THE USE OF OCHRE FOR POTTERY DECORATION IN THE MIDDLE NEOLITHIC AT VĂDASTRA

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The aim of this paper is to present on the basis of some analytic arguments the red ochre problems in the Middle Neolithic at Vădastra relating to the raw mineral sources and the details of the ochre application on ceramics.

At the same time, hand in hand with the progress of archaeological researching methods, the coloured materials discovered in the settlements and tombs, particularly the red ochre, have called the attention of archaeologists and have been subject of their investigation.

The red and yellow ochre and other coloured materials have been reported in numerous Aurignacian settlements and tombs related to parietal and corporal painting, and possibly, as we believe, also to tattoo. At Grimaldi, crouched skeletons painted with red ochre belonging to the Cro-Magnon type have been known since 1874.

The Roche site presented, according to the analyses made at the beginning of the 20th century, seventeen samples of colouring matters: ochre with nuances from red to yellow and pigments with shades from brown to black.

The dye stuffs were prepared by pounding and decantation from the cavities (godets) and were preserved in reindeer horn cases discovered in France as early as the last decades of the 19th century.

As for the Solutrean and Magdalenian ages, the ochre detections are very numerous. In the Noailles grotte many ochre fragments wet in water had seven hues from red to yellow. These materials can be found in the neighborhood in a natural state.

During the Mesolithic and the Tardenoisian ages, particularly in the Neolithic, ochre was very much used. In the last decades of the 19th century in France, Spain, as well as in the Central and Eastern Europe tombs and necropolises were discovered, in which red and yellow ochre was found. In settlements the ochre was discovered under the form of sticks, whereas in tombs red colour was applied on the head or on other parts of the body. Often some ochre fragments were situated not far from the skull. An outcrop is known where ochre was exploited at Cornelia in Dordogne.

At the same time with the discoveries made in Western Europe and in other European countries, in our country the first testimonies of the use of ochre and coloured pigments were releaved. In 1870, at the Piscul Crăsani settlement excavation, Dimitrie C. Butculescu pointed out the presence of a piece of cinnabar found at 2.30 m depth (in the Neolithic stratum). He also mentioned in a posthumous paper the red ochre found in this excavation.[1]

A set of scientific bases beginning with Ioan Andrieşescu's excavation, developed especially after the first world war. In the neolithic settlements in Câmpia Română stone evidence was found of the use of

[1] D. C. Butculescu, *Localităţile explorate de Dim. C. Butculescu de la 1867 la 1895*, Musée d'Histoire de Bucarest, 1902, I, p. 57.

ochre, particularly remains on ceramics. Some explorers mentioned ochre, while others only named it simply "red matter" but more often ochre passed unnoticed under the calcareous crust as the ceramic materials were not treated in the laboratory in order to remove the crust.

In the excavations at Vădastra and also in other old time settlements in the Lower Danube region numerous remains of ochre raising problems such as the origin and the utilisation technique, were discovered in the strata and on ceramics. The physico-chemical analyses started by Em. Protopopescu-Pache lacked comparative material with reliable statigraphy in other settlements. Interrupted because of his death (1967), the analyses were resumed after a decade and the results are presented in this paper.

The Vădastra settlement is situated on a hill fragment of a Danube middle terrace known by the natives under the name of "Dealul Cişmelei" located at 14 km north-northwest from Corabia. The new excavations were started in 1946 by Corneliu N. Mateescu[3] and began with a ditch setting out from the center of "Măgura Fetelor" southwards. Then, a larger surface was dug (10 × 7 m) in the northern edge of the ditch end.[2]

In the following years the diggings were led from the northern end of the ditch to the north up the "Obârşia" brook. Afterwards, in 1962, from the center of the hillock to the west, the investigation of the settlement went on towards the four cardinal points.

At the bottom of the archaeological strata there is a Palaeolithic layer (prolonged middle Aurignacian) built up under a climate with sylvo-steppe vegetation under some 400-500 mm rainfalls and an average temperature of 10-11°C. Above the Palaeolithic layer an intermediate layer without archaeological remains is superposed; built up under a climate drier and colder than the Palaeolithic layer formation.[3]

Together with the arrival of an optimum climate in the Lower Danube region, we ascertain a neolithic population about the end of the sixth millennium and the biginning of the fifth, on "Măgura Fetelor". The layers Vădastra I and Vădastra II (middle Neolithic) are built up under a climate with a higher temperature and humidity than during the Palaeolithic layer formation.[4]

About the end of the fifth millennium the temperature becoming a little colder and the humidity lower, the population of the Vădastra II phase left the settlement while the sites were occupated by bearers of the Sălcuţa culture (late Neolithic).

The post-Sălcuţa layers werw washed away by rainfalls and agricultural works but they are recorded by the tombs of the Coţofeni civilization (transition from the Neolithic to the bronze age) and 14th century tombs, corn pits, cots and other archaeological material.

Materials and analytical methods

Among the Neolithic ceramics discovered at Vădastra, fragments with ornamental red belonging to the Starčevo-Criş, Vădastra and Sălcuţa cultures were investigated. Comparative samples of red application were analysed on the Starčevo-Criş fragments at Schela Cladovei,[5] Cârcea,[6] Şimnic[7] and Dulceanca[8] (Fig 1).

Ornamental red samples were also analysed on the Vădastra pottery at Crusovu[9] and Hotărani[10] and on other ceramics from the Lower Danube area: Boian at Vidra, Aldeni, Spantov and Radovanu,[11]

[2] Corneliu N. Mateescu, *Săpături arheologice la Vădastra*, Materiale, 5, 1959, p. 2.

[3] Em. Protopopescu-Pake, Corneliu Mateescu, Al. V. Grossu, *Formation des couches de civilisation de la station de Vădastra en raport avec le sol, la faune malacologique et le climat*, Quatar, 20, 1969, p. 139.

[4] Arlette Leroi Gourhan, Cornelius N. Mateesco, Em. Protopopescu-Pake, *Contribution à l'étude de la station de Vădastra du Paléolithique supérieur à la fin du Néolithique*, Bulletin de l'Association française pour l'étude du Quaternaire, 4,1967, p. 274-277.

[5] Ceramic material offered by V. Boroneant, diggings performed in 1971. The authors are very grateful to all the archaeologists who kindly placed at their disposal the ceramic materials from their diggings.

[6] Ceramic material offered by Marin Nica, diggings performed in 1971-1975. The authors are very grateful to all the archaeologists who kindly placed at their disposal the ceramic materials from their diggings.

[7] Materials from diggings performed in 1967-1971 by Doina Galbenu.

[8] Ceramic materials offered by Eugen Comşa from Suzana Dolinescu Ferche's diggings.

[9] Corneliu N. Mateescu, *Săpături arheologice la Cruşovu*, Materiale, 3, 1957, p. 105-109.

[10] Ceramic material offered by Marin Nica from his diggings in 1972.

[11] Ceramic materials offered by Eugen Comşa from the diggings in 1973 and 1980.

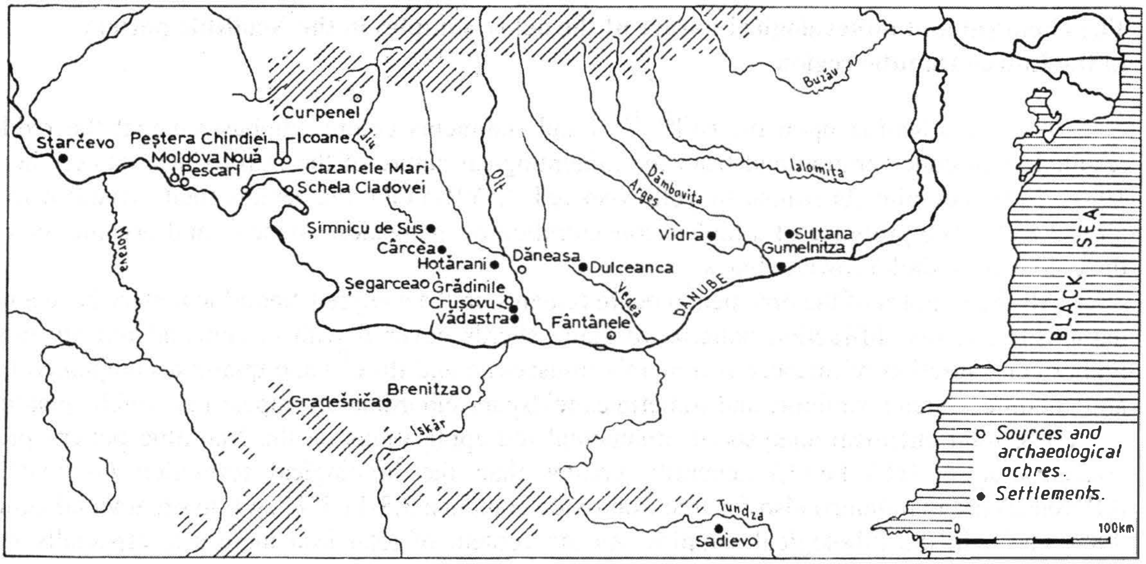

Fig. 1. Settlements of the Lower Danube area referred to in the text.

Gumelniţa at Sultana[12], Vlădiceasa[13] and the eponymous settlement[14] as well as other contemporaneous cultures in the northern part of Bulgaria at Gradesnitza and Brenitza.[15]

In order to specify the probable sources of ornamental ochre, ochre archeological samples of the Epipalaeolithic were investigated at Peştera Chindiei, Icoane Mehedinţi[16] and Octrovul Mare[17] as well as of the Starčevo-Criş culture at Cuina Turcului[18] and Cârcea,[19] of the Vădastra culture at Vădastra[20] and of Gumelniţa culture at Sultana[21] and Gumelniţa[22]. In addition, ochre samples were analysed at Sadievo Nova Zagora[23], in a Coţofeni tomb at Vădastra[24] and from a stick of La Tène ochre at Fântânele Zimnicea.[25]

As presumtive sources samples of terra rossa were analysed (Cazanele Mari, Moldovita-Banat), as well as red rendzinas (Pescari, Valea Marcoviei), loams (Grădinile-Romanaţi, Dăneasa-Olt and Roşiorii de Vede) loams and clays with reddish hue (Curpenelul-Gorj, Şimnic, Podari and Segarcea-Dolj and Vădastra-Malul Roşu).

All the samples were analytically characterised taking into consideration their content of total[26] and free iron,[27] their infra-red absorption spectra[28] and their X-ray diffraction patterns.[29]

The nuances of the colours were estimated by means of Munsell tables[30] and their redness rate was calculated by the equation RR = (10-R) C/V where RR is the redness rate, R – hue, C – chrome and V – the colour value in the Munsell tables.[31]

[12] Ceramic material from diggings by Eugen Comşa in 1976.

[13] Diggings by Done Şerbănescu in 1978.

[14] Ceramic materials offered by Vladimir Dumitrescu and Barbu Ionescu from their diggings in 1963.

[15] Ceramic materials offered by Bogdan Nicolov from his diggings in 1974.

[16] Diggins of V. Bernet in 1967-1969.

[17] Diggings of V. Boroneanţ in 1979

[18] Diggings of V. Bernet in 1968.

[19] Diggings of Marin Nica in 1975.

[20] Diggings of Corneliu N. Mateescu in 1946.

[21] Diggings of Eugen Comşa in 1975.

[22] Diggings of Vladimir Dumitrescu in 1963.

[23] Material offered by Mitic Cancev from his diggings.

[24] Gh. Gâţă, Corneliu N. Mateescu, *On the provenance of the red ochre from the tomb with ritually cut skeleton of Vădastra (Coţofeni Civilisation)*, Dacia, N.S., 27, 1-2, 1983, p. 7.

[25] Stick of La Tène red ochre offered by Barbu Ionescu.

[26] The total iron, expressed as % Fe_2O_3 was brought into solution with fluorhydric and perchloric acids and was determined by means of atomic absorption. M. L. Jackson, *Soil chemical analyais*, New York, Constable, 1958, p. 283.

[27] The free sesquioxides (non bonded as silicates) were dissolved by electrochemical reduction. Elena Gâţă, Gh. Gâţă, *Séparation et détermination des sesquioxides libres des sols et des sédiments. 4th International Congress of Soil Sciences*, Bucharest, 1964, III, p. 50.

[28] The infra-red spectra were obtained by potassium bromide disk technique. Gh. Gâţă, *Determinarea continutului de minerale din grupa bioxidului de siliciu prezente în soluri*, Analele Institutului de Cercetări pentru Pedologie şi Agrochimie, 48, 1987, p. 55-71.

[29] X-ray diffraction patterns were made directly on the sherds. Gh. Gâţă, *Contribuţii la studiul metodelor de determinare a structurii şi compoziţiei chimice a mineralelor argiloase din soluri şi sedimente*, Ph.D.Thesis, Polytechnic Institute Bucharest, 1972, p. 93.

[30] Munsell scale is a colour code adopted by seventh approximation of Soil Classification in the U.S.A. to reduce subjectivism in assessing the colour of a soil. A code of figures and letters is used for colours. For exemple, the *terra rossa* at Cazanele Mari has the colour 10 R 4/6.

[31] J. Torrent, U. Schwertmann, D. G. Schulze, *Iron oxide mineralogy of some soils of two river terrace sequences in Spain*, Geoderma, 23, 1980, p. 191.

The chemical and mineralogical nature of the ochre samples in the Neolithic pottery in the Lower Danube region

The investigation upon the soil[32, 33, 34] and sediments colour emphasized that the reddish colour resulted from concentration and from the mineralogical nature of the iron oxides, namely hematite (red 10R-2.5YR, goethite (brownish to yellowish red 2.5YR-5YR) and lepidocrocite (reddish to yellowish 5YR-7.5YR. The presence of a higher concentration of manganese oxides[35] and organic matter changes the colour from dark brown to black.

All the samples of the ornamental ochre remains on the analysed Neolithic pottery have a mineralogic nature. The X-ray diffraction patterns of some sherds covered with ornamental red always present a higher concentration of micaceous minerals (muscovite and illite[36]) and quartz accompanied by hematite and goethite. Calcite, kaolinite and sometimes feldspars subordinately appear in variable quantities.

The total chemical analysis of ornamental red applications on the Neolithic pottery presents iron concentraion (7-16% Fe_2O_3) generally greater than the lithological formation (4-15%Fe_2O_3). The differences are maintained also for free iron oxide contents: 3.8-11.1%Fe_2O_3 in ornamental ochre and 0.7-9.8% Fe_2O_3 in the lithological samples. As the content of total iron increases, especially of free iron oxide, the red colour or the samples is more intense.

All these analytical data indicate that the ochre on the very old pottery is a loam or a clay with considerable content of iron oxides. Therefore the sources of ornamental red must be sought in the lithological formation with such a colour and with such a particle size.

The alteration of ornamental red in the archaeological strata

The high fluctuation of the red colour nuance on the ceramics in each settlement and culture and the partial or total wiping of the pigments raise the problem of the causes which conduct to the eventual changes of the colour nuance and even to the removing of the ornamental application.

It is sure that the vessels were thrown off and broken after their use. It is also possible that the ornamental red was wiped partially or even entirely during the vessel utilisation, especially if the decoration was made with the raw ochre less adherent or with organic dye stuff. This asumption would explain the presence on some rim sherds at Vădastra I of some very small red ochre spots placed under the rim.

During the burial into the archaeological layers, for several millennia, the ceramic fragments were in direct contact with the soil solution permanently regenerated by infiltration of rainfalls which also brought some components from the descomposition of domestic remainders, animal dejection, hearth ash, a.s.o. The soil solution could dissolve or at least modify to a small or a large extent the iron and manganesse oxides of the ornamental ochre.

In the sylvo-steppe climate at Vădastra[37] with relatively weak percolation, the soil evoluated to a haplic or luvic phaeozem (udic haptustoll or argiuztoll in a seventh approximation, U.S.A.) with weak textural differentiation. In these soils in the settlement neighbourhood at Vădastra the carbonate accumulation horizon lies at about 0.85-1 m depth and in the ploughing horizon the pH does not take down under 6.6 value although chemical fertilizers which acidify the soil were frequently used.

Even more, the base saturation degree always exceeds 90%. This remainder of pH and base saturation degree[38] at high values in the soil horizons of the settlement neighbourhood is due to the eolian

[32] U. Schwertmann, W. Lentze, *Bodenfarbe und eisenoxidesform*, Zeitschrift fur Pflazenerrnahfung, Dugung und Bodenkunde, 115, 1966, 3, p. 210.

[33] C. Crăciun, V. Bălăceanu, Elisabeta Marian, Aurelia Meghea, Lucia Vlad, *Aspecte mineralogice la nivelul fractiunii coloidale din unele soluri cu caracter rodic*, Publicaţiile Societăţii Naţionale Române de Ştiinţa Solului, 23 B, 1987, p. 60.

[34] C. Crăciun, Adriana Grigorescu, Aurelia Meghea, Lucia Vlad, C. Curelaru, N. Florea, *Fierul III în fracţiunea argiloasă a unor soluri rodice şi implicaţile lui cromatice la nivelul acestei fracţiuni*, Publ. SNRSS, 1989, 25 A, 1989, p. 164.

[35] U. Schwertmann, R. M. Taylor, *Iron oxides*, Mineral in Soil Environments. SSSA Book Series 1, p. 422.

[36] Em. Protopopescu-Pake, Cornelius N. Mateescu, Al. V. Grossu, *op.cit.*, 147.

[37] Em. Protopopescu-Pake, Cornelius N. Mateescu, Al. V.. Grossu, *op.cit.*, 147.

[38] Gh. Blaga, I. Rusu, S. Udrescu, D. Vasile, *Pedologie*, 1996, p. 144.

contribution with about 2% $CaCO_3$. The estimation indicates that in this region a material bed with a thickness of about 2-3 cm/century settled in average from the Neolithic time till today.

In addition, the layers of the archaeological profile have a large quantity of carbonate (6-30%$CaCO_3$)[39] due to the horizon mixing as a result of the excavation of pit huts, grain pits and adobe[40]. In these conditions the anthropic contribution (domestic remainders, dejection and hearth ash) accentuated to a little extent the ornamental red alteration as results from the diffusion (1-2 mm) of ochre into the sherd walls and from the very rare spots of natural iron oxides deposited on some ceramic fragments. Moreover, natural ochre bits from Coţofeni tombs discovered at Vădastra[41], were a little dispersed in the soil (only 1-1.5 mm).

In conclusion, we may say that under the circumstances of the cultural layers at Vădastra, the ochre applied before firing remained practically unchanged while the ochre applied after firing could undergo small changes of colours and also partial wiping.

The decoration with ochre of the pottery at Vădastra

The investigated archaeological materials indicate that the ochre ornamentation was used during the whole Neolithic in the Lower Danube area within the Starčevo-Criş, Vădastra, Boian, Sălcuţa and Gumelniţa cultures.

As for the Starčevo-Criş culture, the ornamental red was used as an application covering the vessel walls, as a background or as a painted colour. In the settlement at Vădastra only a few body fragments of small vessel with red application were discovered, but in a neighbouring settlement, at Linia Mare at Vădastra, vessel fragments with red application were found, as well as pots with red background painted with black-brown.

The red colour seems to have a weaker extent in the Vădastra I ceramics decoration. Among the numerous fragments of the Vădastra I vessels discovered in the settlement, only a few vessel rims and some pot bodies as well as figurines have application with red chrome.

In the Vădastra II eponymous settlement, the ornamental red frequently covers the pot rim, the polished parts which are not included in the decoration itself, obtained by incision and filled with white, the strokes dividing the ornamental areas and borders which frame the decorations (Fig 2). Ornamental red is less frequent on the Sălcuţa pottery in the settlement and appear on the rim of some cups belonging to this culture.

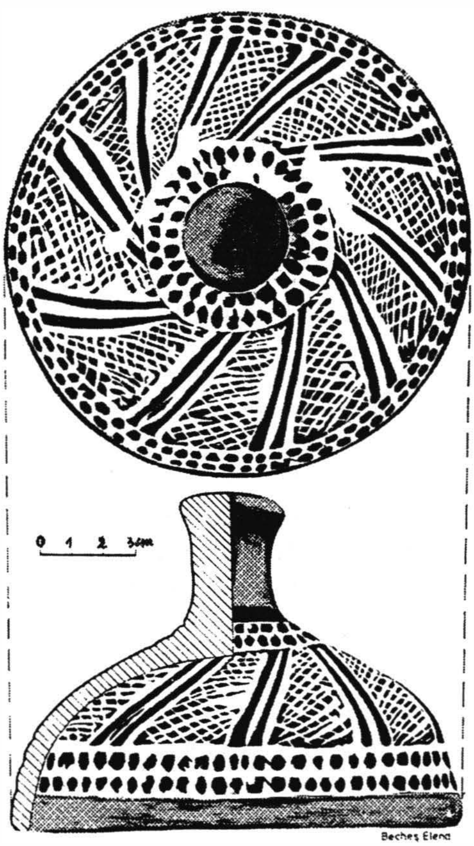

Fig. 2. Vădastra. Lid of ornamental vessel decorated by incision and filling with white chalky paste; red colour (screed area) on rim and shoulder (Vădastra II phase).

[39] Em. Protopopescu-Pake, Cornelius N. Mateescu, Al. V. Grossu, *op.cit.*, p. 154-156.

[40] *Ibidem*, p. 140-141.

[41] Gh. Gâţă, Corneliu Mateescu, *op.cit.*, p. 8.

The nuances of the ornamental red on the pottery at Vădastra

Not all the ochre application left on the Neolithic ceramics at Vădastra can be investigated analytically. In some cases the quantity on the sherd is much too small, in other cases the colour detaching includes a great part of the ceramic mass and there are cases when the natural ochre pellicles are visibly corroded with chrome similar to the oxido-reduction spots on the soil aggregates. The samples obviously alterated are not taken into consideration because they do not represent the red application made by Neolithic artisans but only an alteration.

In order to include in the research, as much as possible, all the ochre remainders in small quantities on the walls of the pots and on figurines, all the unaltered ochre applications were investigated at the beginning by the colour estimation and the calculation of the redness rate.

Among the few fragments of Starčevo-Criş pots discovered at the Vădastra settlement three had a red layer (10R4/6-10R4/8) which had been allplied before firing.

On the ceramics of the Vădastra culture, ochre was applied before and after firing. In the Vădastra I phase, 16 ochre nuances appear on the pottery, from red[42] to reddish brown,[43] reddish yellow,[44] brown[45] and very rarely light grayish brown.[46] In the Vădastra II phase 22 ochre nuances appear from red[47] to yellowish red,[48] reddish brown[49] and very rarely brown.[50]

From the comparation of the ochre nuance on the ceramics of the two phases at Vădastra it results that in the Vădastra II phase the preference for the red hue is accentuated. Consequently we may conclude that the ochre sources were more numerous and permitted the colour choice or the exchanges with ochre and raw materials for more developed preparation as well as the techniques for ochre processing. The potters from the settlement had good knowledge about the evolution of the ochre colour during the firing.[51] The scanty use of ochre applied after firing and the preference of ochre painted before firing in the Vădastra II phase indicate the evolution of the application methods of ochre on ceramics and prove that the Neolithic potters had noticed the better adhesion of ochre paint before firing.

In the late Neolithic at Vădastra on Sălcuţa ceramics only ochre with red hue appears.[52]

The distribution of the ornamental ochre on the Neolithic pottery at Vădastra as a function of the colour expressed by the redness rate appears to be bimodal and asymmetrical (fig. 3). The chief maximum appears at the value of 16 RR and a twice second maximum lies in the interval 3-4 RR. The colour of some samples with a redness rate under 6 is due to a thin ochre layer which does not succeed in covering the colour of the pot walls. These thin pellicles are due to the ochre application manner on ceramics in a thin layer or to the superficial dissolution of the application during the millennia while the sherds were buried in the archaeological stratum.

As results from the histogram in Fig. 3 about 21.6% from the investigated samples appear on the Vadastra I ceramics and 78.4% on the pottery of Vădastra II phase.

Ochre painted after firing appears on 32% of the applications of Vădastra I phase and only 6% of the Vădastra II ceramics. Thus the more frequent use of ochre painted after firing on the pottery Vădastra I phase and its more rare use on Vădastra II ceramics are also statistically confirmed.

On a Vădastra II pot rim unfired ochre was painted on fired ochre, probably to correct the application shortcomings. The chrome of unfired ochre (5YR 5/6) is lighter than that of fired ochre (2.5YR 4/6) and suggests a correction with the same colour suspension immediately after firing and cooling of the pot. It is confirmed in this way the accentuation of the red hue during ceramics firing due to the forming of hematite from the goethite of the sedimentary sources.[53]

[42] Munsell colours 10R 4/3-4/4-4/6-5/6; 2.5 YR 4/6-4/8-5/6.

[43] Munsell colours 2.5 YR 4/4; 5 YR 4/3-5/3-6/4.

[44] Munsell colour 5 YR 7/6.

[45] Munsell colours 7.5 YR 5/4-5/6-6/4.

[46] Munsell colour 10 YR 6/2.

[47] Munsell colours 10 R 3/6-4/4-4/6-4/8-5/6-5/8; 2.5 YR 4/2-4/6-4/8-5/2-5/6-5/8-6/6.

[48] Munsell colours 5 YR 4/8-5/6-5/8-6/6.

[49] Munsell colours 5 YR 5/3-5/4-6/4.

[50] Munsell colours 5 YR 5/3-5/4-6/4.

[51] During the firing the ochre colour becomes more reddish due to a gradual transformation of goethite into hematite. R. C. Mackenzie, *Differential thermal investigation of clay*, Central Press, 1957, p. 300.

[52] Munsell colours 2.5 YR 4/6-5/6.

[53] R. C. Mackenzie, *op.cit.*, p. 301.

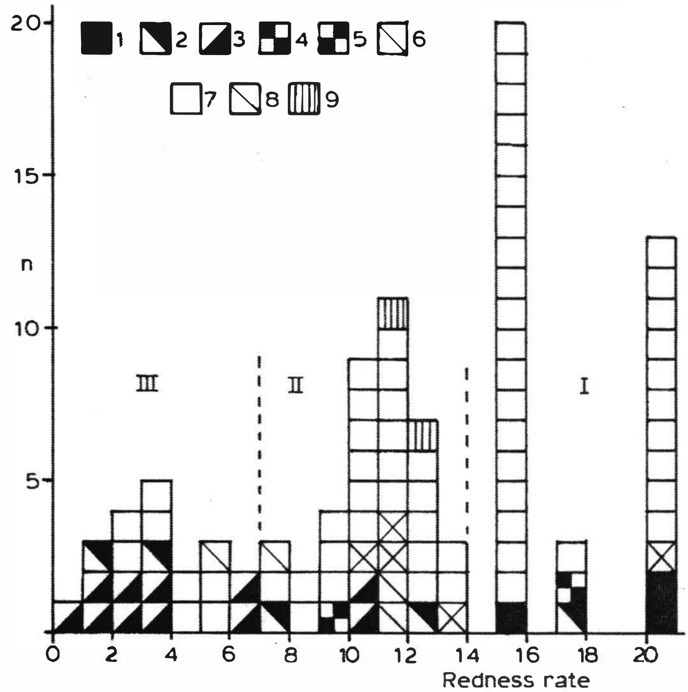

Fig. 3. Distribution of the redness rate (RR) in ornamental ochre on the sherds at the Vădastra settlement. Starčevo-Criş: vessels – 1; Vădastra I vessels after firing – 2; before firing – 3; Vădastra I figurines: after firing – 4; before firing – 5; Vădastra II vessels: after firing – 6; before firing – 7; Vădastra II figurines before firing– 8; Sălcuţa cups, before firing– 9.

The distribution of the application according to the redness rate indicates that we may conventionally separate three types of ochre: type I, red (RR>14), type II, dark red (RR between 14 and 7) and type III, reddish-brown (RR<7). The three types of ochre appear on the Vădastra I and Vădastra II ceramics in proportion of 16%, 26%, 50% and 42%, 46% ,12%, respectively in the Vădastra settlement. The small number of ochre samples on the Starčevo-Criş and Sălcuţa pottery at Vădastra does not allow to estimate the repartition of different ochre types in these cultures at Vădastra.

The ochres remains on the Vădastra figurines at Vădastra have always a dark-red (type II) or a red (type I) hue and show the high care for figurine decoration which could suggest a ritual value of the ornamental red.

In order to know the ornamental ochre hue on the ceramics of other cultures and to have a general image of the chrome repartition on the whole Neolithic pottery in the Lower Danube areas some ochre samples on Starčevo-Criş ceramics were investigated at Schela Cladovei, Cârcea, Şimnic, Vădastra – Linia Mare point and Dulceanca, on Boian pottery at Vidra and Aldeni, on pottery of Boian-Gumelniţa transition at Spantov and Radovanu, on Gumelniţa ceramics at Sultana, Gumelniţa and Vlădiceasa and on the pots from Northern Bulgaria at Gradesnitza and Brenitza.

The distribution of ornamental ochre on the Starčevo-Criş ceramics due to the redness rate (Fig. 4) emphasised the presence of the three chrome types already specified in the middle pottery at Vădastra (Fig. 3). As compared to the Vădastra histogram for the Starčevo-Criş samples, the red hue (type I) has the higher frequency at 16 RR, too. The maximum frequency with the red hue (type II) is placed in the interval 10-11 RR and that of the reddish-brown ochre samples (type III) in the interval 0-1 RR. The better accentuated shift of the redness value accompanied by an increased symmetry of the chrome is due to the inclusion of all the application samples with dark brown colours which appear on the fine ceramics of this culture.

In the middle and late Neolithic ages the three colour types are also utilised (Fig. 5).The distribution of the three ochre types of the samples belonging to different cultures in the Lower Danube area is similar to that of the Vădastra Neolithic and presents the same maxima as the samples of the Starčevo-Criş culture (Fig. 4). We may conclude that during the whole Neolithic age in the Lower Danube region, although the red hues were more and more numerous, the same ochre types originated in red loams and clays were used. The increasing hue numbers and the preserving of the ornamental ochre types show the special extension and the growth of the ochre exchange ways with ochre and raw materials for decorative colours while maintaining the same types of sources.

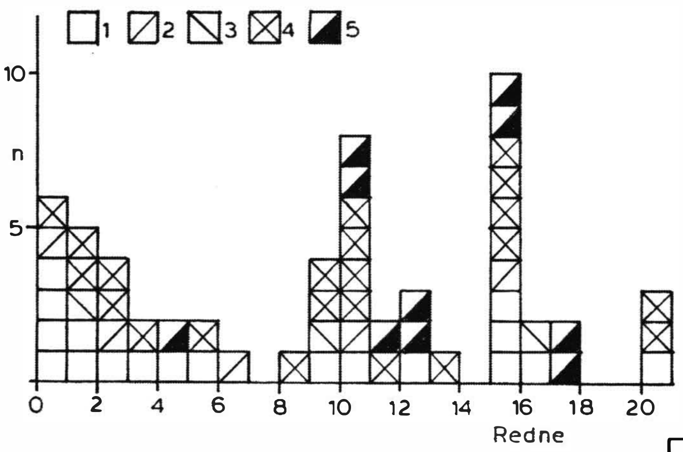

Fig. 4. Distribution of the redness rate in ornamental ochre on the Starčevo-Criş pot sherds: Şimnic-1, Schela Cladovei-2, Cârcea-3, Vădastra Linia Mare-4, Dulceanca-5.

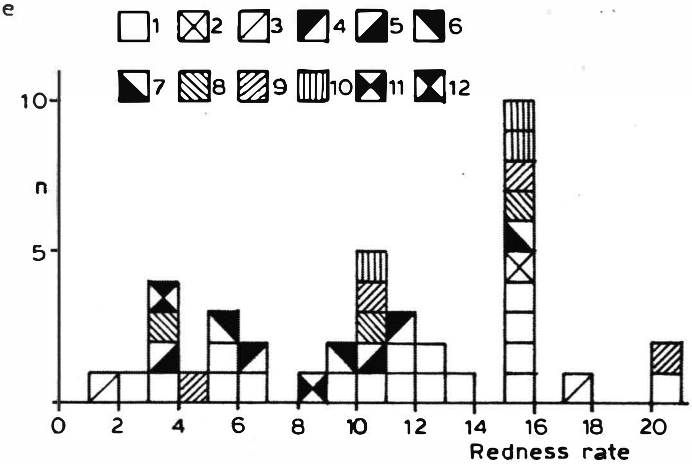

Fig. 5. Distribution of the redness rate in the ornamental ochre on the sherds: Vădastra culture at Cruşovu (vessels-1, figurines-2) and Hotăreni pots-3; Boian vessels at Vidra-4 and Aldeni-5; transition at Spanţov-6 and Radovanu-7; Gumelniţa pots at Sultana-8, Gumelniţa-9 and Vlădiceasa-10; north of Bulgaria at Gradesniţa-11 and Breniţa-12.

The ornamental ochre sources in the in the Neolithic age at Lower Danube

The Neolithic potters in the Lower Danube region selected their ornamental ochre and raw materials from natural loams and clays. From the epipalaeolithic period, in the tombs or in the archaeological layers, natural clods of ochre with different colour spots were discovered (Peştera Chindiei, Ostrovul Mare) or pieces with a single colour, probably proceeding from the powder consolidated by humidity (Icoane-Mehedinţi, Cuina Turcului). Sometimes the archaeological ochre samples have a uniform colour and well defined shapes of little flat cakes (Vădastra, Sultana, Gumelniţa), powders (Gumelniţa) or sticks (Sultana, Fântânele-Zimnicea).

Since heating the oxides accentuated the red hue,[54] the ochre samples properties which remain unchanged during the ceramic firing were chosen for comparison. In a diagram showing the relative intensity of the infra-red absorption band of quartz at 800 cm^{-1} and the iron content of the samples (Fig. 6), the representative points are spread enough but they are placed along a hyperbole with the form $y = 100/(0.86x-1.45)$ having a correlation coefficient of $r = 0.561$ (99% confidance) for the thirty analysed samples.

The existence of such a relation indicates that the archaeological ochre samples and the presumptive sources have the same chemical and mineralogical nature as the red lams and clays. The points are divided into two distinct groups. The principal group appears on the left with values under 10% Fe_2O_3 (Fig. 6B), while a less numerous group is right-hand with values higher than 11% Fe_2O_3 (Fig. 6C). This latter group with a lesser density includes the terra rossa and rendzinas at Moldoviţa, Cazanele Mari and Pescari from Clisura Dunării together with the epipalaeolithic ochres at Peştera Chindiei II and Icoane Mehedinţi and the Starčevo-Criş ochres at Cuina Turcului. The correspondent points of possible sources and the ochre samples from the tombs and from the archaeological strata are mixed to one another. This proves the provenience of the archaeological ochre from such types of sources.

[54] R. C. Mackenzie *op.cit.*, 302.

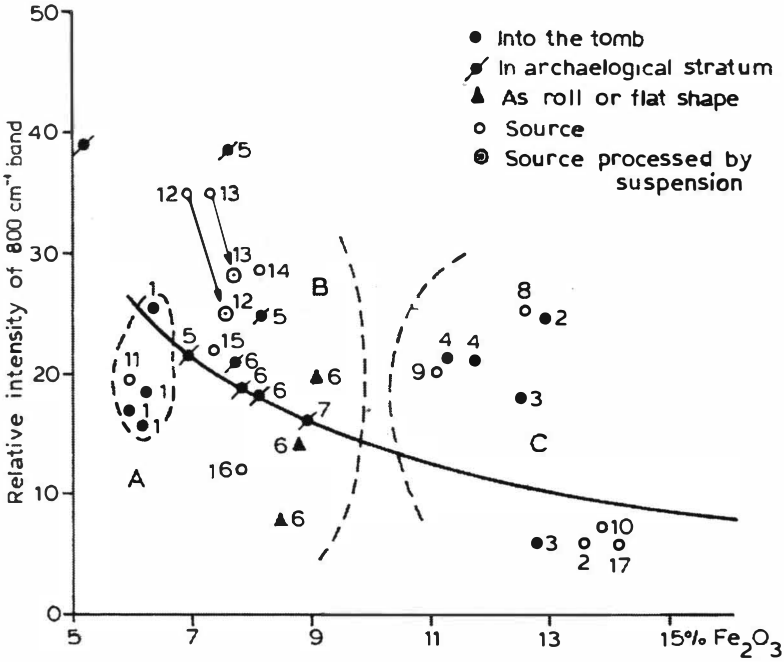

Fig. 6. Distribution of the archaeological ochres and their sources in regard to their 800 cm^{-1} infra-red band intensity and the Fe_2O_3 content. Ochre samples: Vădastra (Coţofeni tombs) – 1; Peştera Chindiei II (epipalaeolithic) – 2; Icoane Mehedinţi (epipalaeolithic) – 3; Cuina Turcului (Starčevo-Criş) – 4; Sultana (Gumelniţa A1) – 5; Gumelniţa (Gumelniţa B1)-6; Vădastra (Vădastra I) – 7. Probable sources: terra rossa Valea Marcoviei – 8 and Cazanele Mari – 17; red rendzinas: Pescari – 9 and Moldoviţa (Banat) – 10; lams: Vădastra Malul Roşu – 11, Grădinile – 12, Dăneasa – 13, Roşiorii de vede – 14 and Şimnic – 15; red clay Curpenelul – 16, terra rosa, Cazanele Mari – 17.

The other group of points (Fig. 6B) contain the samples of the red loams and clays at Vădastra (Malu Roşu), Grădinile, Dăneasa, Roşiorii de Vede, Şimnic, Curpenelu and the ochre from the tombs and from the arhaeological strata at Vădastra (Coţofeni tombs), Sultana (Gumelniţa A1 si A2) and Gumelniţa (B1). In this area the points corresponding to the probable sources and archaeological ochre are also mixed to one another. This proves that the archaeological ochre originates in the red loams and clays of such type. Indeed, in area A (Fig. 6) there are four ochre samples from the Coţofeni tombs at Vădastra and one of red loam at Malul Roşu located in the next neighbourhood of the settlement. The close grouping of the points indicates that this natural loam was directly used as ritual ochre put into the tomb and left on the skeleton.[55]

The loams with red hue at Grădinile and Dăneasa (Fig. 6 points 12 and 13) situated far away from the statistical curve were dispersed in water and the finer part was separated. The corresponding points of the samples separated in this way moved towards the curve at a little higher iron concentration. This new position suggests that many ornamental ochre samples could be processed as suspensions before their application.

The distribution of some sources and ornamental red applications in regard to the ratio of the iron and manganese contents divided the samples into two groups (Fig. 7). The first group with the ratio value under 50 includes the terra rossa and red rendzinas at Clisura Dunării (Moldoviţa, Cazanele Mari, Valea Marcoviei and Pescari) and the ornamental red on some Starčevo-Criş fragments at Schela Cladovei, Şimnic, Vădasatra with Linia Mare point and Vădastra at the eponymous settlement.

The second group with the Fe_2O_3/MnO ratio over 60 value includes red sedimentary loams and clays (Şimnic, Grădinile, Dăneasa, Curpenelul, Vădastra) and the ornamental ochre on some Starčevo-Criş fragments (Şimnic, Vădastra), Vădastra (Vădastra, Crusovu) and Gumelniţa culture (Sultana, Gumelniţa).

The repartition of the samples in histograms (Fig. 7) suggests that during the Starčevo-Criş culture in Oltenia the sources from Clisura Dunării took their exchange way to Schela Cladovei-Şimnic-Vădastra-Linia Mare) and continued to be used with a lower frequence in the Middle Neolithic at Vădastra.

Beginning with the Starčevo-Criş culture at Şimnic and Linia Mare-Vădastra ochre samples were used processed as suspensions prepared from red loams and clays from Central and Southern Oltenia and possibly from Central and Southern Muntenia. This processed ochre became prevalent in the Vădastra culture (Vădastra, Crusovu, Hotarani) and in the Gumelniţa culture (Sultana and Gumelniţa).

[55] Gh.Gâţă, Corneliu Mateescu, *op.cit.*, p. 5.

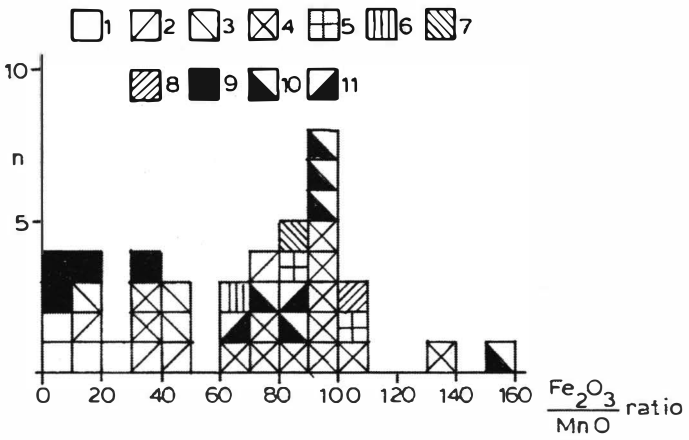

Fig. 7. Distribution or the Fe_2O_3/MnO ratio in ornamental ochres and sources. Ornamental ochres: Starčevo-Criş culture at Schela Cladovei – 1, Şimnic – 2 and Vădastra – linia Mare – 3; Vădastra culture at Vădastra – 4; Gumelniţa culture at Gumelniţa – 5. Ochre in archaeological strata: Vădastra – 6, Gumelniţa – 7 and Sultana – 8. Sources: terra rossa and red rendzinas at Clisura Dunării – 9; red lams at Şimnic, Gradinile Daneasa and Roşiorii de Vede – 10; reddish clays at Curpenelul and Segarcea – 11.

Some applications of ornamental ochre (Vădastra, Gumelniţa, Sultana and Sadievo Nova Zagora) and a La Tène ochre stick at Fântânele-Zimnicea have in their composition significant quantities of opal. This indicates exchange ways and sources from the south and downstream of the Danube, ways which were preserved from the Middle Neolithic to the La Tène.

Application of the firing technique of ochre at the same time with the pottery

Ornamental ochre applied on Vadsatra ceramics was a clayey material coloured in a red hue by iron and manganese oxides. The accentuated variation of ornamental ochre from red to yellowish brown is due not only to the source composition but also to the preparation mode, the application methods and the firing technique.

A graph of the redness rate RR-fineness index I_A[56] of the investigated samples (Fig. 8) gathers the analysed samples, the probable sources and the applications in three areas. In the A area red loams and clays drawing in Oltenia and Muntenia appear. The processed sources as suspensions and therefore prepared as finer material increase their redness and fineness index and pass into the B area (11 and 12 points, Fig. 8). In the latter area the red application appears on the Starčevo-Criş pottery at Şimnic and Linia Mare-Vădastra, on the Vădastra I, Vădastra II and Sălcuţa ceramics at Vădastra and on the Gumelniţa pots at the eponymous settlement.

The position of the points in A and B areas (Fig. 8) suggests that fairly all the red sources of Oltenia and Muntenia type were processed as suspensions and enriched in clay fraction and in iron oxide. In this case the redness was intensified during the firing when the vessel walls exceded the temperature of 300-400°C.[57] In the C area (Fig. 8) the sources of the Clisura Dunării (Moldoviţa, Cazanele Mari, Pescari) appear beside some ornamental ochre samples on the Starčevo-Criş ceramics at Schela Cladovei and Vădastra-Linia Mare and a few applications on the Vădastra II pottery at Vădastra. Generally the corrresponding points of the sources have a lesser fineness than the applications but there are also corresponding points of applications which have the same fineness as the sources. It results that partially some sources were applied directly and some others were improved as suspensions before their application. At Schela Cladovei, for example, a red of the background painted pottery (Fig. 8 point 1a) was applied as improved suspensions while on some vessels with red application (Fig. 8 point 1) natural ochre was directly painted. Other times, as at Şimnic (Starčevo-Criş) both types of sources originated from the samples of terra rossa (Fig. 8 point 2a) and local loams and clay from Oltenia and Muntenia (Fig. 8 point 2) were used.

The ornamental red was applied on the Neolithic pottery by many methods: passage of natural moisten clods on the vessel walls, coverage with viscous paste of the surface to be decorated with a hard and porous tool of an adequate shape, paint of more fluid suspensions on the pot walls and pouring of a suspension on the face to be decorated. In the thin sections only two kinds of application may be

[56] The fineness index was estimated by the width of the half heighth of the diffraction line at 10 A. The greater the index, the smaller the mean dimension of the particles.

[57] R. C. Mackenzie, *op.cit.*, p. 301.

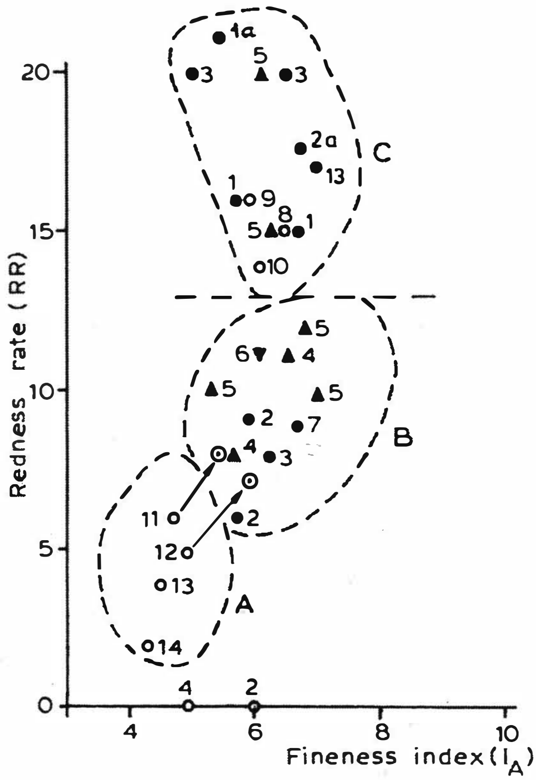

Fig. 8. Redness rate depending on the fineness index of the ornamental ochres and sources. Ochre applications: Starčevo-Criş at Schela Cladovei – 1, Şimnic – 2 and Vădastra Linia Mare – 3; Vădastra settlement as Vădastra I – 4, Vădastra II – 5 and Sălcuţa – 6; Gumelniţa B1 at Gumelniţa – 7. Sources; Cazanele Mari – 8, Moldoviţa – 9, Pescari – 10, Grădinile – 11, Dăneasa – 12, Şimnic – 13 and Vădastra Linia Mare – 14.

distinguished: drawing by pressing the ochre clods and viscous paste and scattering a little more fluid suspension over the surface to be decorated. In the first case the tickness layer is not uniform while in the second the colour layer is much more uniform and thinner.

The ochre layer drawn by means of natural clods as is observed on some Starčevo-Criş sherds at Schela Cladovei, Şimnic and Vădastra-Linia Mare often has slightly different chromes. For the reddish and grayish brown applications from Starčevo-Criş these chromatic variations are themselves ornamental manners giving beautiful nacreous lustre. The ochre paste was taken on a porous and hard tool of an adequate form and drawn on the moisten vessel surface for decoration. The repeated drawing with ochre paste with an application tool produced a thicker, more compact and more adhesive layer.

Such a tool was discovered in the settlement at Vădastra. It is a rim fragment of a Vădastra II pot on which natural ochre is imprinted on the tearing section. The section surface with ochre was polished while using the tool for ochre application.

Ornamental ochre in a thin layer with the uniform colour and tickness was drawn on the vessel walls with a kind of brush using a more fluide suspension. Sometimes brushing traces can be seen. The procedure was used for the concave surfaces with more accentuated curvature such as the vessel rims turned outside up and particular ornaments like a mask on the Vădastra II pot wall at Hotărani. For most ceramics fragments at Vădastra I and Vădastra II at the eponymous settlement, ochre was brushed after firing on the previous polished surfaces as is noticed when ochre was detached and only the polished slip remained.[58] In most cases ochre seams having been painted at the same time with a little viscous suspension because there are not traces of letting out the coloured surface.

Other times the surfaces for decoration were covered several times till the desired colour was obtained. In these cases the ornamental ochre is detached from the vessel walls as successive pellicles and proves that the red was brushed and allowed to dry before the next layer was added.

The red applications on the Starčevo-Criş ceramics were realized sometimes by aspersing the surface with a more diluated suspension. The coloured layer allowed each sprinkling to dry. The scattering of the colour by aspersing was continued till the wished chrome was obtained.

[58] The "slip" is a fine application obtained from the suspensions of the lithologic materials of the ceramic paste.

Other times the suspension was poured on the faces for the decoration and left free to impregnate the surface. The eventual excess was removed by letting out. This method was used for the inner faces ornamentation or for decoration of irregular surfaces such as the bottom of a little table of Vădastra II type with four legs at Vădastra.

In most cases the ornamental red was polished as indicates the accentuated orientation of mica minerals on the X-ray diffraction patterns of the pot faces.[59]

The firing period of Vădastra pottery at Vădastra was relatively short in a reduced atmosphere which led to an incomplete ceramics firing. Generally the external wall of the pot reached 350°-500°C but the vessel and figurine interior seldom reached 300-400°C. In these conditions the iron oxide transformation to hematite took place in different proportions and intensified more or less the red chromes of the applications.

Conclusions

The ornamental ochre that remained on the Vădastra pottery and also on all the Neolithic ceramics in the Lower Danube region has a mineral nature as show the X-ray diffraction analyses of the applications. The components which give their qualities to the ceramic dye stuff are the concentrations of the clay fraction and iron and manganese oxides. The ochre adhesion is due to the clay fraction content and the red nuances are due to the content and mineralogic nature of the iron and manganese oxides. It results that the potters from the Neolithic Age in the Lower Danube area selected the sources of ornamental red according to the nuances of the lithologic raw materials and to their adhesive properties on the pot walls.

At Vădastra and probably in other settlements too during the millennia of the sherd burial in the archaeological layers, natural ochre is little dissolved and can modify its nuance while fired ochre practically remains unchanged.

On Vadstra ceramics and also on all the others from the Neolithic age discovered in the Lower Danube region there are nuances from red to yellowish brown (Figs. 2-3-4) which may be grouped into three types: red (type I), dark red (II) and reddish brown (III).

The increase in the nuance numbers of ornamental ochre from 16 in the Vădastra I phase to 22 in the Vadstra II phase at Vădastra suggests an increasing demand of ochre and of raw materials for ochre inclusively, accompanied by the multiplication of the number of sources and the extension of the exchange area at Vădastra, Gumelniţa, Sultana, Brenitza, Sadievo Nova Zagora and Fântânele-Zimnicea (La Tène).

The remaining ochre on the Vădastra figurines at Vădastra and Crusovu has always a red colour (I and II types) and proves the care for the selection of ornamental ochre. In the red of the figurines and pots one notices the differences of nuance but not of the composition, which confers the colour a ritual meaning. It is possible that the ornamental ochre had been tried previously on pots and afterwards used on the figurines.

The ochre sources are loams, clays and terra rossa from the Central and Southern parts of Oltenia and Muntenia, the Northern parts of Bulgaria, Clisura Dunării and probably the Northern part of Serbia.

All the ochre samples discovered in the Lower Danube region in the epipalaeolithic, Starčevo-Criş and Coţofeni tombs were local samples and indicate the continuity of using a ritual from the epipalaeolithic to the Bronze Age in this region.

There are two ochre qualities in the Neolithic Age in the Lower Danube region. One is similar to the samples of terra rossa and redzinas from Clisura Dunării. This appears on some Starčevo-Criş fragments at Schela Cladovei, Şimnic and Vădastra-Linia Mare and in the Vădastra culture at Crusovu and Vădastra.

Another type originates from the red loams and clays of the Oltenia and Muntenia sediments and from the south of the Danube and appears on the sherds from Starčevo-Criş at Şimnic and Vădastra-Linia Mare, on Vădastra ceramics at Vădastra, Crusovu and Hotărani, on Boian pottery at Vidra, Aldeni,

[59] The more oriented the mica minerals. the sharper and more intense the diffraction lines at 10 A.

Spantov and Radovanu and on Gumelniţa ceramics at Sultana and Gumelniţa. The fineness of the ornamental ochre (Fig. 8), the significant content of iron oxides and the small quartz content indicate above all that the sedimentary sources were enriched with suspensions of particles with smaller dimensions.

Some samples with opal at Vădastra, Gumelniţa, Sultana, Sadievo Nova Zagora and Fântânele Zimnicea originate from the southern part of the Danube and may include also the region in the downstream of the confluence of the Olt with the Danube. These are connected with the geological formation of the continental Aptian from Dobrogea and indicate the continuous use of this ochre type from the Middle Neolithic Age till La Tène.

The continuous improvement of the ochre technique in the Neolithic at Vădastra and in the Lower Danube area results from the application adherence, the preference for the ochre applied before firing and the more and more frequent red colours.

Ochre was applied on the Neolithic ceramics in the Lower Danube region by many methods. The application by drawing of the colour with natural clods produce an ornamental red with chromatic variations and was used in the Starčevo-Criş culture at Schela Cladovei, Şimnic and Vădastra-Linia Mare.

The drawing of an ochre paste with a strong and porous tool such a ceramic fragment with an adequate shape is the most frequent method in Vădastra II phase at Vădastra and Crusovu.

The application on the pots of a fluider suspension by means of a kind of brush was used particularly for the ornamentation with ochre after firing on some Vădastra I and II vessels at the eponymous settlement.

The ochre pellicle made on the whole vessel surface by spattering with a diluted suspension appears on Starčevo-Criş sherds at Schella Cladovei, Şimnic and Vădastra-Linia Mare.

In the Vădastra culture, the pourring of the suspension on irregular concave surfaces is used more rarely, as appears on a Vădastra II little table with four legs at Vădastra. The red application on the inner part of some Starčevo-Criş vessels at Schela Cladovei was also obtained by this method.

The location of the ochre sources including the Lower Danube flow with Serbia, Banat, Oltenia, Northern Bulgaria, Muntenia and possibly Dobrogea, points out the intensive exchange relation during the Vădastra culture and suggests the maintaining of these connections in all the Neolithic time.

Like ceramics, ochre brings many explanations in connection with the inhabitants' activities, the connexions and exchanges with the neighbouring populations, in other words contribute to the completion of the historical image of the Middle Neolithic at Vădastra and the Lower Danube area.