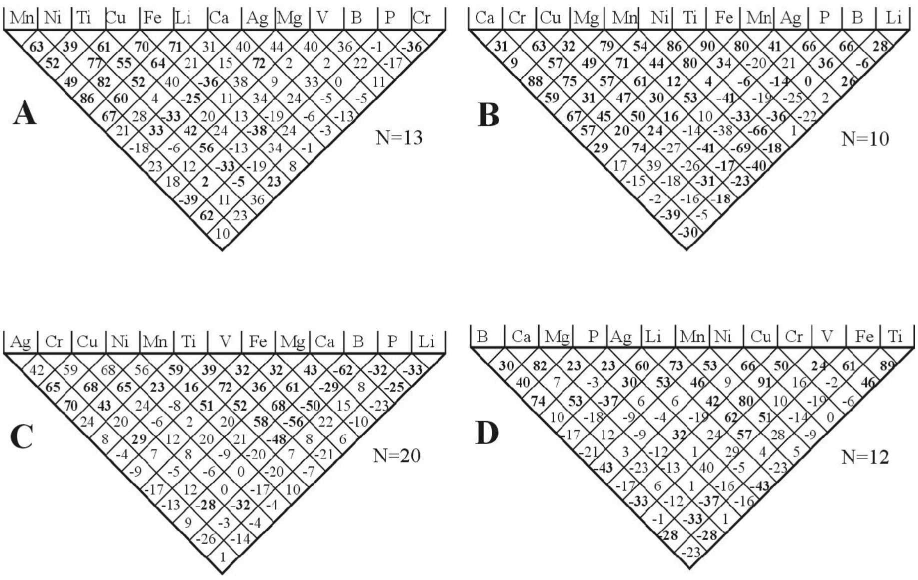

Fig. 5. Correlative relations ($r_{xy}10^{-2}$) of chemical elements determined in flint (*A*) and silicified gaize (*B*) artefacts from Kretuonas and flint artefacts from Margionys (*C*) and Biržulis (*D*). Significant correlation coefficients are marked for q = 0.05. *N* – number of samples

5 pav. Kretuono titnago (*A*) ir silicifikuotos opokos (*B*), Margionių (*C*) ir Biržulio (*D*) titnago archeologiniuose dirbiniuose nustatytų cheminių elementų koreliacijos ryšiai ($r_{xy}10^{-2}$). Išryškinti reikšmingi koreliacijos koeficientai esant q = 0,05. *N* – tirtų mėginių skaičius elements in the Devonian siliceous nodules from the Nemunėlio Radviliškis outcrop are comparable to the ones in the artefacts from Kretuonas. Preservation of the groups [Mg–Ca–Mn–(Ti)] and [Li–Ag–(P)–(B)] is also characteristic.

Data of the correlation analysis allow distinguishing three types of CR. One of them is distinguished by a stable group [V–Ti–Fe] sometimes joined by Mg, Mn, Ca. The second type is characterized by the group [B–Ca–Mg] sometimes joined by P, Mg, Cu, Cr. The third type is distinguished by the group [Mg–Ca–Mn] sometimes joined by Fe, Cr, Ti. The CR of the first type are related with the Upper Cretaceous flint concretions and artefacts made of them (Margionys, partly Biržulis, Kalniškiai, Dreniai, and Kretuonas). The CR of the second type are identified in the ancient settlements of Biržulis area alone and are related with the spread of the Upper Cretaceous flint (silicified gaize) in Žemaitija Region. The CR of the third type are presumably characteristic of the Upper Devonian siliceous rocks and are preserved in the distribution pattern of chemical elements in the Kretuonas and Jara archaeological artefacts.

Results of cluster analysis

Cluster analysis of all samples was performed by Ward's method, expecting, at least partly, to group them according to localities. According to the distribution of the main chemical elements, 143 available samples were grouped into seven large clusters of various ranks. Samples taken from one siliceous nodule and from the same investigation objects (ancient archaeological settlements, Nemunėlio Radviliškis outcrop, etc.) were classified into large clusters. After elimination of gaize and silicified gaize samples from Šūkainiai and Jonikaičiai (marked by anomalous values of some chemical elements), the number of large clusters reduced to two. Each of them was subdivided into 2–3 smaller ones. Typically, that samples of Nemunėlio Radviliškis Devonian siliceous rocks are included into one group. The artefacts from the ancient settlements, though in groups of 4–8, are included into different large clusters. A similar test with the samples from three settlements (Margionys, Biržulis and Kretuonas) ended in distinguishing three large clusters including artefacts from all the three objects. All the three clusters include samples of silicified gaize, flint of different colours with patina and without it. These results were presumably predetermined by a high sensitivity (precision) of the method and the similarity of the chemical composition of siliceous rocks. Different results were obtained generalizing by this method the average values of geochemical data from 16 relatively homogeneous groups (Fig. 6). The grouping of samples in three larger clusters is rather easily explained. One cluster included samples of Nemunėlio Radviliškis Devonian siliceous rocks and Šūkainiai and Jonikaičiai gaize and its silicified varieties. The second cluster included silicified gaize from Kalniškiai, Dreniai and Kretuonas and flint artefacts from Biržulis and Dreniai. The third largest group included samples of flint artefacts from Margionys, Kalniškiai, Ožnugariai and Kretuonas and of some flint concretions from Grodno (Grandichi), Margionys and Sviliai.

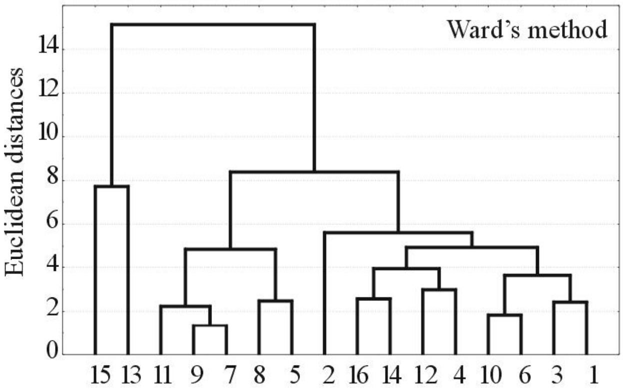

Fig. 6. Similarity of the groups of siliceous nodules, concretions and archaeological artefacts according to the average values of chemical elements determined by cluster analysis. *1* – Margionys (field concretions); *2* – Margionys (samples of one concretion); *3* – Margionys (flint artefacts); *4* – Jara (flint artefacts); *5* – Kretuonas (artefacts of silicified gaize); *6* – Kretuonas (flint artefacts); *7* – Dreniai (flint artefacts); *8* – Dreniai (artefacts of silicified gaize); *9* – Biržulis (flint artefacts); *10* – Ožnugariai (flint artefacts); *11* – Kalniškiai (artefacts of silicified gaize); *12* – Kalniškiai (flint artefacts); *13* – Šūkainiai and Jonikaičiai (gaize, silicified gaize); *14* – Grodno (Grandichi) (artefacts of one flint concretion); *15* – Nemunėlio Radviliškis (samples of the Upper Devonian siliceous nodules); *16* – Sviliai (samples of one Upper Devonian flint concretion)

6 pav. Silicitų gniutulų, konkrecijų ir archeologinių dirbinių grupių panašumas pagal klasterinės analizės pagrindu nustatytų cheminių elementų vidurkius.
1 – Margionys (konkrecijos iš lauko); *2* – Margionys (mėginiai iš vienos konkrecijos); *3* – Margionys (titnago dirbiniai); *4* – Jara (titnago dirbiniai); *5* – Kretuonas (silicifikuotos opokos dirbiniai); *6* – Kretuonas (titnago dirbiniai); *7* – Dreniai (titnago dirbiniai); *8* – Dreniai (silicifikuotos opokos dirbiniai); *9* – Biržulis (titnago dirbiniai); *10* – Ožnugariai (titnago dirbiniai); *11* – Kalniškiai (silicifikuotos opokos dirbiniai); *12* – Kalniškiai (titnago dirbiniai); *13* – Šūkainiai ir Jonikaičiai (opoka, silicifikuota opoka); *14* – Grodno (Grandiči) (mėginiai iš vienos titnago konkrecijos); *15* – Nemunėlio Radviliškis (mėginiai iš viršutinio devono silicito gniutulų); *16* – Sviliai (mėginiai iš vienos viršutinio devono titnago konkrecijos)

GENERALIZATION AND DISCUSSION

Comparison of trace element composition in the Phanerozoic rocks revealed that the median concentrations of many trace elements in the Devonian marls and domerites were smaller than in analogous rocks of other systems. Higher values of Cu were determined only in the Upper Devonian marls and of Zr in dolomites. This happened due to a higher input of terrigenous material into the then basin (Kadūnas et al., 2004). The concentrations of most trace elements, except Sr, in carbonaceous sediments of the Upper Cretaceous are even smaller. A reduction of allothigenous and allothigenous accessory trace elements (Ga, Ba, Zr, and Ti) is observed. Unfortunately, these data are not fit for comparison, because for different reasons some trace elements were eliminated from the study. A comparison of the background values of trace elements in the Upper Devonian dolomites and Upper Cretaceous carbonates (partly in marl) revealed considerably higher concentrations of Cr, Cu, Mn, Ti, and V in dolomites (Kadūnas et al., 2004). In the Upper Cretaceous aleurites, which were the source of gaize, the concentrations of Cr, Li, Ni, and V are higher than in dolomites. The concentration of Ti is comparable. However, analysis of flint artefacts and concretions did not prove an analogous pattern of element distribution in them. It is known that the siliceous nodules of the Upper Devonian Įstras Formation are found in the interlayers of dolomite powder, which are a product of gypsiferous dolomite washout (Водзинскас, 1966). Because of this, the chemical composition of siliceous rocks contained in them may have specific features of the enshrouding siltstone.

The flint concretions of Lithuania are associated with the Cretaceous carbonaceous flint outcrops and especially with the chalk and chalk marl blocks transported by glaciers and their washout residuals in South Lithuania. The *in situ* lying Cretaceous silicified gaize of West Lithuania, where glaciers exposed these rocks, was easily available for use (Radzevičius, 2004). The geological environment of Kretuonas and Jara ancient settlements is related with the Upper Devonian strata sometimes containing siliceous nodules and flint concretions. They are found in Middle and East Latvia, North Belarus and North Lithuania (Biržai district in particular).

As the chemical composition of siliceous rocks with regard to oxides composing rocks is rather comparable (only the concentration of SiO_2 ranges within 94–99%), the study of trace element composition was expected to be informative. The differences of discrete parts of nodules and concretions became especially evident after a cluster analysis of analytical data. The uneven distribution of trace elements (Ag, B, Cr, Cu, Li, Mn, Ni, P, Ti, and V) associated with the sparse insoluble residue in concretions (nodules) was predetermined as early as at the beginning of rock formation. Specific sedimentation conditions (salinity of the primary basin, silicified silt or rock, etc.) predetermined the chemical composition of concretions (nodules) recorded during analysis of archaeological artefacts.

A review of the data allows distinguishing the Nemunėlio Radviliškis, Šūkainiai and Jonikaičiai siliceous samples. Samples from Dreniai (B, Ca, Mg, and Ti), Jara (Ag, Cr, Mn, and Ni) and Kalniškiai (Ag, Ti) resemble them according to the groups of some chemical elements. The composition of flint and silicified gaize artefacts from Kretuonas, Dreniai and Kalniškiai ancient settlements sometimes contains smaller concentrations of B or Ag, Cr and Mg or higher concentrations of Cu, Fe, Mn, P, and Ti. According to the concentrations of these elements, the flint artefacts from Kalniškiai and Dreniai are comparable.

A comparison of some ancient artefacts with the average values of the geochemical composition of flint artefacts from the Margionys ancient settlement has shown that according to many chemical elements the artefacts from Margionys are most comparable with the finds of Kretuonas, Jara, Ožnugariai and Biržulis. The Nemunėlio Radviliškis, Jonikaičiai and Šūkainiai siliceous items differ from Margionys artefacts in most chemical elements. Samples from Kalniškiai and Dreniai are less different.

Data of correlation analysis show three types of CR. A stable group of elements [V–Ti–Fe], sometimes associated with Mg, Mn and Ca, characterizes the first type. Group [B–Ca–Mg], sometimes associated with P, Cu, and Cr, represents the second type. Group [Mg–Ca–Mn], sometimes associated with Fe, Cr and Ti, represents the third type. The CR of the first type are related with the Upper Cretaceous flint concretion and artefacts made of them (Margionys, partly Biržulis, Kalniškiai, Dreniai, and Kretuonas). The CR of the second type are characteristic only of Biržulis ancient settlements and are related with the Upper Cretaceous stratified flint (silicified gaize) widespread in Žemaitija. The CR of the third type are characteristic of the Upper Devonian siliceous items preserved in the distribution pattern of chemical elements of Kretuonas and Jara artefacts.

The average values of the geochemical data of 16 relatively homogeneous groups were generalized by the method of cluster analysis (Fig. 6). In three large clusters, the grouping of samples is simple. One group includes samples of silicified varieties of Šūkainiai and Jonikaičiai gaize, the second includes artefacts of silicified gaize from Kalniškiai, Dreniai and Kretuonas and flint artefacts from Biržulis and Dreniai, and the third includes only flint artefacts from Margionys, Kalniškiai, Ožnugariai and Kretuonas and some concretions from Grodno (Grandichi), Margionys and Sviliai.

The available data show that the siliceous artefacts studied may be made of raw material of triple genesis: the Upper Cretaceous concretions, the Upper Cretaceous stratified flint (silicified gaize) and the Upper Devonian siliceous rocks. The artefacts of Margionys ancient settlement *a priori* can be identified as made of the local flint concretions. However, the raw material of Biržulis artefacts is presumably of two types: the flint concretions imported from the south and the local stratified flint (silicified gaize). The higher petrographic and chemical diversity of Biržulis artefacts proves the existence of the latter type of material. It seems likely that the raw material of Kretuonas and Jara artefacts is also of two types. They are the Upper Cretaceous flint imported by the population of the Stone Age from the south south east and the relatively local Upper Devonian silicites. The distribution pattern of the latter in Lithuania has not been sufficiently well investigated. It is attributed to the spread of the Upper Devonian Frasnian stratified siliceous rocks, siliceous nodules and flint concretions in the Middle and East Latvia and North Belarus, comparable to the ones found in the environs of Nemunėlio Radviliškis and Sviliai settlements (Гравитис, 1963; Сорокин, 1963; Махнач, Гулис, 1993а; Махнач, Гуделис и др., 2004). Though very solid, the investigated and described siliceous nodules found in the environs of Nemunėlio Radviliškis Township have not yet been recognized among the archaeological artefacts.

CONCLUSIONS

1. The trace element composition of siliceous rocks (Ag, B, Cr, Cu, Li, Mn, Ni, P, Ti, and V) is mainly related with a sparse insoluble residue of carbonaceous siltstone (authigenous and allothigenous part). Their uneven distribution in siliceous concretions and nodules was predetermined in the beginning of rock formation.

2. The distribution patterns of chemical elements in flint concretions, siliceous nodules, rock debris and siliceous artefacts from ancient settlements allowed distinguishing a group of similar siliceous artefacts from the Nemunėlio Radviliškis, Šūkainiai and Jonikaičiai ancient settlements. The artefacts from the Dreniai, Jara and Kalniškiai settlements resemble them in some groups of elements.

3. Data of correlation analysis allow distinguishing three types of CR. The CR of the first type are related with the Upper Cretaceous flint concretions and artefacts made of them (Margionys, partly Biržulis, Kalniškiai, Dreniai, and Kretuonas). The CR of the second type are identified in the ancient settlements of Biržulis area alone and are related with the spread of the Upper Cretaceous flint (silicified gaize) in Žemaitija. The CR of the third type are presumably characteristic of the Upper Devonian siliceous rocks and are preserved in the distribution pattern of the chemical elements of Kretuonas and Jara archaeological artefacts.

4. According to data of cluster analysis, one group includes samples of Nemunėlio Radviliškis Devonian siliceous and Šūkainiai and Jonikaičiai gaize and its silicified varieties. The second group includes silicified gaize from Kalniškiai, Dreniai and Kretuonas and flint artefacts from Biržulis and Dreniai. The third, largest group includes samples of flint artefacts from Margionys, Kalniškiai, Ožnugariai and Kretuonas and of some flint concretions from Grodno, Margionys and Sviliai.

5. The artefacts, most likely, are made of the raw material of triple genesis: Upper Cretaceous concretions, Upper Cretaceous stratified flint and Upper Devonian siliceous rocks. The artefacts of Margionys ancient settlement can be identified as made of the local flint concretions. However, the raw material of Biržulis artefacts is presumably of two types: flint concretions imported from the south and the local stratified flint (silicified gaize). The raw material of Kretuonas and Jara artefacts is also of two genetic types: the imported Upper Cretaceous flint from the south south east and the relatively local Upper Devonian siliceous.

6. The distribution pattern of the latter in Lithuania has not been sufficiently well investigated. It is attributed to the spread of the Upper Devonian Frasnian stratified siliceous rocks, siliceous nodules and flint concretions in the Middle and East Latvia and North Belarus.

ACKNOWLEDGEMENTS

The authors thank the Lithuanian Science and Studies Foundation for support of the project (Reg. No T-05044), archaeologist Dr. Habil. A. Girininkas, geologists Dr. A. Radzevičius, Dr. R. Zinkutė and Dr. R. Taraškevičius for their aid in the realization of the planned work and consultation. The authors are also grateful to D. Ostrauskienė from the Lithuanian National Museum, B. Poškienė from the Geological Museum of the Institute of Geology and Geography, personnel of regional museums of Švenčionys, Panevėžys and Biržai towns and regional researcher V. Kučas for permission to use their collection material.

References

1. Bácskay E. 1984. Prehistoric flint mines (exploitation sites) in Hungary and their role in raw material supply. In *Third seminar on Petroarchaeology. Plovdiv, 27–30 August, 1984, Bulgaria*. 127–145.
2. Balcer B. 1988. The Neolithic Flint Industries in the Vistula and Odra Basins. *Przegląd Archeologiczny*. ***35***. 49–100.
3. Baltrūnas V. 1995. Pleistoceno stratigrafija ir koreliacija. Vilnius: Academia. 179 p.
4. Baltrūnas V. 2001. Paviršiaus geologinės sąlygos. Akmens amžius Pietų Lietuvoje (red. V. Baltrūnas). Vilnius. 82–89.
5. Baltrūnas V. 2002. Stratigraphical subdivision and correlation of Pleistocene deposits in Lithuania (methodical problems). Vilnius: Geologijos institutas. 74 p.
6. Baltrūnas V., Karmaza B., Kulbickas D., Pukelytė V. 2004. Mineralinės žaliavos bei jų paplitimas Virvytės, Minijos ir Varduvos aukštupiuose. *Acta Academiae Artium Vilnensis / Vilniaus dailės akademijos darbai. Dailė*. ***34***. 33–44.
7. Becker C. 1959. Flint mining in Neolithic Denmark. *Antiquity*. ***33***. 87–92.
8. Binsteiner A. 2004. Materialinterferenzen im Verbreiterungsgebiet bayrischer Jurahornsteine in Mittel- und Osteuropa. *Archäologisches Korrespondenzblatt*. ***34***. 169–175.
9. Biró K. 1997. Petroarchaeology of Siliceous Rocks and Source Determination in the Carpathian Basin. In *Siliceous rocks and culture. Aucheulian site of Arriaga (Spain)* (eds. A. Ramos-Millán and M. A. Bustillo). 353–359.
10. Guobytė R. 2001. Lietuvos geomorfologinis žemėlapis. *Geologijos akiračiai*. ***3***. 23–35.

11. Hoika J. 1986. Die Bedeutung des Oldenburger Grabens für Besiedlung und Verkehr im Neolithikum. *Offa*. ***43*** [= Festschr. A. Bantelmann]. 185–208.
12. http://www.flintsource.net
13. Jablonskytė-Rimantienė R. 1966. Paleolitinės titnago dirbtuvės Ežerynų kaime (Alytaus raj. Raitininkų apyl.). *Mokslų akademijos darbai. A serija*. ***2(21)***. 187–199.
14. Kaczanowska M. 1986. Kontakte zwischen der Ostslowakei und Kleinpolen angesichts der Importe von Steinrohstoffen. In *Urzeitliche und frühhistorische Besiedlung der Ostslowakei in Bezug zu den Nachbargebieten*. 77–83.
15. Kadūnas V., Katinas Vl., Radzevičius A., Taraškevičius R. 2004. Nuosėdų mikroelementinės sudėties kaita sedimentacijos paleobaseinuose ir geocheminių anomalijų susidarymas. *Lietuvos Žemės gelmių raida ir ištekliai* (ats. red. V. Baltrūnas). Žurnalo „Litosfera“ leidinys. Vilnius. 123–144.
16. Kobusiewicz M. 1997. Sources of flint on the West Polish Plain. In *Man and Flint* (eds. R. Schild and Z. Sulgostowska). Warszawa, Institute of Archaeology and Ethnology Polish Academy of Sciences. 83–90.
17. Konda J. 1986. The Mesozoic siliceous rocks of the Transdanubian Mid-Mountains. In *International Conference on Prehistoric Flint Mining and Lithic Raw Material Identification in the Carpathian Basin. Conference Proceedings* (ed. K. Biró). 165–168.
18. Kozłowski J. K., Manecki A., Rydlewski J., Valde-Nowak P., Wrzak J. 1981. Mineralogico-geochemical characteristic of radiolarites used in the Stone Age in Poland and Slovakia. *Acta Archaeologica Carpathica*. ***21***. 171–209.
19. Lietuvos geologija. 1994. Sud. A. Grigelis, V. Kadūnas. Vilnius: Mokslo ir enciklopedijų leidykla. 447 p.
20. Lietuvos TSR atlasas. 1981. Red. koleg. pirmininkas A. Drobnys. Maskva. 216 p.
21. Lietuvos Žemės gelmių raida ir ištekliai. 2004. Ats. red. V. Baltrūnas. Žurnalo „Litosfera“ leidinys. Vilnius. 700 p., 211 žemėl., CD.
22. Nielsen L. 1997. Raw material provenance in the early Neolithic. A comparative study of thinbutted flint axes from two regions in Jutland, Denmark. In *Man and Flint* (eds. R. Schild and Z. Sulgostowska). Warszawa, Institute of Archaeology and Ethnology Polish Academy of Sciences. 262–267.
23. Ostrauskas T. 2000. Tyrinėjimai Margionių titnago kasyklų ir dirbtuvių komplekse 1999 m. In *Archeologiniai tyrinėjimai Lietuvoje 1998 ir 1999 metais*. Vilnius. 50–51.
24. Rimantienė R. Akmens amžius Lietuvoje. Vilnius. 1984.
25. Quaternary deposits and neotectonics in the area of Pleistocene glaciations. May 12–16, 1997, Belarus. Excursions guide book (ed. A. Matveev). Minsk. 36 p.
26. Radzevičius A. 2004. Prekvartero stratigrafija ir uolienų litogenezė. Kreida. *Lietuvos Žemės gelmių raida ir ištekliai* (ats. red. V. Baltrūnas). Žurnalo „Litosfera“ leidinys. Vilnius. 90–97.
27. Skuodienė I., Katinas Vl. 1981. Nuo paleolito iki rankraštinių kultūros paminklų atsiradimo. In *Lietuvos TSR geologijos istorija* (sud. A. Grigelis). Vilnius. Mokslas. 4–18.
28. Šatavičius E. 2002. Titnago kasyklos ir apdirbimo dirbtuvės prie Titno ežero. *Archeologiniai tyrinėjimai Lietuvoje 2000 metais*. Vilnius. 22–24.
29. Археалогія и нумізматыка Беларусі. Энцыклапедыя. Мінск. 1993. 338–340.
30. Бруяко И. В., Видейко М. Ю., Сапожников И. В. 2005. Виктор Федорович Петрунь и его вклад в археоминералогию. *Археоминералогия и ранняя история минералогии*. Материалы международного семинара. Сыктывкар, Республика Коми, Россия, 30 мая–4 июня 2005 г. 17–22.
31. Водзинскас Э. В. 1966. Верхнедевонские доломиты северной Литвы. *Литология и геология полезных ископаемых Южной Прибалтики* (отв. ред. В. В. Микайла). Вильнюс: Минтис. 69–88.
32. Галибин В. А., Тимофеев В. И. 1993. Новый подход к разработке проблемы выявления источников кремневого сырья для культур каменного века Восточной Прибалтики. *Археологические Вести*. ***2***. Санкт-Петербург. 13–19.
33. Гравитис В. А. 1966. Силициты даугавской свиты. *Франские отложения Латвийской ССР*. Рига: Изд-во АН ЛатвССР. 243–262.
34. Калечиц Е. Г. 1984. Первоначальное заселение территории Белоруссии. Минск. 159 с.
35. Майорова Т. П., Волокитин А. В. 2005. Геохимические особенности кремня древних индустрий и выявление источников сырья. *Археоминералогия и ранняя история минералогии*. Материалы международного семинара. Сыктывкар, Республика Коми, Россия, 30 мая–4 июня 2005 г. 118–119.
36. Махнач А. А., Гулис Л. Ф. 1993a. Желваковые кремни в карбонатных отложениях девона и мела Беларуси. Сообщение 1. Петрография и минералогия. Литология и полезные ископаемые. ***1***. 49–63.
37. Махнач А. А., Гулис Л. Ф. 1993b. Желваковые кремни в карбонатных отложениях девона и мела Беларусии. Сообщение 2. Геохимия. *Литология и полезные ископаемые*. ***2***. 78–87.
38. Махнач А. А., Гулис Л. Ф. 1993c. Желваковые кремни в карбонатных отложениях девона и мела Беларуси. Сообщение 3. Генезис. *Литология и полезные ископаемые*. ***6***. 55–68.
39. Махнач А. А., Гулис Л. Ф., Махнач Н. А., Стрельцова Г. Д., Шиманович В. М., Михайлов Н. Д. 2004. Постседиментационные изменения отложений платформенного чехла. *Основы геологии Беларуси*. Минск. 215–233.
40. Петрунь В. Ф. 1971. О геологической позиции и обработанном кремне мезолитической стоянки Белолесье. *Материалы по археологии Северного Причерноморья*. Вып. *7*. Одесса. 110–117.
41. Селиванова Н. Б. 1984. Методика определения источников кремня для археологических памятников. *III Seminar on Petroarcheology*. Plovdiv. 93–102.
42. Синицына Г. В. 2005. Сырьё на финальнопалеолитических стоянках Валдая. *Археоминералогия и ранняя история минералогии*. Материалы международного семинара, Сыктывкар, Республика Коми, Россия, 30 мая–4 июня 2005 г. 115–116.

43. Сорокин В. С. 1963. Выделения аутигенного кремнезема в карбонатных породах даугавской свиты. *Франские отложения Латвийской ССР*. Рига: Изд-во АН ЛатвССР. 263–298.

Valentinas Baltrūnas, Bronislavas Karmaza, Dainius Kulbickas, Tomas Ostrauskas

SILICITINĖS UOLIENOS KAIP PRIEŠISTORINIŲ DIRBINIŲ ŽALIAVA LIETUVOJE

S a n t r a u k a

Atlikto darbo tikslas – nustatyti priešistorinių silicitinių dirbinių žaliavos paplitimo, sudėties ir genezės ypatybes bei galimas jos transportavimo kryptis Lietuvoje. Tam peržiūrėta archyvinė ir literatūrinė medžiaga silicitų sudėties, genezės ir paplitimo Lietuvos, Latvijos ir Baltarusijos ikikvarterinių uolienų sluoksniuose klausimais. Peržiūrėti ir petrografiškai apibūdinti gerai ištirti akmens amžiaus senovės gyvenviečių titnaginiai dirbiniai Lietuvos nacionalinio muziejaus, kitų muziejų fonduose, paimti ėminiai laboratoriniams tyrimams, taip pat ir iš atodangų bei karjerų. Siekiant geocheminiu požiūriu apibūdinti silicitus ir išryškinti skirtumus, buvo nustatomas kai kurių makro- ir mikroelementų pasiskirstymas įvairiose jų atmainose (konkrecijų titnage, sluoksniniame titnage – silicifikuotoje opokoje), taip pat archeologiniuose titnago dirbiniuose naudojant daugiaelementinę (grupinę) emisinę spektrofotometrinę analizę.

Statistinis bei grafinis duomenų apibendrinimas leido padaryti išvadą, kad tirtieji silicitiniai dirbiniai gali priklausyti trijų tipų žaliavai: viršutinės kreidos titnago konkrecijoms, viršutinės kreidos sluoksniniam silicitui (silicifikuotai opokai) ir viršutinio devono silicitams. Jeigu Margionių (Pietų Lietuva) senovės gyvenvietės dirbinius galima priskirti vietiniam ledynų perklostytam konkrecijų titnagui, tai Biržulio ploto (Šiaurės Vakarų Lietuva) senovės gyvenviečių dirbinių žaliava greičiausiai yra dviejų tipų – tai gyventojų atneštinis iš Pietų Lietuvos konkrecijų titnagas ir vietinis sluoksninis silicitas (silicifikuota opoka). Kretuono ir Jaros (Rytų Lietuva) senovės gyvenviečių dirbinių žaliava, atrodo, taip pat priklauso dviem genetiniams žaliavos tipams: atneštiniam iš pietinių Lietuvos rajonų viršutinės kreidos konkrecijų titnagui bei vietiniams viršutinio devono silicitams. Pastarojo tipo žaliavos paplitimas Lietuvoje nėra pakankamai ištirtas ir daugiausia grindžiamas viršutinio devono franio aukšto sluoksninio silicito, silicito gniutulų ir titnago konkrecijų, panašių į aptiktus prie Nemunėlio Radviliškio ir Svilių, paplitimu Vidurio ir Rytų Latvijoje bei Šiaurės Baltarusijoje. Prie Nemunėlio Radviliškio aprašytieji ir tyrinėtieji silicito gniutulai, nors ir pasižymi dideliu kietumu, tačiau tarp archeologinių dirbinių kol kas neatpažinti.

Валентинас Балтрунас, Брониславас Кармаза, Дайнюс Кульбицкас, Томас Остраускас

СИЛИЦИТОВЫЕ ПОРОДЫ КАК СЫРЬЕ ДЛЯ ДОИСТОРИЧЕСКИХ ИЗДЕЛИЙ В ЛИТВЕ

Р е з ю м е

Цель настоящей работы – выявить особенности распространения, состава и генезиса сырья доисторических силицитовых изделий, а также направления их транспортировки в Литве. Изучены архивные и литературные материалы по составу, генезису и распространению силицитов в дочетвертичных отложениях Литвы, Латвии и Беларуси. Осуществлены осмотр и петрографическое определение силицитовых (кремневых) изделий из хорошо изученных древних поселений каменного века в фондах Литовского национального музея, других музеев; отобраны пробы для лабораторных исследований. Отбор проб осуществлен также из обнажений и карьеров. Для геохимической характеристики и выявления различий силицитов определялось распределение некоторых макро- и микроэлементов в отдельных их разновидностях (конкрециях кремня, окремненных силицитовых прослоев – силицитизированной опоки), а также в археологических силицитовых изделиях путем применения многоэлементного (группового) эмиссионного спектрофотометрического анализа.

Статистическое и графическое обобщение полученных данных позволили сделать вывод о том, что изученные силицитовые изделия могут быть отнесены к сырью трех типов: кремневые конкреции верхнего мела, окремненные (силицитизированные) прослои верхнего мела и силициты верхнего девона. Если сырьем изделий древнего поселения Маргенис (Южная Литва) являются переотложенные ледниками местные кремневые конкреции, то сырье окрестностей оз. Биржулис (Северо-Западная Литва) скорее всего является двоякого происхождения: принесенный из Южной Литвы кремень верхнемеловых конкреций и местный из окремненных прослоев опоки верхнего мела. Сырье изделий древних поселений на участках приозерья Кретуонас и ручья Яра (Восточная Литва), по-видимому, также можно отнести к двум генетическим типам: принесенному из южных районов Литвы кремневых конкреций верхнего мела и местным девонским силицитам. Распространение сырья последнего типа на территории Литвы изучено недостаточно и в большой мере обосновывается распространением силицитовых прослоев и желваков, а также кремневых конкреций франского яруса верхнего девона в Средней и Восточной Латвии и Северной Беларуси. Изученные и описанные верхнедевонские силицитовые желваки в обнажении вблизи мест. Нямунелио Радвилишкис (Северная Литва), хотя и характеризуются большой твердостью, среди археологических изделий пока не выявлены.