MISCELLANEA GEOGRAPHICA – REGIONAL STUDIES ON DEVELOPMENT
Vol. 27 • No. 3 • 2023 • ISSN: 2084-6118 • DOI: 10.2478/mgrsd-2023-0008

Microscale iron spherules as a trace of metallurgical activity in Old-Polish Industrial District river valleys

Abstract

In the last few centuries, a metallurgical industry based on forges driven by water wheels developed in many European valleys. One such area is the Old-Polish Industrial District (OPID) in Central Poland, which is one of the largest of this type of industrial center. Metallurgical activity developed here from the Prehistoric to modern times. The transformation of metallurgical technology led to the collapse of production, and the ongoing renaturation obliterated most of the traces of former industrial activity. The Magnetic Spherule Separation method used so far in Western Europe, and recently in Poland, has been able to detect traces of former metallurgical activity preserved in alluvium. Fluvial deposits contain microscopic, perfectly spherical iron hammerscales formed during metallurgical production. The results of the study of the alluvium of selected rivers in the OPID indicate the presence of iron spherules of various origin and facies in the sediments of the floodplain, which accumulated during the period of metallurgical activity and were redeposited in modern times. This allows us to estimate, among other things, the age, rate of accumulation and impact of anthropopressure on sedimentation conditions.

Keywords

Old-Polish industrial district • magnetic spherule separation method • holy cross mountains • flood plain sedimentation

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Received: 8 May 2023
Accepted: 31 July 2023

Introduction

In the 21st century, much attention is being paid to the anthropogenic transformation of fluvial systems (Kalicki 2006; Xing et al. 2014; Gibling 2018; Elznicova et al. 2021), especially in intensively industrialized areas in the past or present, such as North Rhine-Westphalia and Benelux (Quik et al. 2020), the Po River basin (Brandolini & Cremaschi 2018), some sections of large river valleys in the USA (Skalak et al. 2013), or the Upper Silesian and Old-Polish Industrial District (OPID) (Przepióra et al. 2016; Rutkiewicz & Malik 2018; Kalicki et al. 2019a, 2020; Fularczyk et al. 2020; Przepióra 2021). Increasingly, the scale of anthropogenic pressure and its impact on fluvial processes, as well as the structure and sediments of floodplains, are being assessed through the prospect of the occurrence of various objects and pollutants of anthropogenic origin in sediments (Kalicki et al. 2019a, 2020; Rutkiewicz et al. 2019; Houbrechts et al. 2020; Hrubý et al. 2021; Kiss et al. 2021; Przepióra et al. 2021, 2022).

The Holy Cross Mountains region (Richling et al. 2021) provides a wide range of opportunities to conduct this type of research, as mining and iron smelting have been developing here for over 2,000 years (Bielenin 1992). This activity was concentrated in river valleys, and directly or indirectly affected the fluvial environment. In the last few centuries, the OPID developed here, where many factories used water energy, and the valley bottoms were zones of accumulation of various objects related to industrial activity in sediments of the channel and overbank facies (Kalicki et al. 2019a; Przepióra 2021).

The studies of the alluvium of the Holy Cross Mountains region, carried out since 2019, allowed the occurrence of iron spherules in these sediments to be identified, both in-situ and redeposited and in the vicinity of OPID metallurgical centers (Kalicki et al. 2021b; Przepióra et al. 2023).

Study areas

The research was carried out on sediments from five sites located in valleys of third-order rivers, draining the area of the OPID in the Polish Uplands (Richling et al. 2021) (Holy Cross Mountains region) (Fig. 1). The material of various facies from the fluvial and limnic sedimentary environment was analyzed, including channel and overbank alluvium, lacustrine and subaqueous fan deposits. This material accumulated near metallurgical plants that functioned in historical times.

Aim of study and methods

The aim of this study was to identify former metallurgical activity traces in alluvia of the OPID using sedimentological methods and archival data. The presented and interpreted results were obtained using the Magnetic Spherule Separation (MSS) method, employed in selected river valleys of the Holy Cross Mountains area since 2019. The study identifies the relationship between the sedimentary environment, which contains iron microspherules (hammerscales) (Fig. 2) in different facies of alluvia, and deposits of other origin in the valley bottom.

The field work consisted of making open pits and boreholes using an Eijkelkamp hand drilling rig and collecting samples for laboratory analysis.

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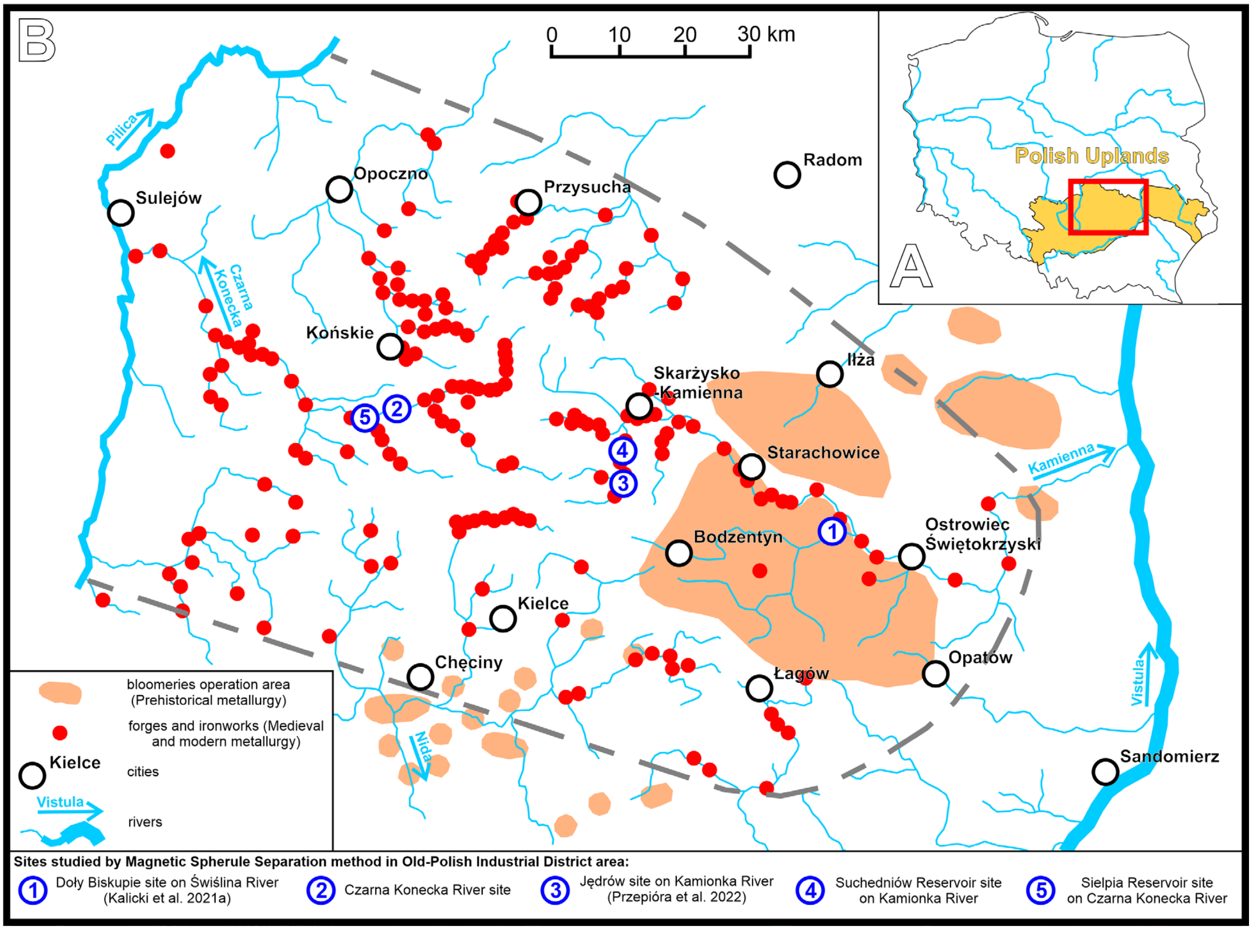

Figure 1. Study area (A) on Polish Uplands (according to Richling et al. 2021) and study sites location (B) within OPID area (according to Bielenin 1992; dashed line)
Source: after P. Przepióra & P. Kusztal

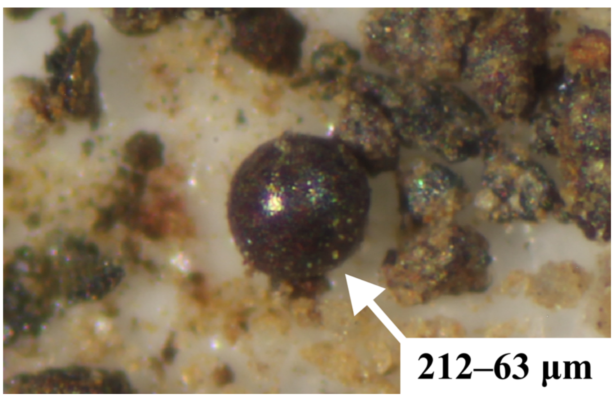

Figure 2. Iron microspherule separated from alluvia (photo P. Przepióra 2019)

The grain size analyses were done using the sieve method (size 2800–63 μm) and laser diffraction (fine fraction) with a Mastersizer 3000 particle size analyzer; $CaCO_3$ content was calculated using the Scheibler method, and pH with the ELMETRON CP-411. The separation of ferromagnetic elements from alluvia was made using the modified MSS method (Richardeau 1977; Houbrechts et al. 2020; Przepióra et al. 2022).

Grain size results with Folk-Ward's (1957) statistic parameters were generated in the GRANULOM program. In the selected sites, the obtained results are presented by the cross-sections. The Digital Elevation Models (DEM) were developed based on LIDAR data from geoportal.gov.pl. A query of archival materials was also carried out, using, among other things, historical (Bielenin 1992) and cartographic records included in digital databases (Maps Arcanum 2023; Mapster 2023; PGI-NRI 2023).

Results

Fluvial study site—Czarna Konecka River valley

The oxbow lake near Piekło is located in the W part of the OPID (Fig. 1), in the middle of Czarna Konecka valley, which is about 1.5 km upstream of Sielpia Reservoir—the local erosion base. As indicated by the cartographic data, the chute cutoff of this bend took place before 1974. At that time, a new channel was created, which initially formed a two-riverbed system with the cutoff meander. From the turn of the 20th/21st century, after the formation of the neck in the inlet part of the oxbow lake and its final cutoff, the new channel functioned independently as a modern riverbed (Fig. 3A).

Two geological profiles were made at the site: Z10 on point bar of abandoned channel; and Z4 into the inlet section of the abandoned channel (Fig. 3A).

Two series in the Z10 profile can be distinguished (Fig. 3B). There are two phases of channel sediment accumulation,

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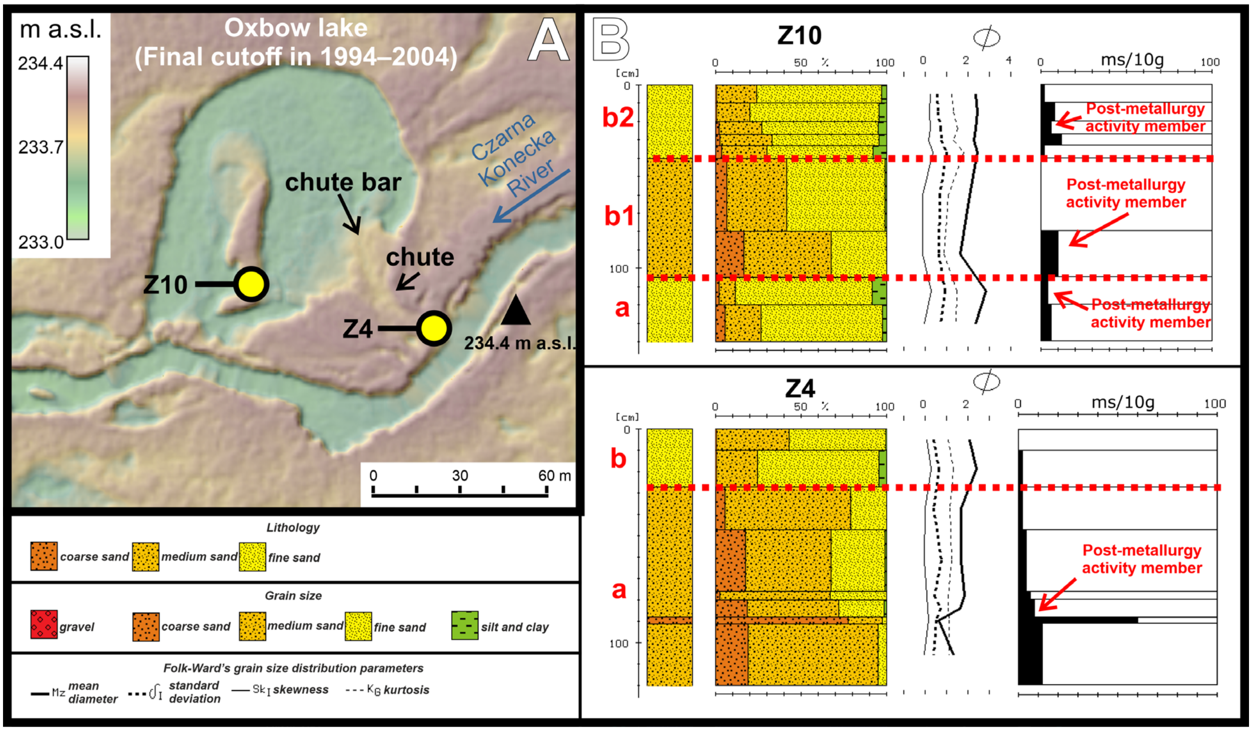

Figure 3. Location of study profiles (A) on the DEM (geoportal.gov.pl) and results of grain size analysis, Folk-Ward's (1957) grain size parameters and microspherule concentration (B)
Source: by K. Fularczyk

probably connected to lateral channel migration and the formation of inclined bedding typical of such river shifting and simultaneous aggradation. The lower unit (a) (140–105 cm) is moderately sorted (σ_I=0.7–0.9) fine sands with (2–9%) silt and clay content (Mz=2.3–2.9ø) and represents the highest part of the point bar facies and therefore has an admixture of a finer fraction. Above this (105–0 cm), there is a second channel sediments unit (b) with fining upward sequence (Mz=1.7–2.4ø), typical for alluvium of meandering rivers. There are two facies of the lower (member b1: 105–40 cm) and upper (member b2: 40–0 cm) point bar. These are slightly better sorted (σ_I=0.6–0.9) mixed and fine sands (b1) with an admixture (<10%) of silts and clays at a depth of 80–0 cm (b2). The alluvium of both units is contaminated by microspherules accumulated in three phases. Such objects were not found exclusively in the layer (80–40 cm) of the lower point bar facies (Fig. 3B).

The Z4 profile is represented by channel sediments of neck in the inflow to the oxbow lake accumulated in two phases (Fig. 3A). In the first stage of its fill (a) there are mostly medium sands (Mz=1.7–1.8ø) with a layer of coarse sands at a depth of 90 cm (Mz=0.8ø). These sediments are well and moderately well sorted (σ_I=0.4–0.7). The oxbow lake was built up with finer channel sediments, in the second stage of fill (b), by well and moderately well sorted (σ_I=0.4–0.6) fine sands (Mz=2.0–2.4ø). The alluvium of both members is contaminated by microspherules accumulated in four phases. Such objects were not found exclusively in the topmost layer (10–0 cm) of the upper member (b), whereas their highest concentration was noted in the layer (a) of coarse sands (Fig. 3B).

Lacustrine study site—Suchedniów Reservoir

This site is located in the NW part of the Suchedniów Reservoir, rebuilt on the central Kamionka River in 1974. Outcrops of bottom sediments were made during hydrotechnical works (dredging) in 2017 (Przepióra et al. 2019) (Fig. 4A). In the 140 cm outcrop, 3 accumulation units are clearly visible (Fig. 4B).

At a depth of 140–120 cm (Fig. 4C), dark, sandy-silty sediments with an admixture of gravel and alternating layers of detritus represent the member of the former industrial pond, in operation from the turn of the 19th/20th century (Fig. 4A). The Mz is 1.9–2.8ø, while the sorting ranges from moderately good to very poor (σ_I=0.5–2.6). The content of $CaCO_3$ is low (0.5–1.25%) and the pH reaches 4.7–5.9 (Przepióra et al. 2019). In these sediments, from 1 to 6 microspherules/g were detected, alternating with layers in which they did not occur.

The middle member, at a depth of 120–50 cm (Fig. 4C), represents the sandy-gravel alluvium of megaripple marks, which was formed during the catastrophic flash flood in 1974, resulting from the dam rupture (Fig. 4B). The Mz of these sediments is 1.3–2.6ø, while the sorting is moderately good and moderate (σ_I=0.6–0.8). The $CaCO_3$ content is about 0.5%, and the pH range is 5.3–5.7 (Przepióra et al. 2019). Only 1–3 microspherules/g of sample were detected.

The upper member is represented by modern reservoir (since 1974) lacustrine sediments (Fig. 4A). There are silty sands with single gravels (Fig. 4C). The share of clayey-silty sediments increases upwards from 8 to 30%. The Mz of these sediments is 1.5–2.8ø, while the sorting is poor and very poor (σ_I=1.1–2.4). The $CaCO_3$ content is 0.7–0.9%, and the pH is 4.7–5.5 (Przepióra et al. 2019). Also, in these sediments, microspherules were detected, at 1–3 microspherules/g.

The low content of $CaCO_3$ in the profile is related to the geological structure of the area, while the acidity of the sediments indicates a concentration of pollutants present related to human activity in the river catchment (Kalicki et al. 2021a).

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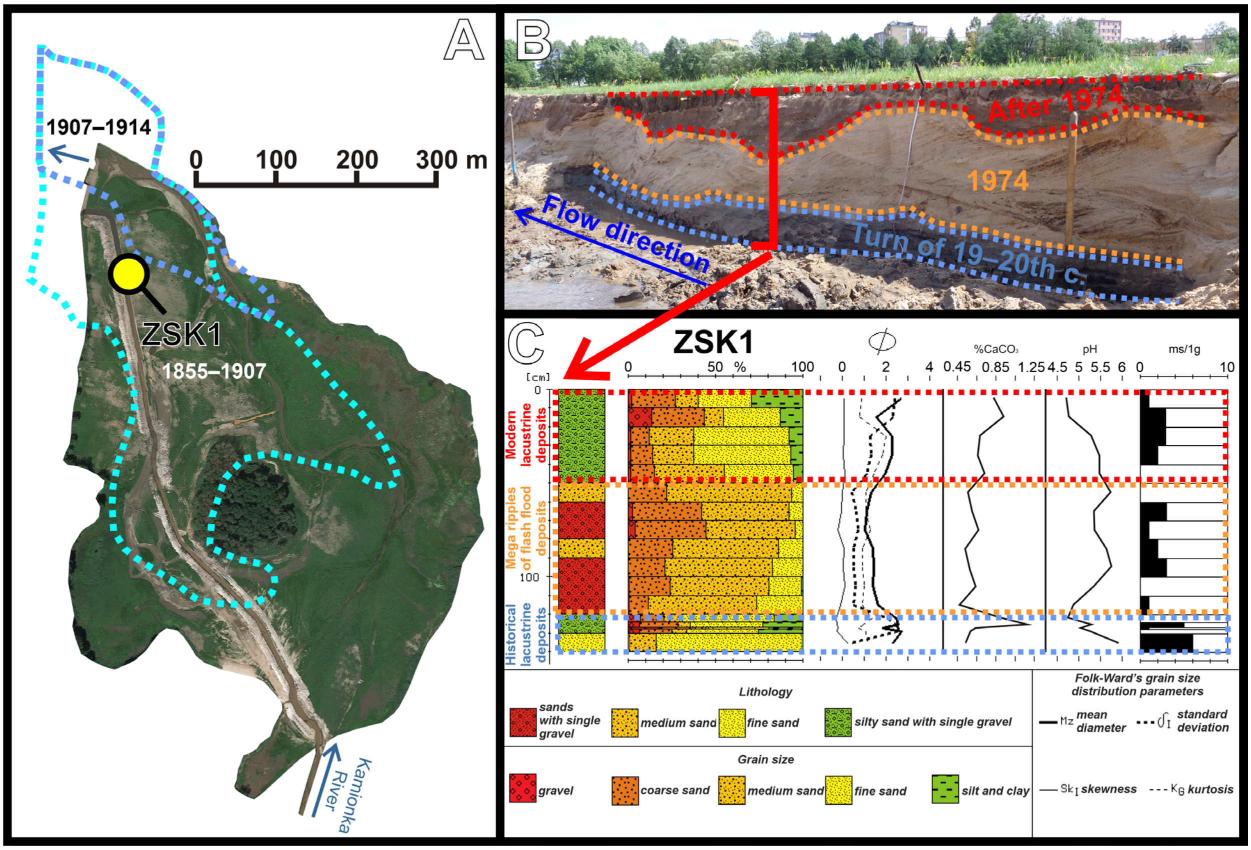

Figure 4. ZSK1 profile location on the actual Suchedniów Reservoir bottom (Google Maps 2023) with historical borders of previous ponds (A), view of the outcrop (photo P. Przepióra 2017) with distinguished members and their age (B), grain size, Folk-Ward's (1957) grain size parameters, geochemical measurements and microspherule concentration in deposits (C)
source: by P. Przepióra

Lacustrine study site—Sielpia Reservoir

This site is located in the middle section of the Czarna Konecka River that flows into the Sielpia Reservoir and forms a high-energy subaqueous alluvial fan (delta)(Fig. 5A). During hydrotechnical works, the field research was carried out in 2019–2020 on the reservoir and incised by the river inland delta. Profiles of 70–90 cm depth were made along the delta to create a longitudinal section A–A' (Fig. 5B) (Przepióra et al. 2021).

The delta is built by well-sorted to poorly sorted (σ_1=0.4–1.8) sediments, mostly sands with single gravel (Mz=0.5–0.8ø), medium sand (Mz=1.0–1.3ø), and silty sands with gravel (Mz=2.3–2.8ø). Among the coarse clastic sediments, there are numerous slag fragments redeposited from the upper section of the river. The front of the delta covers a lacustrine sediment built by a darker layer of finer deposits with detritus layers. In a longitudinal section, three accumulation phases are marked with one lacustrine sediment (I) and two phases (II, III) of delta deposition (Fig. 5B) (Przepióra et al. 2021).

Only a small amount of iron microspherules in several samples of 10–20 g weight was found (Fig. 5B). A very small number of detected microspherules (1–3 in the sample) made it possible to draw a subjective member of their accumulation in the delta sediments (30–60 cm depth). Single spherules also appear in upper part sediments (5 cm depth). The number of spherules is highest in the lower part of the DSW6P profile, covering part of the lacustrine sediments (depth 60–90 cm). A visible level of microspherules deposition coincides mainly with the lithological accumulation phase II and some in phase I. The greatest number of iron spherules was detected in the top of lacustrine deposits and in the bottom of delta sediments (Przepióra et al. 2021).

Discussion

Iron microspherules can be a product of natural phenomena occurring on the Earth (Stankowski et al. 2002); however, the documented intensive metallurgical activity in the OPID area (Bielenin 1992) suggests their anthropogenic origin in the studied sites. They are a product of iron smelting and forging in metallurgical facilities (Dungworth & Wilkes 2007). They occur in various facies of alluvia and sediments of different origin accumulated in valley bottoms.

In the previous studies in Świślina valley (Kalicki et al. 2021b) (Fig. 1), microspherules were detected in the overbank sediments of the Holocene lower floodplain. This is mainly composed of sandy silts, also referred to by Klatka (1958) as anthropogenic muds. There is no historical data on the activity of the Medieval and modern iron smelting and processing plants in the vicinity of the site and in the river basin. However, many of the Prehistoric bloomery traces were found in the Świślina catchment (Orzechowski 2007). The microspherules occur in the study profile only above the flood sandy layer and probably represent the intensive activity of the Prehistoric smelting in the entire catchment (Fig. 1). This material was redeposited by the river in the last two millennia. Recently, its redeposition may have also occurred during catastrophic events—namely, the flash flood after the dam rupture in 2001 (Ciupa 2012) or during previous similar events on the river in the last millennium (Kalicki et al. 2021b).

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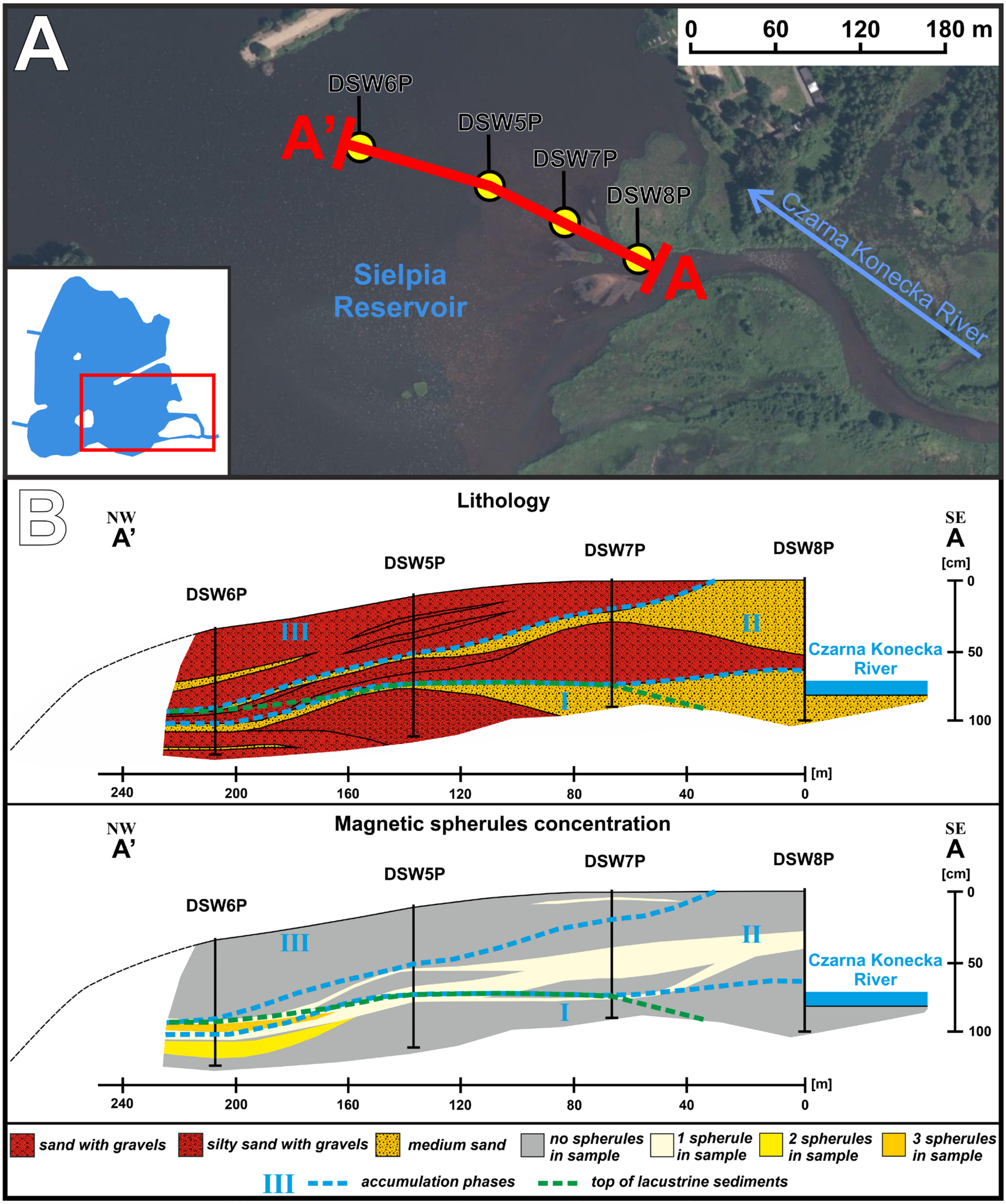

Figure 5. Study profiles and A–A' section across the Sielpia Reservoir bottom location (Google Maps 2023) (A), lithology and microspherule concentration in the deposits (B)
source: by P. Przepióra

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In the Czarna Konecka valley (Fig. 3), the Medieval and modern metallurgical activity was dominant. There were many forges on the river, on which industrial ponds were built (Fig. 1), forming an anthropogenic small-scale water retention system (Kalicki et al. 2020). The channel regulation and the subsequent industry decline led to further changes in the sediments delivered to the river. Iron spherules, accumulated in the form of several layers, were detected in both profiles made on the point bar (Z10) and abandoned channel fill (Z4) (Fig. 3). In profile Z4, a noticeable increase in the number of microspherules may be related to the former forge pond dam rupture at Małachów, located 4.5 km upstream (Kalicki et al. 2019b). The other overlying members represent their subsequent redeposition. Archival and cartographic data (Maps Arcanum 2023; Mapster 2023; PGI-NRI 2023) documented the palaeomeander cutoff at the turn of the 20th/21st century. Microspherules from the Czarna Konecka River were detected in smaller amounts in younger channel sediments, which clearly indicates their multiple redeposition, similar to the Kamionka valley (Przepióra et al. 2022). The youngest alluvia can be dated with Pb and Cs (conclusive in terms of redeposition), while so far this kind of sediment has been dated using, among others, the OSL method in the Kamionka valley (Przepióra et al. 2022).

Sediment fill of the Suchedniów Reservoir is characterized by a large diversity of the sedimentary environment (Fig. 4), including two members of lacustrine deposits and flash flood deposits, forming sandy-gravel megaripples. The occurrence of microspherules in the former pond of lacustrine sediments indicates that they were accumulated during the nearby forge activity, which, until the end of the 19th century, was located approximately 50 m westward of the current reservoir dam (Przepióra et al. 2019). Their presence in the flash flood deposits (dam rupture in 1974) and the present lacustrine sediments indicates their redeposition from the upstream part of the catchment, where at least 5 forges also operated until the end of the 19th century (Bielenin 1992).

The Sielpia Reservoir had already been dredged in the 20th and 21st centuries, which was confirmed by archival documentation and maps (Maps Arcanum 2023; Mapster 2023; PGI-NRI 2023). The reason for the formation of a high-energy subaqueous fan at the confluence of the Czarna Konecka into the reservoir (Fig. 5) was catastrophic flash floods caused by failures of dams in the upstream section (Kalicki et al. 2019b). The small number of microspherules present in its sediments was probably caused by washing out and their further redeposition. Also, the accumulation was during the flood of sands washed out from the Pleistocene terraces and the Holocene floodplain alluvia with subfossil trunks formed during the Subatlantic (Kalicki et al. 2021c), and by bottom erosion, in which microspherules do not occur. The iron spherules form a layer that goes deeper towards the front of the fan. It buried the lacustrine sediments of the modern reservoir, where the largest number of microspherules was detected. As in the Suchedniów case (Fig. 4), the spherules were accumulated in the sedimentation basin of the Sielpia Reservoir. The connection of sedimentological data with historical, cartographic, archival and media information is confirmed by the very young age of these sediments (21st century) (Przepióra et al. 2021).

Conclusions

The MSS method can be successfully applied in places in Central and Western Europe (the same as in Belgian Wallonia), where the Prehistoric and historical metallurgical activities developed. A slightly modified MSS method can be used for the deposits of a different origin (e.g., fluvial and lacustrine), different facies (channel, overbank) of floodplain alluvia, as well as for deposits of different sedimentation type by secular or catastrophic processes. The MSS method can be used to detect the Prehistoric, Medieval and modern (forges and blast furnaces) smelting activities in various sediment facies. Redeposited microspherules are good determinants of smelting activities in the sediments of the OPID river valleys. The obtained data gains new interpretation possibilities based on the results of the MSS method; for example, they show the rate of sediment accumulation and its relative age. Careful interpretation of the reasons for the occurrence of single microspherules in sediments should include their redeposition, bioturbation, and possible formation as a result of a meteorite impact. Historical data describing the time of operation of the forges and blast furnaces on the OPID rivers allow for the potential dating of iron microspherules in alluvia by connecting their presence with increased metallurgical activity periods, or floods related to the dam ruptures of former industrial ponds.

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Vol. 27 • No. 3 • 2023 • ISSN: 2084-6118 • DOI: 10.2478/mgrsd-2023-0008

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