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Re-identification of plant impressions on prehistoric pottery from Ukraine

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ABSTRACT

In this paper, we report newly obtained data on cereals from the Neolithic to Bronze Age from the northern side of the Black Sea. One part of the North Pontic area, located within present-day Ukraine, is one of the focal points of discussions on the agricultural dispersal between West and East Eurasia; however, existing reliable cereal data are scarce. In this study, we employed an improved silicone casting method to obtain impressions from pottery artifacts and conducted a survey of more than 30,000 pottery samples from different times. Observations of seed surface texture using scanning electron microscopy further improved the identification accuracy. The results showed no reliable cereal impressions in Ukrainian sub-Neolithic pottery, despite the predicted exceptionally early start of arable agriculture prior to the 6th millennium BC. In contrast, in Linear Band and Eneolithic pottery, cereals originating from West Asia were identified, although Chinese millet was absent. Meanwhile, abundant *Panicum miliaceum* impressions appeared abruptly with incidental barley and wheat in Late Bronze Age pottery. The Chinese millet species *Setaria italica*, which is clearly distinguishable from *P. miliaceum* using our method, was not identified. The archaeobotanical dataset obtained in this survey is an important achievement in examining food globalization that crossed the east and west of Eurasia.

1. Introduction

One part of the North Pontic area, located within present-day Ukraine, is one of the focal points of discussions on agricultural dispersal in Eurasia (Motuzaite-Matuzeviciute, 2020; Dal Corso et al., in press). Agriculture is thought to have started exceptionally early in Ukraine, dating back to before the 6th millennium BC (Pashkevich, 2000; Kotova, 2003). However, this conclusion is based on unreliable identification of cereals from impressions in pottery (Motuzaite-Matuzeviciute et al., 2013a; Stevens et al., 2016). On the other hand, Motuzaite-Matuzeviciute (2012) carried out archaeobotanical investigations in eastern Ukraine and southwest Russia but did not find any cereal impressions at 15 sites from the 8th to 6th millennium BC, suggesting that arable agriculture was introduced there around the second half of the 5th millennium BC. For pottery impressions of broomcorn millet dating back to the Neolithic period on the north sides of the Black Sea, it has been pointed out that re-evaluations of these identifications and time ratios are necessary (Hunt et al., 2018). Thus, the authors of the current paper think it is necessary to revise the previous conclusions regarding start of not only broomcorn millet cultivation but also arable agriculture as such in the whole territory of Ukraine before 6000 BC. A base of such re-evaluation is already founded by a new analysis of the directly radiocarbon-dated macrobotanical remains.

Recently, a package of cultivated plants of West Asian origin was recovered from Ratniv-2 in western Ukraine, a Linear Band Pottery Culture (LBK) site. Among them, two *Triticum dicoccum* samples dated to 5471–5230 cal BC were identified (Motuzaite-Matuzeviciute and Telizhenko, 2016), representing the earliest cereals confirmed in Ukraine by

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accelerator mass spectrometry (AMS). Meanwhile, Salavert et al. (2021) compared the plant composition of two archaeological sites, Buh-Dniester Culture and the LBK, on the South Buh River[1] in south-western Ukraine and found cereals of West Asian origin at only the LBK site, which ranges from the late 6th millennium to the early 5th millennium BC. Therefore, it will be necessary to determine whether cereal cultivation was limited to the LBK group in Neolithic Ukraine or whether it had spread to indigenous eastern groups.

In the 5th and 4th millennia BC, a great number of Eneolithic Trypillia settlements were located in southwest Ukraine and in the western side of the Middle Dnipro River region. These groups are recognized as one of the first agricultural societies in the area, with the scale of the settlements thought to represent the largest mega-sites in Eurasia during the 4th millennium BC (Videiko, 2012; Müller et al., 2016). However, Chapman (2017) questioned the archaeobotanical data, suggesting that the size of the settlements was not correlated with an increase in the production of cereals. In addition, isolated charred *Panicum miliaceum* has been also reported at the Trypillia sites (Müller et al., 2016), suggesting that re-evaluation of archaeobotanical data is necessary to clarify the nature of agriculture in Trypillian society.

The emergence of *P. miliaceum*, a millet of East Asian origin, and its route of spread within Europe remains an important topic among archaeobotanists. Recent direct AMS dating of charred European *P. miliaceum* revealed that the oldest sample was one found in Ukraine that dated to between the 17th and 15th centuries BC (Filipović et al., 2020). However, in the article, the authors also point out the insufficiency of data from the North Pontic region. Meanwhile, three possible dispersal routes of *P. miliaceum* have been suggested: the Balkans via Anatolia, the Caucasus corridor, and the West Eurasian Steppe (Riehl, 1999; Miller et al., 2016; Wang et al., 2019; Trifonov et al., 2017; Motuzaite-Matuzeviciute, 2020). Thus, to confirm which of these routes existed, multiple reliable data from Ukraine are also required. Impressions of *P. miliaceum* from pottery belonging to the Usatovo Culture have already been re-examined using plants seed replication via obtaining a cast of clay instead of silicon but no reliable *P. miliaceum* impressions have been found (An et al., 2017). However, this methods can create problems in terms of the resolution; therefore, high-resolution analysis of cereal impressions from the Neolithic to Bronze Age pottery in Ukraine is needed.

Japanese scientists have pioneered this casting method and they have successful case studies (Endo and Leipe, in press). In the Japanese archipelago, as in Ukraine, data based on the flotation method are limited, and impression surveys conducted over the past 20 years have successfully revealed the emergence of cereal cultivation at the end of the Final Jomon period, with the combined cultivation of rice and millet (Nasu and Momohara, 2015). Notably, the presence of *P. miliaceum* and *Setaria italica* was precisely recognized using impressions in pottery (Nakazawa, 2009; Sasaki et al., 2010; Endo and Takase, 2011; Nakayama, 2014). Flotation is also advantageous for discussing the utilization of plants based on quantitative analysis. However, considering the limited surveys of charred remains, the high-resolution method we use can effectively provide reliable large datasets on previously excavated and stored pottery within a short period of time and at low cost (Endo, in press).

In the present study, we conducted the new survey of cereal impressions obtained from pottery dated from the Neolithic to the Bronze Age with the aim of clarifying the emergence of cereal cultivation in Ukraine.

Originally, the term agriculture includes both arable agriculture (cultivation) and pastoral agriculture, but in this paper, only arable agriculture is considered from archaeobotanical data, therefore, hereinafter, the term agriculture refers to arable agriculture.

2. Chronology

Dating of plant specimens from impressions tends to be based on the typological chronology of the pottery itself; however, correlations of this chronology with the absolute chronology of some Ukrainian archaeological cultures remain controversial (e.g., Motuzaite-Matuzeviciute, 2013; Haskevych et al., 2019). In addition, many potsherds with impressions are small and lacking decoration, and thus the typological approach is not always effective. Moreover, even with potsherds excavated from a layer with a well-defined time frame within a multi-stratified site, contamination from another period can occur. In Ukraine, the start of "Neolithic" traditionally refers to the emergence of the first pottery, not the spread of agriculture (for more details, see Motuzaite-Matuzeviciute, 2013). Thus, the presence of early-looking prehistoric potsherds at a site is considered sufficient for classifying it as Neolithic. When considering the Ukraine Neolithic in terms of Marek Zvelebil's availability model (Zvelebil, 1986), the Buh-Dniester, Kyiv-Cherkasy, Surskyi, Donets, and Azov-Dnipro cultures should be attributed to communities at the availability stage. The influence of farming groups on their hunter-gatherer neighbours can be seen only in the sporadic exchange of prestigious goods, as well as in attempts to imitate the decorations and forms of pottery from the Criş, Vinča, and Trypillia and some other Western cultures. Thus, it would be more accurate to refer to cultures situated east of the agricultural frontier as "para-Neolithic," "sub-Neolithic," or "ceramic Mesolithic" rather than "Neolithic".

The criteria of the Eneolithic (Chalcolithic or Copper Age) also remain uncertain. In Ukraine, all sites with finds of copper items are usually considered to belong to that period. The Eneolithic attribution of the classical farmers of the Trypillia Culture and the first steppe pastoralists with copper tools and burials under mounds is not in doubt. However, their settlements and graves are not found everywhere in Ukraine. Next to their areas, there are many synchronous sites that are traditionally interpreted as belonging to the Neolithic and/or sub-Neolithic.

Both the end of the Eneolithic and the beginning of the Bronze Age are usually associated with the first bronze items spread in the area of Ukraine in the end of 4th millennium BC (Klochko et al., 2020). Therefore, many late Eneolithic sites are *de facto* synchronous with the Early Bronze Age. The same applies to the end of the Bronze Age, when the first iron tools and weapons began to appear. The start of mass diffusion of iron in the 8th century BC was linked with the Cimmerians, the first people to inhabit the territory of Ukraine who are mentioned in written records.

For a long time, chronological studies of Neolithic to Bronze Age Ukraine were based only on comparisons with more accurately dated material from the Mediterranean, Balkans, and Central Europe. This was based on both the typological method and the not numerous so-called "import" finds at local sites. From the mid-1970s, radiocarbon measurements began to be used, and from the 1990s, radiocarbon dating became systematic and widespread. According to approximate calculations, about 250 dates each have been obtained for the Ukraine Neolithic and Eneolithic, and more than 500 dates have been obtained for the Ukraine Bronze Age. However, there are no complete catalogues of these dates. The most significant series for the Neolithic and sub-Neolithic were published by Telegin et al. (2002), Burdo (2001–2002), Kotova (2003), Manko (2006), and Haskevych et al. (2019), whereas Telegin (1985), Burdo and Videiko (1998), Kotova and Videiko (2004), Rassamakin (2011; 2012), and Diachenko and Harper (2016) provided a catalogue for the Eneolithic. Numerous dates of Bronze Age sites were also published in a special volume of the "Baltic-Pontic Studies" edited by Końko and Klochko (2003).

[1] In the article, all Ukrainian geographical names as well as names derived from the archaeological sites and cultures are based on those written in Ukrainian. Of the dozen ways of Romanizing the Ukrainian alphabet, the standard officially adopted by the Ukrainian government in 2010 is employed here.

These dates were obtained in different years, in different laboratories, and by using different methods and samples, including human and animal bones, charcoal, shells, and organic temper from the clay paste of pottery. Thus, not all dates are equally informative. Critically reviewing them is an impossible task for this study. Therefore, we collected and generalized data for the approximate boundaries of time intervals representing only those cultures under investigation (Fig. 1).

3. Materials and methods

3.1. Materials

The search for impressions was carried out by 30,753 finds from the collections of 64 sites kept at the Institute of Archeology of the National Academy of Sciences of Ukraine in Kyiv and the Archeology Research Laboratory at Faculty of History and Philosophy of Borys Grinchenko Kyiv University (for details see Appendix A: Table A.2).

3.1.1. Sub-Neolithic

Nearly 14,350 potsherds from 35 sub-Neolithic sites were observed (Fig. 2, Appendix A: Tables A.1 & A.2), including 27 specimens that have previously been reported as yielding crop impressions. The vast majority of the processed material belongs to the Buh-Dniester Culture and Kyiv-Cherkasy Culture.

Pashkevich (2000: 405; personal communication) previously identified 14 impressions of six cultivated plant species on 11 potsherds of the Buh-Dniester Culture vessels from seven sites in Ukraine. These collections were re-examined, along with material from 15 more sites of the Buh-Dniester Culture. Accordingly, a total of 3,540 potsherds from 22 sites in the South Buh River basin were examined (Appendix A: Tables A.1 & A.2).

The area of the Kyiv-Cherkasy Culture is the Middle Dnipro region, which ranges from the mouth of the Sozh River in the north to the mouth of the Orel River in the south. Pashkevich (2000: 407) previously identified impressions of 36 seeds from nine cultivated plant species on

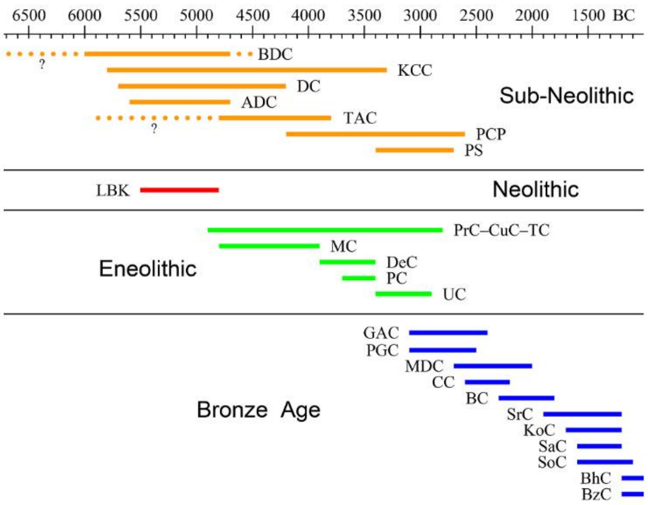

Fig. 1. Chronology of the late prehistoric cultures of Ukraine, the materials of which were processed and discussed in this study. Abbreviations: ADC – Azov-Dnipro culture, BC – Babyne culture, BDC – Buh-Dniester culture, BhC – Bilohrudivka culture, BzC – Bilozersk culture, CC – Catacomb (Katakombna) culture, CuC – Cucuteni culture, DC – Donets culture, DeC – Deriivka culture, GAC – Globular Amphorae culture, KCC – Kyiv-Cherkasy culture, KoC – Komariv culture, LBK – Linear Band Pottery culture, MC – Malice culture, MDC – Middle Dnipro culture, PC – Pyvykha culture, PCP – Pit-Comb Pottery cultural unity, PGC – Pit Grave (Yamna) culture, PrC – Precucuteni culture, PS – Pustynka-5 type sites, SaC – Sabatynivka culture, SoC – Sosnitsa culture, SrC – Srubna culture, TAC – Tash-Air culture, TC – Trypillia culture, UC – Usatove culture.

34 potsherds from six sites that are now attributed to the Kyiv-Cherkasy Culture and sites of the Pustynka-5 type. In addition, Pashkevich also recorded a number of wild species. Collections from four of these sites were reviewed here, along with material from three more sites of the Kyiv-Cherkasy Culture. Therefore, nearly 7,900 sub-Neolithic fragments from these sites were examined (Appendix A: Tables A.1 & A.2).

3.1.2. Neolithic

Only 449 potsherds from the classical Neolithic LBK were observed (Appendix A: Tables A.1 & A.2), 21 of which represented debris of a few import vessels found on the Buh-Dniester Culture sites of Dobrianka-3 and Bazkiv Ostriv. The remaining 428 samples represent a small portion of material from the confirmed LBK sites of Hnidava[2], which was excavated by Kuchera in 1968–1969, and Holyshiv-2, which was excavated by Okhrimenko in 1988.

More than 60 years ago, two vessels containing charred grains of wheat, spelt, and peas were found at a burial within the Nezvysko site in the Middle Dniester region (Passek and Chernysh, 1963: 20). Recently, charred grains of cultivated plants were also discovered using the flotation method in two LBK sites. The first was Ratniv-2 in Volhynia, which is located only 9.3 km from Holyshiv-2 and 11.3 km from Hnidava, with samples yielding finds of *Hordeum vulgare*, *Lens culinaris*, *Linum usitatissimum/catharticum*, *Pisum sativum*, *Triticum/Hordeum* sp., *Triticum monococcum*, and *T. dicoccum* (Motuzaite-Matuzeviciute and Telizhenko, 2016). The second site was Kamyane-Zavallia in the Middle Southern Buh region, which yielded samples of *T. turgidum* cf. subsp. *dicoccon* (*T.* cf. *dicoccum*, emmer), *Hordeum* sp. / *T.* sp. (barley/wheat), *T.* cf. *monococcum* (einkorn) and cf. *Hordeum* sp. (Salavert et al., 2021: 355). Earlier, Pashkevich (2000: 409) discovered 42 impressions of cultivated plants on pottery from four Volhynian LBK sites, including 10 impressions from Holyshiv-2.

3.1.3. Eneolithic

More than 11,700 potsherds and 65 vessels from 32 Eneolithic sites were observed. Almost all of the samples comprising 11,337 potsherds and 64 vessels belonged to the Trypillia Culture, and the vast majority plus a large quantity of daub were from 15 confirmed Trypillia (phases A to C) sites. In addition, 620 Trypillian potsherd samples from 16 multi-stratified sites representing mainly sub-Neolithic finds were also observed. The remaining 391 potsherds and 1 vessel were non-Trypillian, 278 samples of which were from the Kukrek site in Crimea. A total of 36 potsherds and 1 vessel were of the Deriivka Culture from both the eponymous Deriivka site and the Kyiv-Cherkasy Culture site of Pyshchyky-2 in the Middle Dnipro region, whereas 76 potsherds of the Eneolithic Malice Culture were from the above-mentioned Holyshiv-2 site in Volhynia (Appendix A: Tables A.1 & A.2).

Yanushevich, Kuzminova, and Pashkevich previously identified six cultivated plant species on impressions in pottery wares and clay figurines, daub, and on macro remains from 64 Trypillia Culture sites located in Ukraine and Moldova (Pashkevich and Videiko, 2006, see their Tables 3–7). Meanwhile, more recent investigations based on studies of macro remains in the Trypillia CI stage mega-site of Maidanetske resulted in identification of the same species, except *P. miliaceum* (Kirleis and Dal Corso, 2016). Remains of tools used by Trypillian farmers, including stone and horn parts from hoes, and flint blades for sickles, are not very numerous. However, it is thought that the development of agriculture provided sufficient food to support the inhabitants of the settlements, including large mega-sites (Shukurov et al., 2015).

3.1.4. Bronze Age

Approximately 3,800 potsherds and 66 vessels of the Bronze Age from 24 sites were observed. A more or less representative series of 570

[2] This site is also known as Rovantsi.

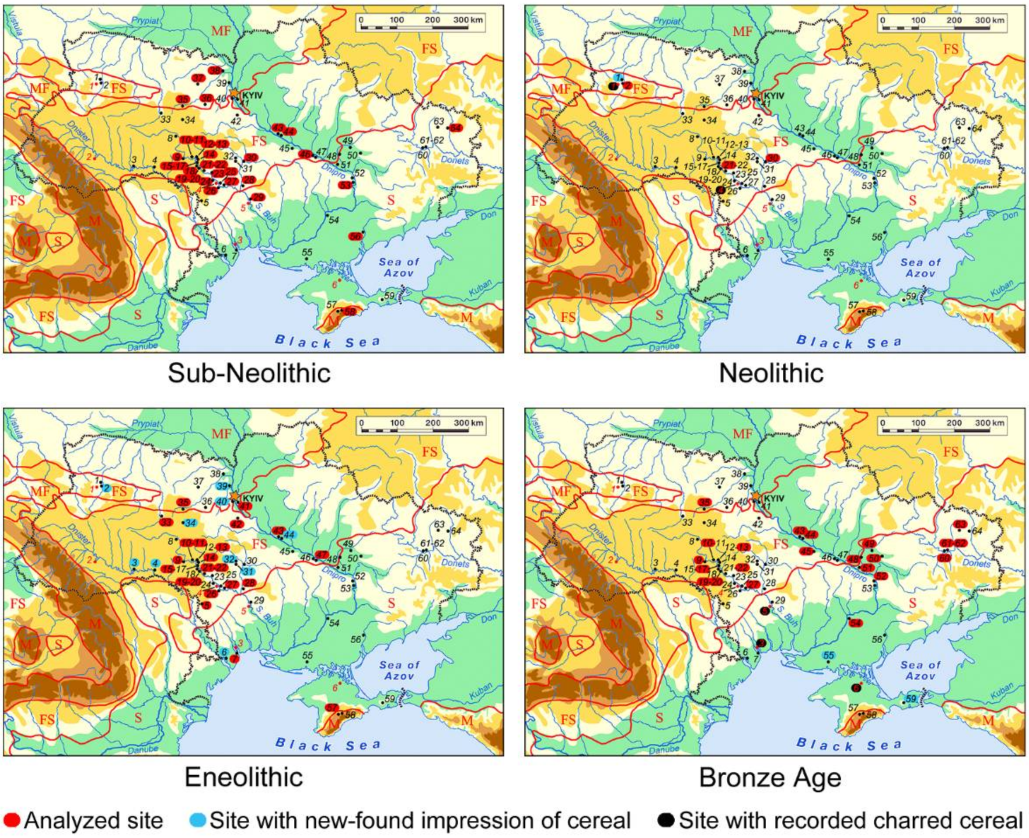

Fig. 2. Location of sites from which charred plant remains and impressions were obtained. Black dots and numbers represent sites from which material was observed by the authors. 1 – Hnidava; 2 – Holyshiv-2; 3 – Luka Vrublivetska; 4 – Bernashivka; 5 – Tymkove; 6 – Maiaky; 7 – Usatove; 8 – Sandraky; 9 – Pechera-1; 10 – Samchyntsi-1; 11 – Samchyntsi-2; 12 – Shchurivtsi-Porih; 13 – Shymonovske-2; 14 – Zankivtsi-2; 15 – Sokiltsi-1; 16 – Sokiltsi-2; 17 – Sokiltsi-6; 18 – Hlynske-1; 19 – Ladyzhin-1; 20 – Ladyzhin-2; 21 – Bazkiv Ostriv; 22 – Mytkiv Ostriv; 23 – Shumylove-Cherniatka; 24 – Haivoron-Polizhok; 25 – Zhakchyk; 26 – Savran; 27 – Melnychna Krucha; 28 – Mykolyna Broyaka; 29 – Puhach-2; 30 – Dobrianka-3; 31 – Hrebeniukiv Yar; 32 – Maidanetske; 33 – Chortoryia; 34 – Troianiv; 35 – Hutir Teterevskyi; 36 – Lazarivka; 37 – Krushnyky; 38 – Hryni; 39 – Kazarovychi; 40 – Chapaivka; 41 – Sofiivka; 42 – Kolomyitsiv Yar; 43 – Pyshchyky-2; 44 – Buzky; 45 – Velyka Andrusivka; 46 – Uspenka; 47 – Deriivka; 48 – Lyskivka; 49 – Yosypivka; 50 – Pereshchepyne; 51 – Sorochyne; 52 – Skelia-Kamenolomnia; 53 – Mykilske-2; 54 – Babyne-3; 55 – Novokyivka; 56 – Kamiana Mohyla-1; 57 – Kukrek; 58 – Buran-Kaya-4; 59 – Kirove; 60 – Hlyboke Ozero-2; 61 – Usove Ozero; 62 – Yampil; 63 – Rudivka; 64 – Hyrlo Biloi. Red dots and numbers represent sites with charred plant remains that were mentioned in the text: 1 – Ratniv-2; 2 – Nezvysko; 3 – Cherevichne; 4 – Kamyane-Zavallia; 5 – Vynohradnyi Sad; 6 – Bolotne. Abbreviations: MF – mixed forest; FS – forest-steppe; S – steppe; M – mountain. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

to 1,300 of the examined fragments originate in settlements of the Pit Grave Culture and Catacomb Culture of the Early Bronze Age (Skelia Kamenolomnia), the Babyne Culture of the Middle Bronze Age (Babyne-3), and the Sabatynivka Culture (Novokyivka) and Srubna Culture (Usove Ozero) of the late Bronze Age. A few vessels from the Srubna Culture settlements Yampil, Hlyboke Ozero-2, Yosypivka were also inspected. It should be noted that, with the exception of the Novokyivka settlement, these represent only a small random sample of potsherds and vessels, not the complete collections. Moreover, several dozen vessels from the Pereshchepyne, Rudivka, and Sorochyne barrow burial grounds, all of which contain material from the above-mentioned cultures, were also examined. All listed sites were located in southern and southeastern Ukraine (Fig. 2). The rest of the examined materials were obtained from the Globular Amphorae Culture and Corded Ware Culture of the early Bronze Age as well as the Komariv Culture, Sosnitsa Culture, Bilohrudivka Culture, and Bilozerka Culture of the late Bronze Age. These samples were neither numerous nor noteworthy, and most represented late admixture introduced in sub-Neolithic material of multicultural sites in the Southern Buh and Middle Dnipro regions (Appendix A: Tables A.1 & A.2).

It is now established that the Pit Grave (Yamna) Culture in Ukraine existed from the last quarter of the 4th to the middle of the 3rd millennium BC and represents the western region of a very large historical-cultural community. Stationary settlements have been found in the Lower Dnipro region, of which 20,000 potsherds from two of these (Mykhaylivka and Skelia Kamenolomnia) were examined by Pashkevich (1991: 15). She identified the impressions of a few species of cultivated plants, with six notable impressions of *P. miliaceum* in the Skelia Kamenolomnia site. Impressions of grains, spikelet forks, and fragments of straw of *T. compactum*, *T. monococcum*, *H. vulgare*, and millet[3] were also recorded by Kuzminova and Petrenko (1989: 120) on vessels from four Pit Grave Culture burial sites in the Lower Dniester.

The Pit Grave Culture was replaced by the Catacomb (Katakombna) Culture in the middle of the 3rd millennium BC. In this culture, finds of cultivated plants were clearly recorded with the discovery of a bag of spikelets from *T. dicoccon* and *T. monococcum* at burial No. 28 under barrow No. 14 near the village of Bolotne in Crimea (Pashkevich, 2000: 412). Impressions of millet and bare wheat grains were also found in Catacomb Culture vessels at burial sites under mounds near the village of Myrne between the Lower Dniester and Lower Danube (Kuzminova and Petrenko, 1989: 120).

The Babyne Culture (also called the Multirolled Ceramic Culture and the Mnogovalikovaya Culture) represents the transition to the Late Bronze Age (23–18/17 centuries BC) in most parts of Ukraine. In pottery samples from the eponymous settlement of Babyne-3 on the Lower Dnipro River, Pashkevich (2000: 414) identified four impressions of wheat, barley, and *P. sativum* grains. Impressions of millet grains were found in pottery from the Strumok barrow burial ground in the Lower Danube region, whereas impressions of *T. monococcum* and barley were found in pottery from the Kiselovo barrow burial ground on the Kuialnyk coastal lake (Kuzminova and Petrenko, 1989: 120).

The Srubna Culture (19/18–13/12 centuries BC) represents the basic

[3] Hereinafter, the authors of the current article use the term millet as it is done in the respective referred papers – without specification to certain species.

formation of the forest-steppe and steppe zones in Ukraine. Very few published findings of plant cultivation are available for this period, and according to Pashkevich (2000: 413), who reviewed more than 7,000 potsherds from the Usovo Ozero settlement on the Seversky Donets River, only 276 impressions of eight species of domestic plant were observed. Charred millet grains were also found at the same site in dwelling No. 4 (Pashkevich 1991: 21).

In contrast, in the Sabatynivka Culture (16–13/12 centuries BC), which represents the basic formation of the North Pontic steppe zone, much evidence of crop farming has been found. Examining nearly 14,200 fragments and tens of kilograms of daub from 11 sites, Pashkevich (2000: 414) identified 165 impressions of nine cultivated plant species, including *P. miliaceum*. In addition, 3,584 samples of charred grain from 10 species of domestic plant were also discovered using the flotation method in the settlement of Vynohradnyi Sad on the South Buh River. Samples included *T. monococcum* (3), *T. dicoccon* (33), *T. spelta* (3), *T. aestivum s.l.* (11), *H. vulgare* var. *coeleste* (1), *H. vulgare* (3464), *P. miliaceum* (53), *P. sativum* (5), *Vicia ervilia* (2), and *Vicia* sp. (9) (Pashkevich 2000: 414). Millet grains caked into a homogeneous mass were also found in the Cherevichne settlement on the Khadzhibey Estuary, whereas impressions of *T. monococcum*, *T. dicoccon*, *H. vulgare*, and *P. miliaceum* grains were recorded by Kuzminova and Petrenko (1989: 120) in pottery from a few settlements in the northwestern Black Sea area and Moldova.

3.2. Impression analysis

Many archaeobotanists have questioned the reliability of plant identifications made from impressions in pottery (Zohary and Hopf, 2000: 5; Hunt et al., 2008; Stevens et al., 2016: 1545), particularly those of seeds at the species level. We therefore employed an improved impression method to increase accuracy, using anatomical observations of un-carbonized plant surfaces obtained via scanning electron microscopy.

3.2.1. Sampling

For the most accurate identification of each item with an impression, which silicone copy was made, in Cyrillic inscriptions on its surface with information regarding localization within the site, depth, inventory No. and year of exploration were recorded in a special database. The part of a vessel and occurrence of decoration was also specified there (Appendix C). However, the main way of unerring identifying the shard or vessel is their photos, which are specified in Appendix C with a unique code. Any of these photos are available upon reasonable request addressed to the corresponding author.

3.2.2. Procedure

These impressions were identified using silicone casting method (called "impression replica" method in Japan) based on Hisa and Katada (2005) and was carried out as follows (Fig. 3):

(1) Inner and outer surfaces and fracture surfaces of pottery were observed with the naked eye and a loupe to determine potential plant impressions.

(2) A solution of 5% Paraloid B-72 in acetone was applied to protect the surface of the pottery.

(3) Silicone resin (Tokuyama Fit Tester; Tokuyama Dental Corp., Tokyo, Japan) was then applied to the impressions.

(4) Casts were removed from the impressions after hardening.

(5) The surface of the impressions was cleaned with 100% acetone to remove any residual Paraloid B-72.

(6) Observations of the cast were performed using SEM (VE-8800; Keyence, Osaka, Japan; provided by Ancient Scientific Institute, Meiji University, Japan), and the anatomical details of seed surfaces transcribed from the impressions were obtained along with photographic images.

(7) Casts were identified based on morphological features by using modern and fossil comparative material and identification criteria (Fuller, 2017; Nasu et al., 2007; Fuller and Zhang, 2007; Jacomet et al., 2006; Tsubakizaka, 1993).

4. Results

Searching the impressions of plant seeds, the authors examined materials of 64 sites in 2016–2018. The total number of analysed finds from them is 30,619 potsherds and pieces of daub plus 134 intact and restored vessels (for details see Appendix A: Table A.2). As a result, 488 silicone casts taken from impressions in pottery items from 50 sites were obtained. Plant species were identified in 122 casts. These were cultivated plants in 101 cases (Appendices B & C).

4.1. Re-identification of published plant impressions

Re-excavation of previously documented potsherds containing plant impressions was very time and effort consuming. Accordingly, a total of 27 sub-Neolithic potsherds that were published as yielding 29 impressions of cultivated plants, were found in the collection at the Institute of

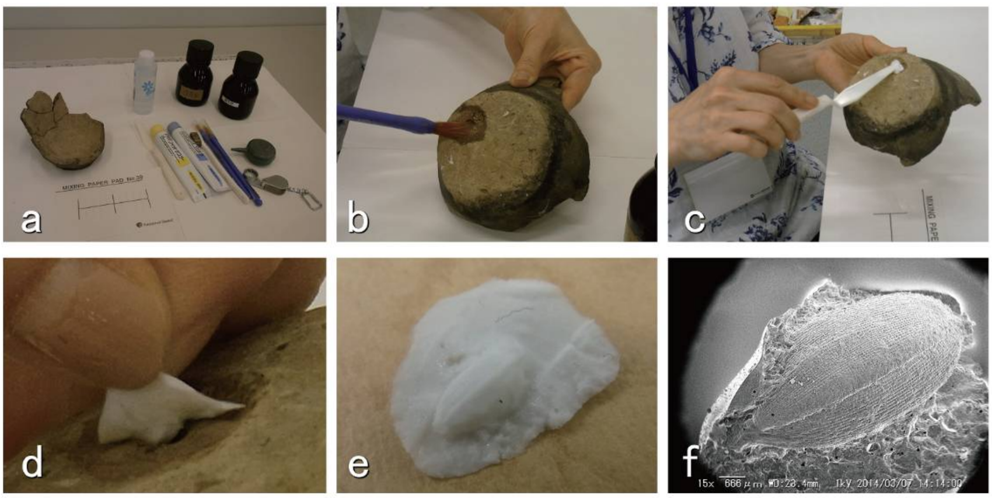

Fig. 3. Procedure for making a silicone cast from an impression. (a) Tools for making casts, (b) Coat the impression and the surrounding part of the pottery with acetone containing 5% Paraloid B72 to protect the pottery surface, (c) Fill silicone into the impression, (d) Remove the cast from the impression after hardening, (e) Silicone cast of the impression, (f) SEM image of the silicone cast.

Archaeology, NAS of Ukraine. Casts were taken from 32 impressions on 25 of these potsherds. However, we were unable to accurately identify any domestic plant samples, and hardly any plant impressions were confirmed (Appendix B: Tables B.1 & B.2, Figs. B.1 & B.2).

4.2. New observations and identification

4.2.1. Sub-Neolithic

In the newly observed sub-Neolithic samples, cf. *Sambucus* (Fig. 4a & b), Asteraceae (Fig. 4c & d), Apiaceae (Fig. 4e), Poaceae (Fig. 4f & g), and snail specimens (Fig. 4h-j) were observed, but no cultivated plants were identified (Appendix B: Table B.1). At the Buzky site, which is located on a low sandy hill on the Dnipro River flood plain, only one possible *Triticum* candidate was identified. Although the Buzky site is thought to belong to the sub-Neolithic, the potsherd containing this impression is considered to belong to the Trypillia Culture (stage B) based on its typology (Fig. 4k & l).

4.2.2. Neolithic

Of 449 observed Neolithic potsherds from LBK pottery, we identified only two that contained impressions of *T.* cf. *dicoccom* (Fig. 4m-o) at the Hnidava site, which is located on the left loess terrace of the Styr River at

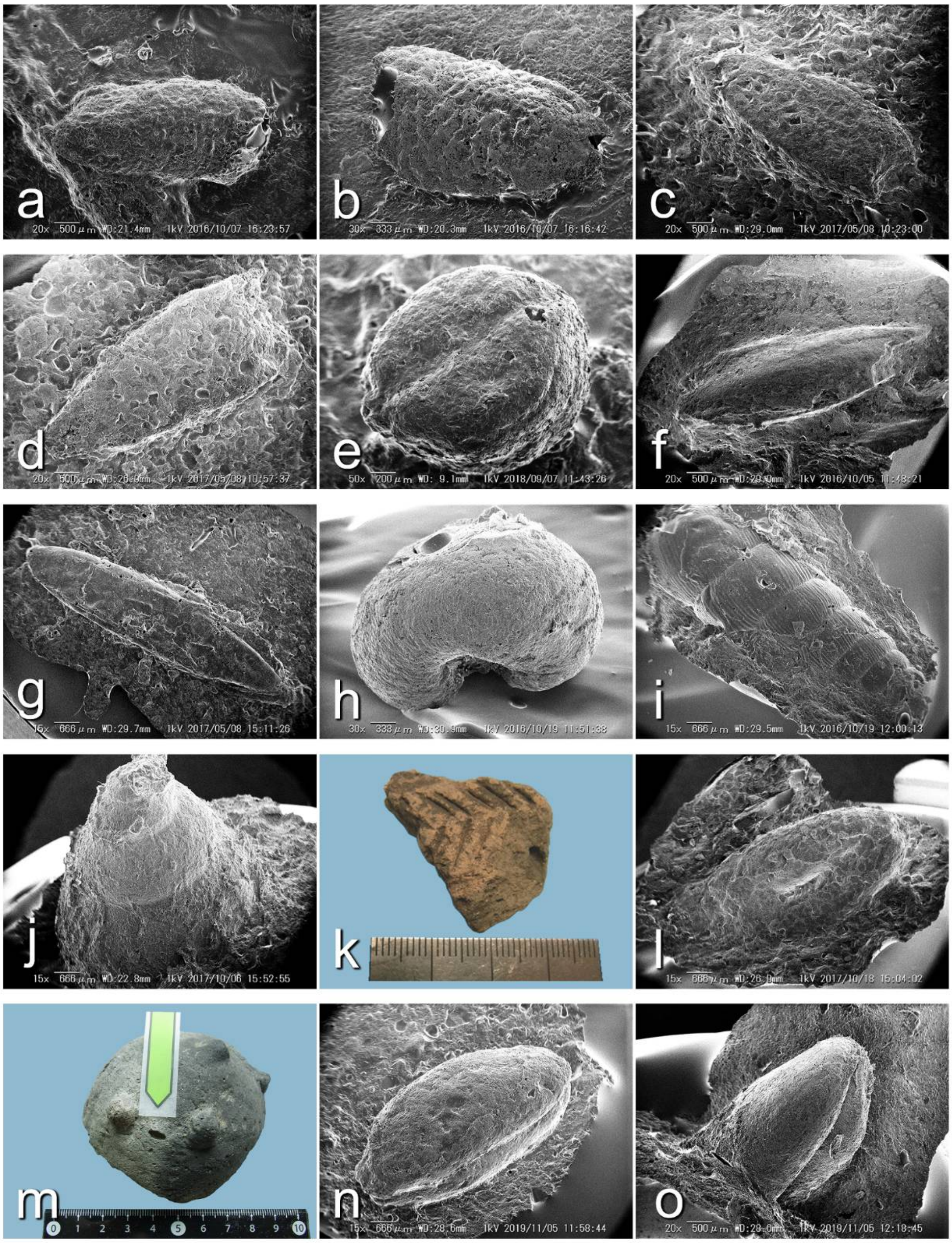

Fig. 4. Images of representative samples and casts obtained from sub-Neolithic (a-j), Neolithic (m-o) and Eneolithic (k, l) pottery from Bazkiv Ostriv (a, b, h, i), Uspenka (c, d), Buzky (e, k, l), Pechera-1 (f), Mykilske-2 (g), Hryni (j), and Hnidava(m-o). Casts are identified to cf. *Sambucus* (a, b), Asteraceae (c, d), Apiaceae (e), Poaceae (f, g), snail specimens (h-j), cf. *Triticum* (l), T. cf. *dicoccom* (n,o).

a height of up to 10 m above the water surface, 11.3 km from the Ratniv-2 site in Volhynia in northwestern Ukraine.

4.2.3. Eneolithic

A total of six *H. vulgare*, one *T. spelta*, three *T.* cf. *spelta*, one *T.* cf. *dicoccum*, two *Triticum* sp., 11 *Hordeum/Triticum*, one *Hordeum/Triticum*?, five Amaranthaceae, one Monocotyledon, and one snail sample were identified from impressions in pottery at 10 Eneolithic sites, with the exception of the abovementioned impression of *Triticum* at the Buzky site (Appendix B: Table B.1).

At the abovementioned Holyshiv-2 site, a multi-stratified settlement representing the Neolithic to Copper Age, one grain of *Hordeum/Triticum* was also identified from a potsherd with a characteristic pattern of the Malice Culture (ca 4800–3900 years BC; Fig. 5a & b). The remaining

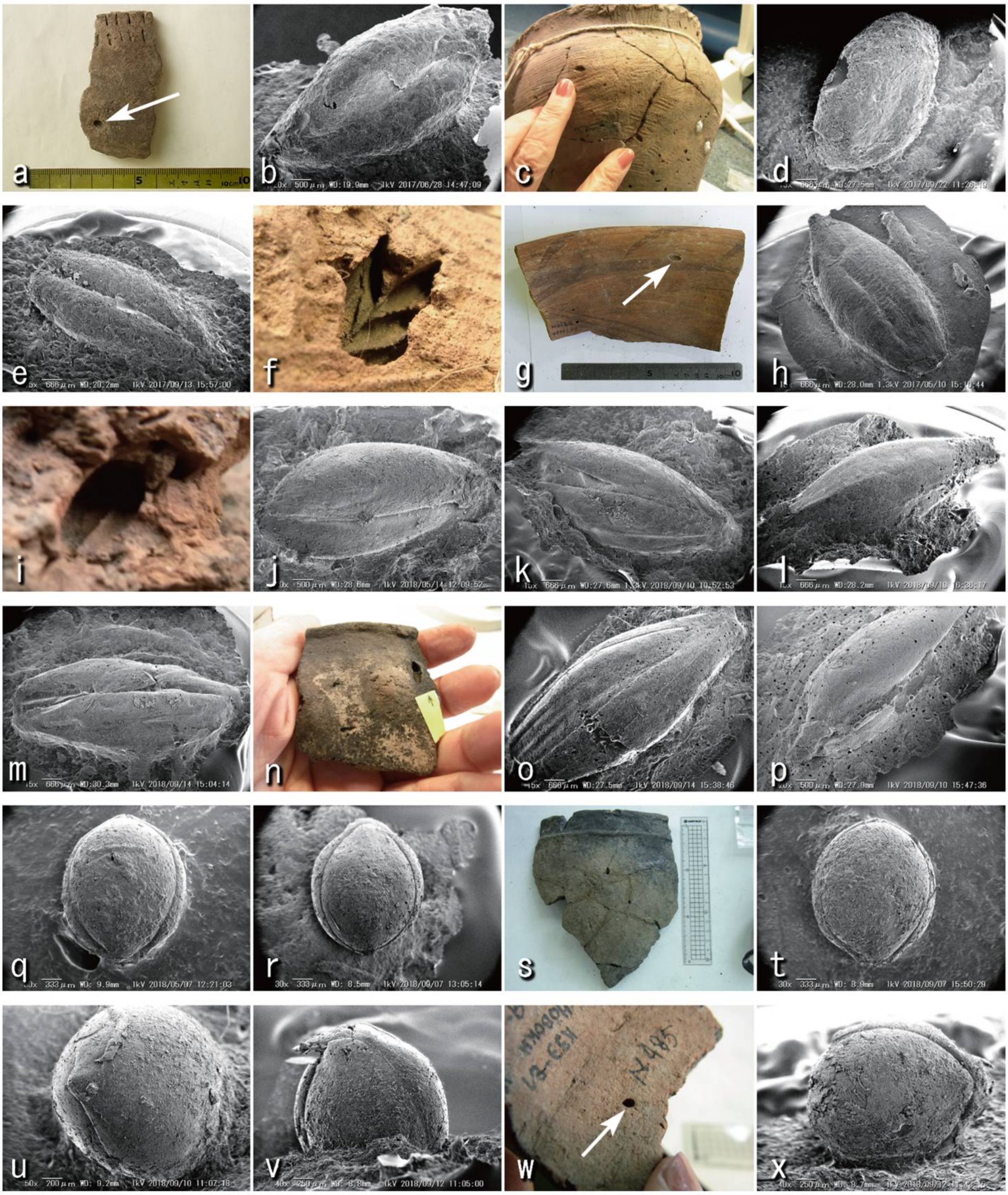

Fig. 5. Images of representative samples and casts obtained from Eneolithic (a-j) and Bronze Age (k-x) pottery, and daub from Holyshiv-2 (a, b), Chapaivka (c-f), Maidanetske (g, h), Maiaky (i, j), and Novokyivka (k-x). Casts are identified to *Hordeum/Triticum* (b), *H. vulgare* (d, h, k-m, o), *Triticum* (e, j, p), *P. miliaceum* (q, r, t-v, x).

Eneolithic cultivated plant casts were obtained from Trypillian pottery.

Among casts from the Chapaivka site (Trypillia BII, 4000–3900 BC) located on top of a loess hill close to the Dnipro River valley in the forest-steppe zone, two *H. vulgare* (Fig. 5d), three *Triticum* (Fig. 5e), and two *Hordeum/Triticum* samples were identified from impressions on the same vessel (Fig. 5c). In addition, a big deep impression thought to be that of an ear of *H. vulgare* (two-rowed barley, hulled; Fig. 5f) was also observed here: however, no cast could be prepared because of the fragility of this impression.

The Maidanetske settlement is a well-known mega-site attributed to the Trypillia CI stage (3800–3700 BC), located on the watershed plateau surrounded by the Talianka River valley and small streams in the forest-steppe zone between the Dnipro and Southern Buh Rivers. A number of charred *Hordeum* and *Triticum* samples were previously reported at this site (Pashkevich and Videiko, 2006, Table 6). Meanwhile, among impressions in potsherds and daub, we identified one grain *of Triticum* sp., two spikelets *of Hordeum vulgare* (one of these is 6- or 2-row, husks removed; Fig. 5g & h), and three samples of *Hordeum/Triticum* chaff.

At the Maiaky site, which is located on the edge of the 3rd loess terrace of the Dnister River estuary in the steppe zone, belonging to both the Trypillia Culture (stage CII) and Usatove Culture dated around 3300–3000 BC, one *Triticum* grain was identified from daub (Fig. 5i & j).

In addition some cereals were identified at the Trypillia A sites of Bernashivka (one *Triticum* and one *H. vulgare* grain) and Hrebeniukiv Yar (one *H. vulgare* grain), the Trypillia BI site of Luka Vrublivetska (one *Hordeum/Triticum* grain), the Trypillia BII to CII site of Kazarovychi (three *Hordeum/Triticum* grains), and the Trypillia CII site of Troianiv (one *Hordeum/Triticum* grain).

4.2.4. Bronze Age

Excluding wild plants, a total of seven *H. vulgare*, three *Triticum* sp., 45 *P. miliaceum*, and 10 cf. *P. miliaceum* samples were identified from impressions in Bronze Age pottery (Table B.1). No *P. miliaceum* was identified from pottery belonging to the Early or Middle Bronze Ages, although numerous samples appeared abruptly from the Late Bronze Age.

The most valuable results were obtained from the Novokyivka site located in the southern Black Sea lowlands to the southward of the Dnipro River lower reaches. Impressions of seven *H. vulgare* (Fig. 5k-o), three *Triticum* sp. (Fig. 5p), and 44 *P. miliaceum* (Fig. 5: q-x) grains with lemma and palea, as well one sample of *Hordeum/Triticum* chaff were identified on pottery from this site. This settlement belongs to the Sabatynivka Culture (Late Bronze Age: 1600–1300/1200 years BC). Some households found here showed archaeological evidence of surface dwellings, with a large number of storage pits (perhaps some for grains), hearths, and stone molds for the production of bronze instruments, all undisturbed by later layers (Gerškovič, 1999: 30-37).

Two vessels and several potsherds from different periods of the Early Iron Age were processed randomly due to an error during the primary identification of ceramics from the multicultural settlements of Sandraki, Yosypivka, and Babyne-3. However, a detailed consideration of the agriculture of this time period is beyond the scope of the present study.

5. Discussion

5.1. The presence of cereals in the sub-Neolithic and Neolithic

In western and southwestern Ukraine, cereals were identified from the Neolithic pottery of LBK, but not from the sub-Neolithic vessels of the Buh-Dniester Culture. Specifically, in this survey, two impressions of wheat grains were identified in the LBK potsherds excavated from the Hnidava site in Volhynia. This is in concordance with previous finds of carbonized grains of barley and wheat in flotation samples from the LBK site of Ratniv-2, located 11.3 km from Hnidava (Motuzaite-Matuzeviciute and Telizhenko, 2016), and carbonized wheat grains from the LBK site of Kamyane-Zavallia site on the Southern Buh River (Salavert et al., 2021). Therefore, our research confirms that cereal cultivation spread to this part of Ukraine with the LBK people in the second half of the 6th millennium BC (Motuzaite-Matuzeviciute and Telizhenko, 2016). In contrast, no cereal impressions were observed in any of potsherds from the sub-Neolithic sites, located in the Southern Buh catchment and to the east of it. Our SEM results suggest that 29 impressions identified previously as cultivated plants using the naked eye (Pashkevich, 2000) are not reliable (Appendix B: Table B.2, Figs. B.1 & B.2). The absence of cereals and pulses is consistent with previous analysis of 4,500 potsherds from 15 sub-Neolithic and Eneolithic sites in eastern Ukraine (Motuzaite-Matuzeviciute, 2012). But in this article of 2012, exceptional pottery shards with impressions of naked and hulled barley were also reported as finds from the layer of the second half of the 5th – the first half of 4th millennium BC at the Zanovskoe site on the Siverskyi Donets River. The authors suppose that these impressions also need to be verified by the new method; however, these crops may indicate a slight interaction with Trypillia Culture during the parallel period. The absence of arable farming activity is indirectly supported by the complete absence of flint blade sections with characteristic gloss (so-called "sickle inserts") in the flint assemblages of the Buh-Dniester, Kyiv-Cherkasy, and Donets cultures as well as in the Crimean sub-Neolithic (Gaskevych, 2003: 13–14). Moreover, the absence of changes in the location of sites of the previously mentioned cultures compared with previous Mesolithic cultures in these regions is also of importance. Bearers of the Mesolithic and sub-Neolithic cultures inhabited small areas of sandy dunes and hillocks directly above the banks of rivers and floodplain lakes, as well as the edges of the low first, occasionally flooded, terraces and islands. They often occupied places with favorable fishing grounds (Appendix A: Table A.1). In contrast, large settlements of agricultural communities, such as Neolithic LBK and Eneolithic Trypillia Culture, tend to be found on high watershed plateaus formed by loess (Appendix A: Table A.1). The presence of a small number of Trypillia materials in multi-layered sites Pechera-1, Bazkiv Ostriv, Melnychna Krucha, Buzky etc. located in river floodplains (Danilenko, 1969; Kotova, 2003) may be explained by seasonal visits of small specialized groups of fishers from large permanent farming villages.

The results are also in good agreement with the results of other natural science analyses. For instance, based on the absence of evidence of caries along with the consistent occurrence of calculus and enamel hypoplasias from burials in Neolithic cemeteries of the Azov-Dnipro Culture in the Dnipro Rapids region (Mykilske, Yasynovatka, and Deriivka), Lillie with co-authors concluded that the diet of the local population during the second half of the 6th to the beginning of the 5th millennium BC was high in protein and, therefore, predominantly meat- and fish-based (Lillie, 1996; Lillie and Richards, 2000; Lillie et al., 2011).

Overall, these findings suggest regional and cultural differences in the establishment of arable agriculture in Ukraine, which was previously thought to date back to the 7th millennium BC (Kotova, 2003). It is evident that the LBK settlements in western Ukraine established arable farming in the latter half of the 6th millennium BC (Motuzaite-Matuzeviciute and Telizhenko, 2016). On the other hand, it is highly likely that agriculture had not been adopted by indigenous groups in areas where the LBK had not spread. Further analysis of each region and each cultural group is therefore important for confirming the establishment of agriculture in Ukraine.

5.2. Arable agriculture in the Eneolithic

Barley, wheat, and even a small amount of *P. miliaceum* have already been detected by the flotation method from the Eneolithic sites mainly from Trypillia Culture (Müller et al., 2017, see their Table 16). On the other hand, from the impression method, cereals of West Asian origin were found in the Eneolithic pottery; however, no millet of East Asian origin was identified. Wheat and barley were detected in pottery of the

Malice Culture and Trypillia Culture, stages A to C (near 5000–3000 BC). No impression of cereals were identified in the Steppe Eneolithic site of Deriivka, although the number of potsherds observed there was perhaps insufficient to draw any definitive conclusions. It is possible that the West Asian model of agriculture spreading across the East Carpathian Mountains, was adopted only by groups of the Trypillia Culture, which were distributed mostly west of the Dnipro River. According to Anthony (2007: 166), "The frontier shifted eastward to the uplands between the Southern Buh and Dnieper rivers. This soon became the most clearly defined, high-contrast cultural frontier in all of Europe." The Dnipro may therefore have been an important boundary of subsistence strategies. However, unfortunately, the total amount of cereal is unknown from the impression data, so it cannot be discussed whether barley and wheat identified from the Tripillian settlements were sufficient as food for the inhabitants of mega-sites.

A possible alternative route of spreading of cereals of West Asian origin into Eastern Europe via Caucasian corridor in Eneolithic was speculated since the recent finds of charred grains of domesticated wheat from the coastal shell midden site of Ardych-Burun in the Crimean peninsula, directly AMS radiocarbon dated to the middle of the 4th millennium BC (Motuzaite-Matuzeviciute et al., 2013b). However, we identified no impressions of cereals in the pottery from the synchronous open-air site of Kukrek, located 34 km northwest of Ardych-Burun. No Chinese millet was identified from our observations of Eneolithic pottery. However, some macrobotanical evidence of *P. miliaceum* has been reported from Trypillian settlements such as Maidanetske, but the possibility of disturbance from younger material was also highlighted (Müller et al., 2017). By newest data, a charred grain of *P. miliaceum* from the Maidanetske site was AMS radiocarbon dated to the Early Middle Age and interpreted to an intrusion (Dal Corso et al., in press).

5.3. The spread of Chinese millet into Ukraine

No *P. miliaceum* was detected from any pottery belonging to the sub-Neolithic, Neolithic, Eneolithic, Early or Middle Bronze Ages. In contrast, at the Novokyivka site, which belongs to the Late Bronze Age (Sabatynivka Culture, roughly 1600–1300/1200 BC), 44 *P. miliaceum* samples with secure botanical characteristics were identified. These findings suggest that the arrival of *P. miliaceum* into Ukraine occurred in the Late Bronze Age. The estimated time range of 1600–1300/1200 BC is also consistent with the previous prediction of around 1500 BC made using an archaeobotanical database of *P. miliaceum* (Stevens et al., 2016). It is also consistent with findings in the surrounding region. In Hungary, for example, carbonized *P. miliaceum* was dated to 1606–1414 BC (Motuzaite-Matuzeviciute et al., 2013a), whereas samples from northwestern Caucasus were directly dated to the 12th–10th centuries BC (Trifonov et al., 2017). In addition, recent extensive programme of AMS-dating of charred broomcorn millet grains from 75 prehistoric sites in Europe revealed that the oldest sample is from the Vynogradnyi Sad site, which is located in the Southern Buh steppe area in southwestern Ukraine and belonged to the Sabatynivka Culture, the same as the Novokyivka site. This sample was directly dated to the 17th–15th century BC (Filipović et al., 2020). Also, the newest AMS dating results of *P. miliaceum* from the Neolithic to the Iron Age in the North Pontic region report the same results as this paper, with the appearance of the millet from the late Bronze Age (Dal Corso et al., in press, 2019).

Wang et al. (2019) predicted three possible dispersal routes for Chinese millet from Central Asia into Europe: the western Steppe, via the Anatolian peninsula toward the Balkans, and northward through the Caucasus Corridor. The Steppe route has been negatively viewed from isotope analysis of human bones in northwestern Kazakhstan (Motuzaite Matuzeviciute et al., 2015). On the other hand, the newly reported results (Martin et al., 2021) supports the northward route through the Caucasus. In addition, finds of Chinese millet in the sites of the Sabatynivka Culture, including Novokyivka, may indirectly support the Balkan route since unlike the Babyne Culture and Srubna Culture, the areas of which covered the territory from Ukraine to Kazakhstan, the Sabatynivka Culture, which extended from the Lower Danube to the Lower Dnipro and some of the Northern Azov region, was historically and economically connected with the farmers and pastoralists in the Balkan Peninsula and the Carpathians/Danube region (Gershkovich, 2003). However, so far neither route has enough data to determine the Chinese millet dispersal into Europe. Further surveys of impressions in pottery of the Bronze Age cultures in the east are required for definitive conclusions on this issue.

It should also be noted that another Chinese millet, *S. italica*, was not identified in our samples. In the archaeobotanical database of *S. italica*, this species is estimated to have arrived in Europe around 500 AD (Stevens et al., 2016). Our results also suggest that its arrival was greatly delayed in comparison with the introduction of *P. miliaceum.* However recently, *S. italica* directly dated to between 2011 and 1771 cal BC was reported in the Caucasus (Martin et al., 2021). Therefore, it is necessary to accumulate similar evidences in the surrounding area, but it will be also necessary to reconsider the arrival timing and intercontinental dispersal route of *S. italica* into Europe. Our improved impression method that can clearly distinguish *P. miliaceum* and *S. italica* will be effective in grasping the trend of two millets in the 3rd-2nd millennium BC in western Eurasia.

6. Conclusions

In this study, we observed 30,753 intact and fragmented pottery artefacts. Silicone casts were taken from 486 possible impressions of plant seeds, then identified 93 grains, three spikelets and five chaff of three species of domestic cereals from 16 sites of different periods.

No cereal was detected from pottery belonged to Sub-Neolithic. Therefore, it is necessary to reconsider the start of arable farming in Ukraine, which was predicted in the 7th millennium BC (Kotova 2003). However, we identified wheat from impressions in the pottery excavated in one site of the LBK that had spread to western Ukraine in the second half of the 5th millennium BC, when the Sub-Neolithic sites existed in territories further east. This is consistent with the result of flotation carried out recently at LBK sites in Ukraine. Also, we have identified wheat and barley originating in West Asia from impressions on pottery of the Eneolithic Trypillia Culture, which settlements have already been reported as yielding a large amount of carbonized wheat and barley grains.

P. miliaceum, which previously was slightly reported from Eneolithic, early and middle Bronze Age by the flotation method as well the clay casts of pottery impressions, was not detected from our silicone casts. Then, in the late Bronze Age, the impressions of the broomcorn millet appeared. The appearance time is also consistent with the direct dating results of carbonized grains. The presence of *P. miliaceum* that was revealed from impressions in this study, at the northern part of the Black Sea during the 2nd millennium BC, can be said to be important data for examining the dispersal route of this millet from Central Asia to Europe.

All the results regarding the timing of appearance of first arable agriculture as well as appearance of broomcorn millet in Ukraine, which we got from the analysis of impression in pottery, are extremely consistent with the dating results of carbonized materials. Thus, the authors believe that the SEM research of silicone casts will be an effective method to fill the gap in areas where macrobotanical data is lacking. Furthermore, it could contribute to the study of East-West food globalization in Eurasia.

CRediT authorship contribution statement

Eiko Endo: Conceptualization, Funding acquisition, Project administration, Investigation, Visualization, Writing – original draft, Writing – review & editing. **Hiroo Nasu:** Investigation, Writing – review & editing. **Dmytro Haskevych:** Conceptualization, Investigation, Resources,

Visualization, Writing – original draft, Writing – review & editing. **Yakiv Gershkovych:** Resources, Writing – original draft. **Mykhailo Videiko:** Investigation, Resources, Writing – original draft. **Olexandr Yanevich:** Conceptualization, Investigation, Resources.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Submission declaration

The authors declare that the work described has not been published previously.

Appendix A. Supplementary data

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