



Oligomictic alluvial aggregates: petro-mineralogical and geochemical evaluation of sandy gravel formations on the middle course of the Danube (Hungary)

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Abstract

This paper evaluates the applicability of Hungarian oligomictic alluvial gravel formations at two sections of the Danube river for construction use. The classification of these aggregates is more challenging than monomictic rock aggregates due to their heterogeneous nature. Multi-source clastic sediments cannot be characterized by single values of physical properties but only a distribution, and they are generally less predictable than monomictic materials. The possible applications in concrete manufacturing were evaluated by complex macro-microscopic petrographic, heavy mineral and nuclear analytical geochemical investigations. The two regions falling on the middle course of the Danube have similar sandy gravel formations, applicable as aggregates for conventional and unconventional concrete building purposes or road construction. Their utilization in open-air constructions might be problematic due to their probable alkali-silica reactivity.

Keywords Multi-source aggregates · Petrographic evaluation · Heavy minerals · Nuclear analytical techniques · Prompt gamma activation analysis · Neutron activation analysis

Introduction

Being abundant at the surface or near-surface localities, clastic sediments are a common subject of raw material exploitation worldwide. Sandy gravel formations are fundamental raw material sources of the construction industry, especially in concrete manufacturing as aggregates. The applicability of these clastic sediments is determined by physical characteristics that affect their durability (against freeze-thaw processes, chemical agents, heat, mechanical stress). Concretes for different purposes require raw materials with specific properties (e.g. high compressive strength for basement constructions, ionizing-radiation resistivity for biological shielding materials) (e.g. Alexander and Mindess 2005).

To qualify clastic raw materials for concrete industrial purposes, international standards (ASTM C 295 2019; DIN

EN 932 1996; DIN EN 933 2015) have been established, applying conventional mineralogical and petrographic methods. In addition, granulometry and grain shape analysis are also fundamental properties to be determined and described in standards (ASTM D 4791 (2019) for coarse aggregate; CRD-C-120 (1994) for fine aggregate; ASTM C 295 (2019) and ASTM D 3398 (2000) for shape analysis).

Aggregates have to meet numerous specifications to become suitable to concrete production (Berubé 2001). One of them is the characterization of the grain population (e.g. size distribution, gradation, grain shape, surface texture). Another aspect is the overall composition (mineralogical and/or chemical) which specifies the presence of the harmful substances in the aggregate (e.g. clay minerals, organic matter, micas, iron- and manganese-oxide staining, fine-grained siliceous material, alteration or weathering in the form of incrustation on grains). Specific characteristics can be set for the surface moisture or water content, absorptivity, specific gravity, bulk density, or void content. Such properties ultimately determine the mechanical performance (resistance against fragmentation, abrasion or polishing), durability (resistance against environmental conditions) and chemical stability (resistance against deleterious chemical reactions including alkali-aggregate reactivity) of the

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concrete. Durability and mechanical resistivity are defined by European Standard test methods (e.g. EN 1097-1 2003; EN 1097-2 2006; EN 1097-9 2005; Erichsen et al. 2011) and affected by initial weathering state of the aggregates and environmental (i.e. climatic and anthropogenic) conditions (e.g. EN 1367-1 2007; EN 1367-2 2010; Šachlová et al. 2016, 2017; Czinder and Török 2019).

Chemical stability belongs to one of the most desirable aspects of aggregates when used as fillers in concrete. From the range of different deleterious chemical reactions, alkali silica reactivity of aggregates is among the most widely studied (Fernandes and Broekmans 2013). This chemical reaction takes place between the alkalis of the pore solutions (cement paste) and certain forms of silica in the aggregates inside the concrete (Stanton 1940; Swamy 1992; Dron and Brivot 1992, 1993; Fournier and Bérubé 2000). Volumetric expansion due to the formation of alkali silica gel presents a harmful process influencing the integrity of concrete structure. Prediction of chemical stability of aggregates in concrete is preferentially done by detailed petrographic examination (e.g. West 1994; Ramachandran and Beaudoin 2000; Šachlová et al. 2016, 2017; Kuchařová et al. 2016; Ahmad et al. 2018) or by well-established physical tests (e.g. ASTM C 1260 2014, RILEM AAR-1,-2 2016). Furthermore, ionizing radiations can induce or accelerate the deterioration of concrete via alkali-silica reaction even if the aggregates were inert to alkali before the irradiation (Ichikawa and Koizumi 2002, 2007). Such damage mechanisms in concrete may be more pronounced in an environment with elevated temperature and humidity. In Hungary, ASR-susceptible rocks are amorphous to weakly crystalline silica containing (volcanic rocks) or deformed quartz containing rocks (quartzite, sand, gravel, metavolcanites) (Fernandes et al. 2018).

There is an elaborate correlation between the petrographic and technical properties of aggregates (Johansson et al. 2015). Multivariate statistical analysis revealed that the mean grain size (on average and for certain mineral grains), the grain-size distribution, the sensitive-mineral content (e.g. mica) and the frequency of microcracks are the main factors that influence the technical properties. Since aggregates are exposed to intensive mechanical, physical and chemical stresses during the service life of the concrete, these clastic materials have to be resistant. All abovementioned properties and requirements of aggregates are fundamentally determined by the mineralogical composition, the fabric and the structure of the material. Thus, the petrographic investigation of aggregates provides a solid basis for the assessment of their performances in a concrete structure.

Polymictic alluvial clastic aggregates differ from monomictic rock aggregates exploited in quarries for construction purposes. That justifies a comprehensive analysis with a special emphasis on the petrographic-geochemical examinations. The study of pebble lithology and the composition of fine particles and heavy minerals are traditional methods in

provenance analysis (Dickinson 1985; Pettijohn et al. 1987; Lihou and Mange-Rajetzky 1996). The size, lithology and maturity of the sediment (i.e. ultimate survival of various lithologies) are determined by the lithology of the source area, the rate and distance of transport and the degree of recycling and weathering. Local outcrops, tributaries to the mainstream and re-deposition from older strata may contribute to the clast population and the mixing along the fluvial course, resulting in multi-cycle clast populations (e.g. Lindsey et al. 2007).

Although potential of freshly exploited monomictic (e.g. andesite) aggregates is intensively investigated (Török and Czinder 2017; Czinder and Török 2019, 2020), there are several extended alluvial sandy gravel formations in Hungary that are dominantly exploited for construction purposes. This phenomenon is characteristically different from the Central European practice and is due to the geological-geographic situation of Hungary being a great sedimentary basin of Danube. This study focuses on two regions of the middle course of the Danube (the main river of the country and the Carpathian Basin): the NW Hungarian and the Mid-Danubian regions. The detailed mineralogical-petrographic and geochemical characterization of different fractions of the clastic sediments provides a solid scientific evidence to classify them according to their industrial applicability. This paper aims to provide a detailed geological description of sand-to-gravel fractions of the NW Hungarian and the Mid-Danubian regions by microscopic petrographic, heavy mineral and bulk geochemical investigations.

Until now, such systematic and comprehensive information was not available concerning Hungarian gravel mining sites. Our investigation does not aim to make a provenance study but to compile the micro-scale characteristics that are relevant to the overall physical properties of the aggregates. Using these data, the applicability of clastic sediments for construction becomes predictable.

Materials and geological setting

The Danube enters the Carpathian Basin at Dévény (Devin) tight where the middle river course type with a high sediment-deposition rate is typical. The geology of the catchment area of the river is varied; thus, varied petrographic composition of the clastic sediments can be expected parallel with high ratio of coarse-grained fractions and moderate (re)working of the grains. In addition to the bulk transported by the Danube from the west of Dévény, the average petrographic composition has been affected by the tributaries inside the basin, as well as the local geology. For instance, in the central part of the Carpathian Basin, the alluvial sediments are supplemented by Miocene volcanic input from the close mountainous area of Visegrád-Börzsöny (Karátson et al. 2000, 2007).

At the NW Hungarian region, the Danube enters from the elevated mountainous background (breaking through the

Hainburg Block and the Little Carpathians) to a depressing/subsiding basin where the river deposits most of its sediments, and especially its coarse-grained sediments, forming a huge alluvial cone (Franyó 1967; Hajósy et al. 1993). The depression is filled with over 2000 m of Pannonian marine mainly sandy-silty and 100–250 m (max. 700 m) of Quaternary fluvial sandy-gravelly sediments. The Quaternary sedimentation was determined by the tectonic activity of the Rába lineament which divided the homogeneous Pleistocene alluvial cone into two separate regions: a continuously depressing basin and an elevated terrace. The Holocene alluvial sedimentation is partly driven also by the transportation of smaller rivers (Rába, Marcal, Rábca, Répce) which — just like the relevant part of the Danube river — provided a low energy depositional environment (slow and shallow river beds, abandoned meanders). The NW Hungarian region of the Danube is characterized both by middle course type meandering beds (Moson-Danube) and lower course type braided channels (Danube major bed). The sedimentation is mainly happened in the major braided channel, while the minor meandering channels helped the lateral distribution of the sediments. The Holocene gravelly formations show decreasing average grain size from west to east (on a 50 km long distance it can decrease from 50–70 mm to 25–35 mm) (Szádeczky-Kardoss 1938; Károlyi 1957; Pécsi 1962). Although the gravelly sediments are fauna-free which makes the litho-chronostratigraphic classification difficult, they are divided into three main members (separated by erosional and tectonic discordances) (Jaskó 1990). The upper member is an Upper Pleistocene-Holocene gravel, gravelly sand formation (with subordinate fine sediments on its top) of the Marcal, Rába and Répce rivers deposited in shallow depressions (10–50 m thickness). The lower member is limited to tectonic trenches but even with 100 m thickness. The cross-bedded sand- to siltstone member is

a fluvial fine-grained formation forming the footwall of the lower member (its thickness is some hundreds of meters in the center of the basin while it is thin on the rims). The upper member can be divided into the silty clayey materials of the Upper Holocene low floodplain, sand and sandy gravel of the Lower Holocene high floodplain and the sandy gravel sediments of the Upper Pleistocene terrace (Kaiser et al. 1998; Jánossy and Krolopp 1994).

At the Mid-Danubian region, the first (Upper Pleistocene-Holocene) and second (a and b, Upper Pleistocene) terraces of the Danube are extended (Burján 2002). The sediments (dominantly 12–16-mm-sized fine gravel) of the terraces 2a and 2b are well-sorted, characterized by distribution curves with two maxima (0.4 and 8–16 mm peak values). Sediments of terrace 2b (medium-coarse-grained sand with gravels) are slightly coarser than those of terrace 2a (fine-medium-coarse-grained sand with gravels). Sediments of the first terrace are located at the flood plain of the Danube (20–25 km wide around the riverbed in this region) (Molnár 1995). Its basement is Pannonian clay-sand formation (Bulla 1941; Pécsi 1959; Erdélyi 1967; Kriván 1960; Sümeghy 1952, 1953). The first terrace sediments are well-sorted and two maxima distribution curves (0.4 and 32–40 mm maximum values) are typical. According to previous research, Danube clastic sediments on the Hungarian course have well-rounded and medium spherical shapes (Pécsiné Donát 1958).

Our study focuses on two selected sections of the river course (Fig. 1). The NW Hungarian region involves the surroundings of Győr with four selected gravel pits (abbreviated as NW-1–4). The Mid-Danubian region is situated to the south from Budapest with four mining sites (abbreviated as MD1–4). All pits operate with shore excavation or bed dredging (Fig. 2). The gravels are washed with the lake water and assorted into four grain-size fractions.

Fig. 1 Simplified geological map of the two investigated regions with the differentiation of Holocene floodplain levels and Pleistocene low-high terraces (or older sediments). The positions of the sampled open-air sandy gravel quarries in the NW Hungarian and the Mid-Danubian regions are indicated with black dots

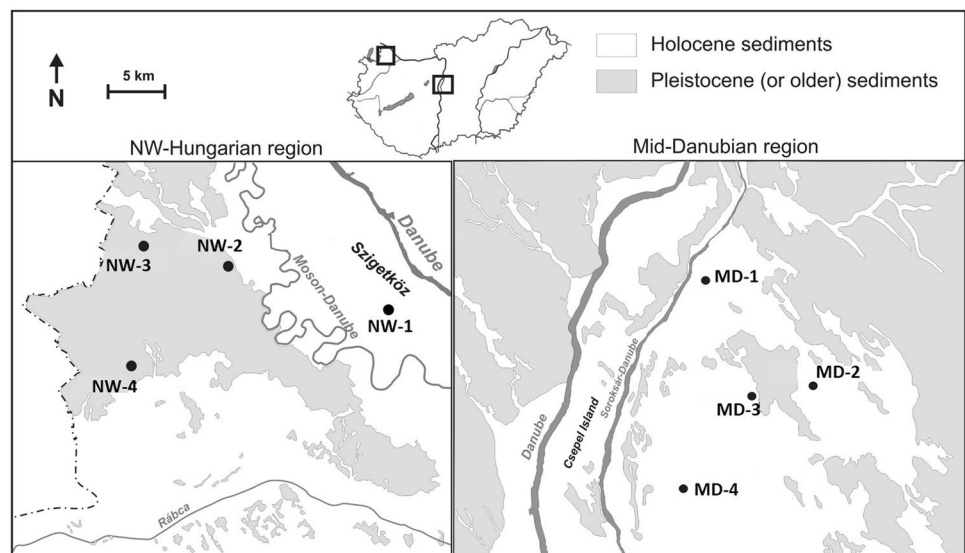




Fig. 2 Gravel pits operating with (a) bed dredging and (b) assorting system

From every selected quarry four sorted (particle size 0–4, 4–8, 8–16, 16–32 mm) and washed samples were collected. The sampling was made with care, and after homogenization, the representative portions were taken from the whole with the halving method.

Methods

Macroscopic, microscopic and heavy mineral studies were made on the different grain fractions. Variability in the quality and nature of individual outcrops were established using a combination of petrographic techniques on different scales. From all the localities, traditional macroscopic gravel petrography was done on the 4–8-, 8–16- and 16–32-mm fractions classifying av. 200 pebbles by fractions (Fig. 3). In addition, the 2–4-mm fraction was selected for fine-grained pebble examinations (FPE; Bradák et al. 2014) which

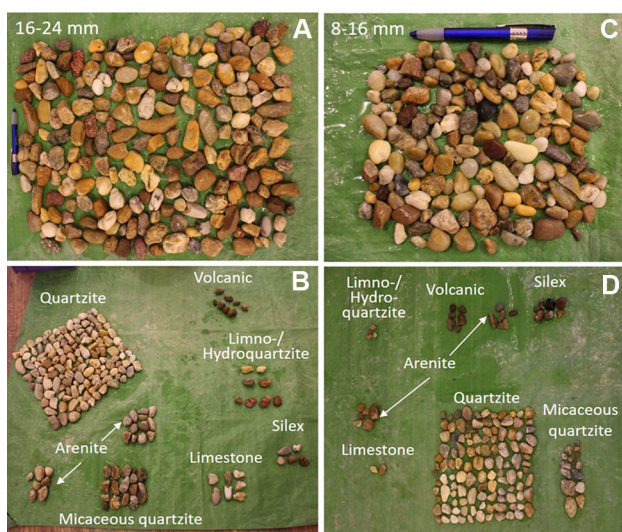


Fig. 3 Macroscopic petrographic examination and documentation of the coarse pebble (MD-1 16–24 mm (a–b) and 8–16 mm (c–d)) fractions

involved the modal composition analysis in petrographic thin sections (embedded in cement, instead of epoxy) containing more than 300 grains per sample. Microscopic petrographic investigations were done on a Zeiss petrographic polarizing microscope. The quantification was based on the grain counting method. As the sieved gravel fractions have uniform grain-size distributions, our results produced by grain counting method are corresponding to the results gained by the conventional point counting method.

Another aspect of microscopic petrographic investigations was the evaluation of potentially alkali-silica reactive (ASR) aggregates. The chemical process is described in the literature (Stanton 1940; Swamy 1992; Dron and Brivot 1992, 1993; Fournier and Bérubé 2000), and in addition to the physical and chemical tests, the petrographic examination of aggregates is a basic method to predict the potential reactivity (e.g. West 1994; Ramachandran and Beaudoin 2000; Ahmad et al. 2018; Šachlová et al. 2016, 2017). In the Hungarian Danube sediments, being silicate-based lithological assemblages, ASR-sensitive forms of silica (micro- or cryptocrystalline quartz, submicroscopic or disordered quartz, strained quartz, opal, chalcedony, tridymite, cristobalite, devitrified glass) are determined and quantified, since those can be harmful in the concrete mixture already in small amounts (e.g. 2 w% opal) (French 1991; Beck et al. 2000; Danielsen and Nebdal Svendsen 1991; Anastasio et al. 2016). In this study, we followed the recommendations of Fernandes et al. (2016); the undulatory extinction angle method proposed by Dolar-Mantuani (1983), West (1991, 1994) and Ramos et al. (2016); and the micro-cryptocrystalline silica content determination emphasized by Andersen and Thaulow (1989) and Ramachandran and Beaudoin (2000).

There were eight samples (one sample per quarry) selected for heavy mineral investigations. The specimens were made of the washed 0–4-mm fraction by subsequent drying (on room temperature), mass weighing, dry-wet-dry sieving (63, 125 and 250 μm sieve sizes), mass weighing of the four fractions, selection of fractions 63–125 μm and 125–250 μm , concentrating the heavy mineral fraction in the two size-fractions by a heavy liquid (bromoform) and repeated rinsing with alcohol, separate mass weighing of heavy and light fractions in both size ranges, and preparation of polished grain section of the two heavy mineral fractions (embedded in synthetic resin, planar burnishing to uncover the complete cross-section of the grains). The analysis involved the qualitative and quantitative determination of the mineral species and their main characteristics by a Nikon OPTIPHOT2-POL polarizing microscope. The photo documentation was done with a Nikon DS Fi1 camera and the NIS Elements software. The quantitative determination was done with the Fleet grain counting method (Fleet 1926; Galehouse 1969) which gives a number percentage, but

— because of definite grain size — these data can be used as area or volume percentages as well (Galehouse 1969).

The elemental compositions of samples (four fractions from each site) were determined by a combination of nuclear analytical techniques. Prompt-gamma (PGAA) and instrumental neutron activation analysis (NAA) were performed at the Centre for Energy Research, Budapest (see the method-description in Szilágyi et al. 2019). The set of elements obtained by these two methods is comparable for some elements (these can be used as an internal quality assurance indicator) and complementary for many other elements. Since the neutrons can penetrate deeply into the sample, the analytical information is highly representative to the bulk of the materials. Peak identification and quantitative analyses of both methods are performed using the k_0 method (De Corte 1987; Révay 2009; Firestone et al. 2014; Molnár 2004). The major-minor element compositions (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, H) of the sandy gravel samples have been determined by PGAA. In silicate matrices, some additional important trace elements (B, Cl, Sc, V, Cr, Co, Nd, Sm and Gd) also can be detected by PGAA. The upgraded Budapest PGAA facility was technically described by Szentmiklósi et al. (2010) in detail. During the analysis, the whole sample (in solid or powdered form) is irradiated by cold neutrons in an external beam (thermal equivalent flux of $1.2 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$, 24 mm^2 to 1 cm^2 beam spot size). During the neutron irradiation, the induced characteristic gamma radiation is detected with a High-Purity Germanium Detector with a Bismuth-Germanate Scintillator active background reduction system (HPGe-BGO) placed next to the beamline. The trace element compositions (As, Ba, Br, Ca, Ce, Co, Cr, Cs, Eu, Fe, Hf, K, La, Na, Nd, Rb, Sb, Sc, Sm, Sr, Ta, Tb, Th, U, W, Yb, Zn, Zr) of sandy gravels have been measured by NAA. The features of the Budapest NAA facility were described by Szentmiklósi et al. (2016). With this technique, we irradiate the powdered samples with neutrons in the research reactor's active zone (thermal neutron flux of $3.0 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$), and the gamma rays from the decay of short- and long-lived radionuclides are observed through a multi-step detection procedure (4–5 and 20–30 days after irradiation) using well-shielded HPGe detectors.

Results

Macroscopic pebble petrography of the coarser fractions

The general results of pebble petrography (of the 16–24-mm, 8–16-mm and 4–8-mm fractions) are presented in Figs. 4 and 5. Concerning their compositions, the gravels can be classified as oligomictic (quartzose) and subordinately polymict (two cases: NW-2 8–16 mm, NW-4 8–16 mm). Quartz pebbles predominate but those are usually below 70%. Comparing the two main regions, it is a fact that limestone is more common among the sedimentary rock types in the NW Hungarian region.

Sediments of the Mid-Danubian region contain several quartz varieties. Yellow and brownish quartzite are the main types, the white type is less common, whereas dark to light grey ones occur subordinately. Quartz and mica (mainly muscovite) containing (metamorphic or metagranitoid) rock fragments were observed in almost all quarries (micaceous quartzite, phyllite, gneiss of Palaeozoic crystalline formations from the Garam river catchment area/Bohemian Massif; Bradák et al. 2014) (7.0–12.5%). Further rock types, such as volcanites, silex, siliciclastites, limno-/hydroquartzite or limestone additionally, are present (22.0–31.5%). Volcanites are grey-black-red andesite clasts with feldspar and amphibole phenocrysts and are originated from the Visegrád and Börzsöny Mts. (Karátson et al. 2000, 2007). Siliceous sedimentary rocks are grey-black sandstone, quartzose sandstone and siltstone or rarely marl-claystone (Szeberényi et al. 2014). Quartz arenite pebbles are remarkable in MD-3 8–16-mm and 4–8-mm fractions (15%). Limestones are white-pale yellow micritic-sparitic inhomogeneous and homogenous textured, fossil-free rocks probably of Mesozoic or Miocene age (Bradák et al. 2014).

Clastic fractions of the NW Hungarian region are also predominated by quartz varieties. White to yellow quartzite is the main type, whereas dark to light grey and brown ones are subordinate. Metamorphic or metagranitoid rock fragments (quartz and mica composition; e.g. micaceous quartzite, phyllite, gneiss) were recognized in all quarries (11.0–27.9%) and possibly could derive from the crystalline formations of the Bohemian Massif (Bradák et al. 2014). Other rock types (volcanites, silex, siliciclastites, limno-/hydroquartzite, limestone) are additionally present (12.0–30.4%). Relevantly higher limestone content (6.5–11.5%) at NW-1 and NW-2 could be observed. Siliciclastites are red-yellow sandstones, light-colored calcareous sandstones (Neogenic of the Leitha Mts.), quartz arenites and green-grey-black siltstones (Mesozoic flysch and molasse sediments of the N, E or Central Alps). Limestones are grey-greyish yellow fine micritic fossil-free rocks of Mesozoic (N, E or Central Alps) or Neogene age (Leitha Mts.) (Szeberényi et al. 2014).

The petrography of pebbles reflects relatively high maturity. The provenance was significantly influenced by the basement and the river catchment area composition. The absolute dominance of quartz or quartz dominated metamorphic-metagranitoid rock fragments indicates the provenance from weathered crystalline rocks (of the Alps/Bohemian Massif) and the longer sediment transportation path. The minor role of sedimentary rocks (e.g. silex, siliciclastic rocks, limestone) and volcanites is dictated by the local geology (influence of smaller river input).

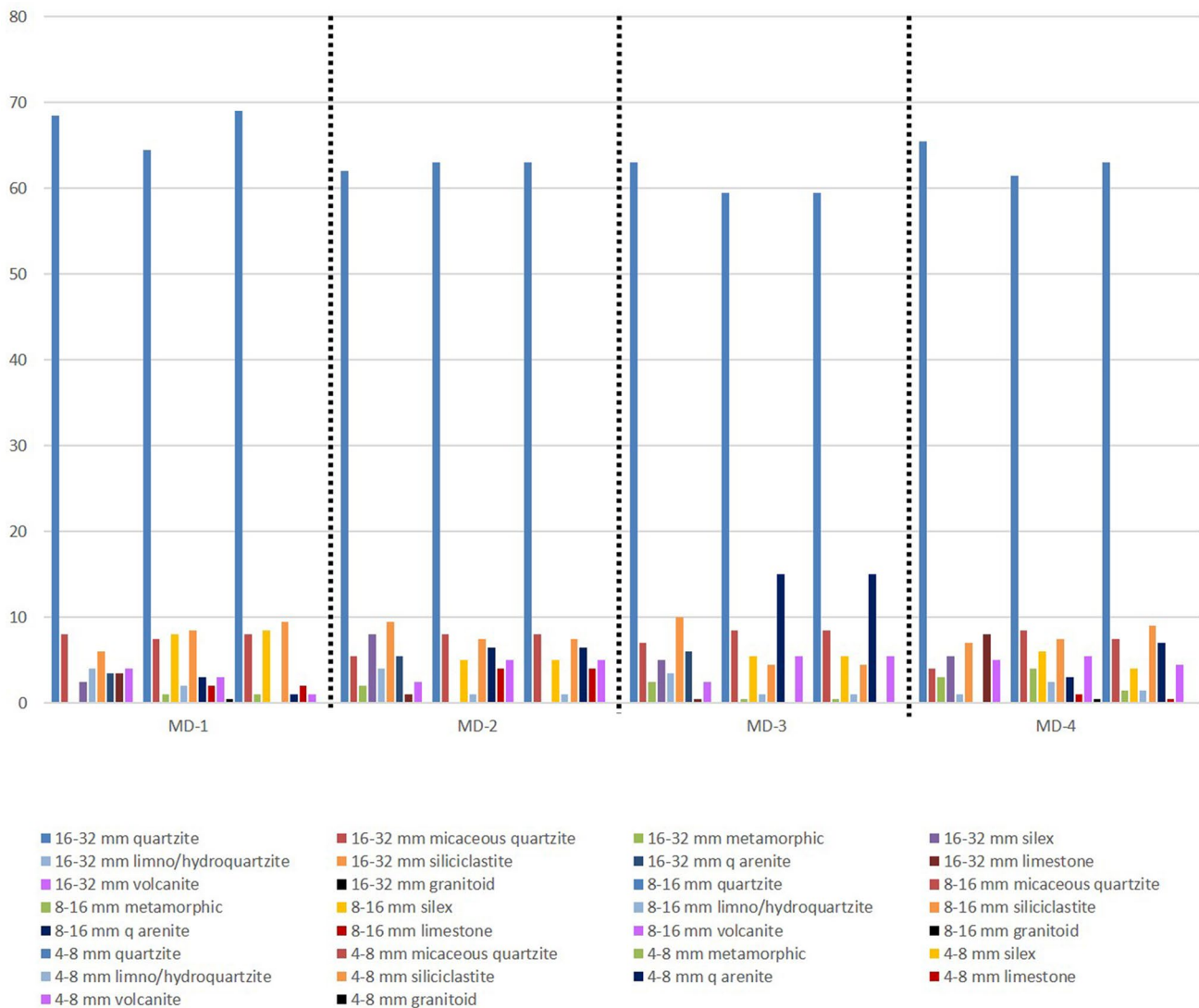


Fig. 4 Modal composition of 16–32-mm, 8–16-mm and 4–8-mm pebble fractions of the Mid-Danubian quarries (based on macroscopic petrographic examination)

FPE of the 2–4-mm fraction

The modal compositions of the studied 2–4 mm sediment fractions are presented in Figs. 6 and 7. A difference in the mineralogical maturity of sediments from the two regions is best indicated by the <50% quartz-quartzite at Mid-Danubian sites (polymictic composition), while it is almost 60% at the NW Hungarian sites (oligomictic composition).

The four raw materials of the Mid-Danubian region have common features that can characterize the territory and the section of the fluvial depositional area (Fig. 6). These features are the predominance of quartzite (mono- and polycrystalline quartz lithoclasts) over other metamorphic

rock types (e.g. micaceous quartzite, mica schist-gneiss-metagranitoid rock fragments; 16–24%), igneous rocks (volcanics and granitoids; 16–19%) and sedimentary rocks (siliciclastites, limestone, silex; 3–10%). The abundance of igneous and especially volcanic rocks (andesite) is significant. The quartzites often have sutured grain boundaries (with schistose oriented texture), but close-to-equilibrium (bulky non-oriented) texture is also present. The intra-grain crystal sizes are varied (50–1500 μ m). Monocrystalline quartz grains are very subordinated. Micaceous quartzite with oriented muscovite flakes is common. Subordinate phyllitic to gneissose rock fragments are also present. Plutonic rock fragments with microcline or plagioclase and

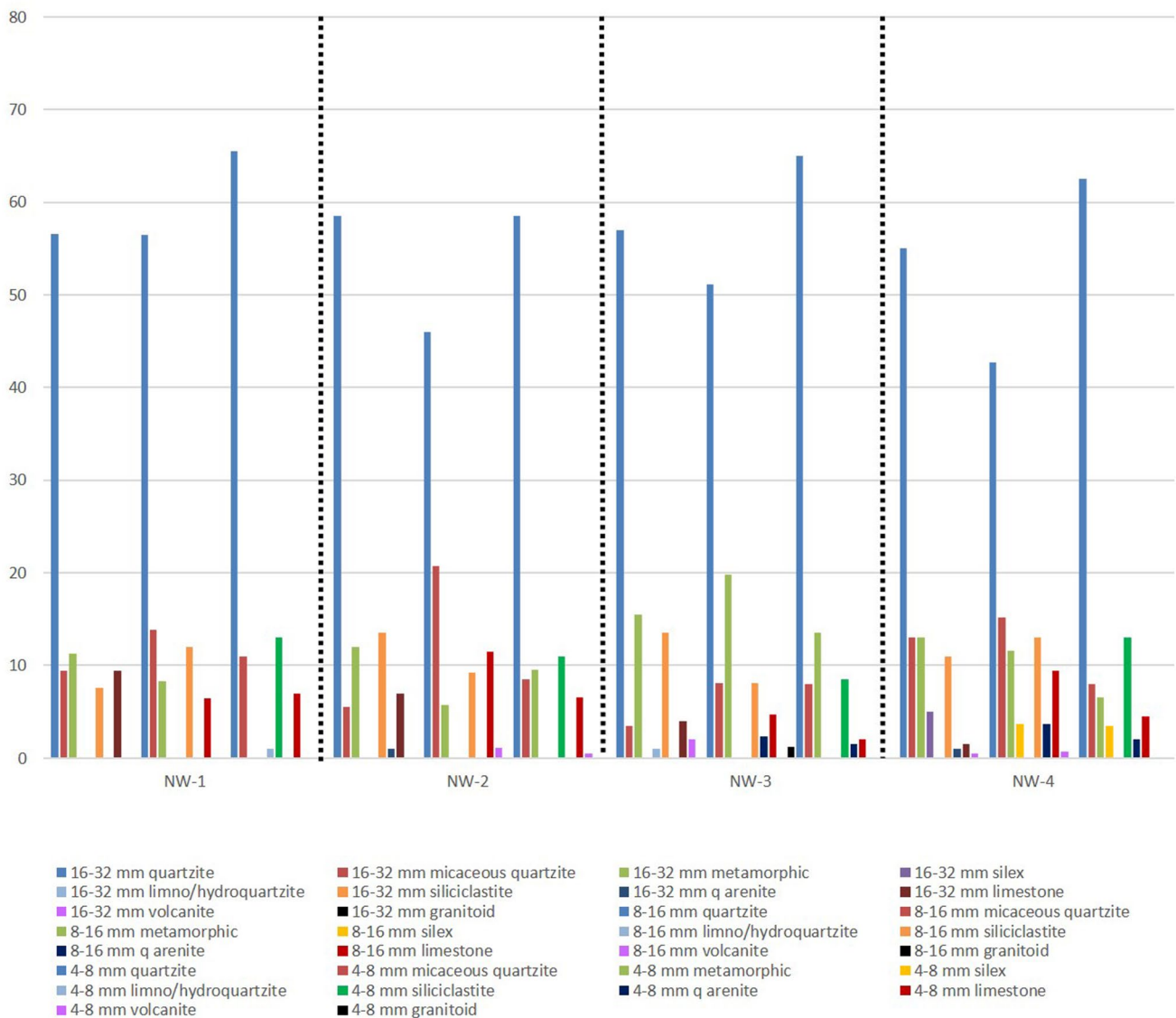


Fig. 5 Modal composition of 16–32-mm, 8–16-mm and 4–8-mm pebble fractions of the NW Hungarian quarries (based on macroscopic petrographic examination)

mafic minerals (e.g. green amphibole) can be observed. Monomineralic K-feldspar (sericitized) or plagioclase clasts are granitoid originated fragments. Volcanics are predominantly andesites with plagioclase and mafic (biotite or amphibole) phenocrysts and glassy to plagioclase laths containing ground mass. Microcrystalline quartz (homogeneous or with veins) and radiolarite (both can be carbonated) are characteristic components. Micritic to sparry limestone, loess clasts and silt-sandstones (dominantly quartz-muscovite composition, but sometimes with calcareous cement or calcite grains) are also typical grain types.

The four raw materials of the NW Hungarian region have common features that can characterize the territory and the section of the fluvial depositional area (Fig. 7). The

predominance of quartzite over other metamorphic rock types (9–16%), igneous rocks (3–11%) and sedimentary rocks (1–9%) is more expressed and typical for these sediments than in the Mid-Danubian region. The compositions of the major rock groups are a bit different from the Mid-Danubian sediments: the igneous rocks are rather represented by feldspar mineral clasts than granitoid fragments (or volcanics); the sedimentary rock types are more varied and characteristic (fossiliferous limestone, carbonatized radiolarite). In these sediments, the quartzites have sutured grain boundaries and variable intra-grain crystal sizes (50–1000 μ m). Monocrystalline quartz grains are more subordinate than in the Mid-Danubian samples. Quartzites that contain oriented muscovite flakes or rare feldspars are common. Subordinate

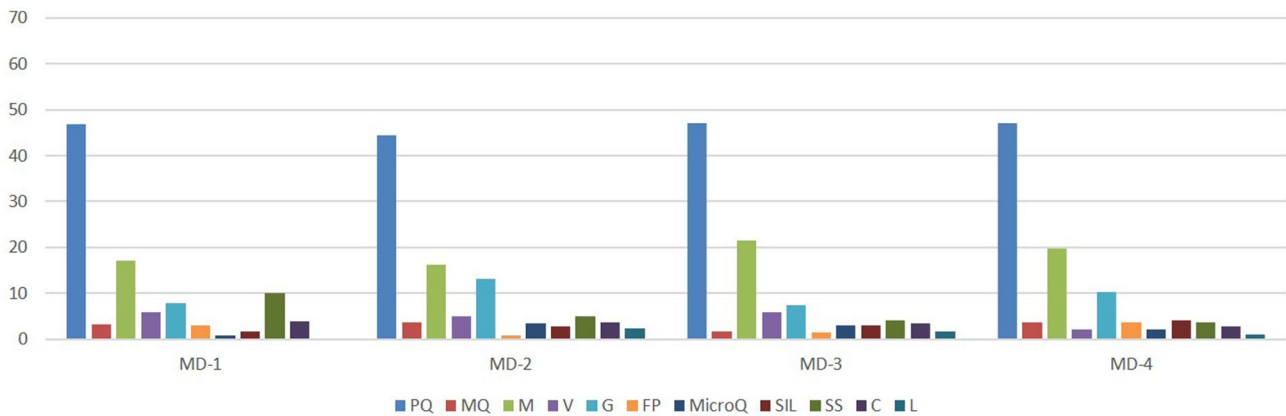


Fig. 6 Modal composition of 2–4-mm fine pebble fractions of the Mid-Danubian quarries (based on microscopic petrographic examination). Abbreviations: PQ, polycrystalline quartz; MQ, monocrystalline quartz; M, metamorphic rock; V, volcanic rock; G, granitoid

rock; FP, feldspar; MicroQ, microcrystalline quartz; SIL, siliciclastite and radiolarite; SS, claystone-siltstone-sandstone; C, carbonatic rock; L, loess

gneissose-metagranitoid rock fragments are also present. Rare granitoid rock fragments and more common monomineralic microcline, sericitized K-feldspar or plagioclase clasts can be observed. Commonly carbonatized microcrystalline quartz (homogeneous or with veins) and radiolarite are abundant and characteristic components. Micritic fossiliferous limestone and claystone-siltstone-sandstone-metasandstone (dominantly quartz-muscovite, sometimes calcareous or glauconitic composition) are also typical grain types.

Macro- and micro-scale pebble petrographies of the samples showed diagnostic differences among the two regions. All sampled sediments are dominated by metamorphic (and especially quartzite) rock originated clasts. Low-quartz-content metamorphics, igneous and siliciclastic sedimentary rock fragments are subordinate to quartzite content. Furthermore, pebbles of the Mid-Hungarian region contain a higher ratio of volcanic rocks, while gravels of the NW Hungarian region represent more mature sediments with a higher proportion of limestone.

The assessment of the potential ASR-sensitivity of aggregates was based on the quantification of micro-cryptocrystalline quartz (<100 μm) and on the measurement of the undulatory extinction angle of 20 quartz crystals per sample after DeHills and Corvalán (1964) and the classification applied by Ramos et al. (2016) (after the Portuguese specification LNEC E 415 1993). According to the RILEM AAR-1.1 (2016) specifications, all the samples, due to their microcrystalline quartz content exceeding 2 w%, have to be treated as class II (see Table 1). The significant amounts of potentially ASR reactive forms of silica in metamorphic rock fragments (Fig. 8) are micro-cryptocrystalline quartz, strained (deformed, stretched) quartz crystals with strain lamellae, subgraining and/or sutured/interlobate boundaries indicating bulging recrystallization (BLG), subgrain rotation recrystallization (SGR) or grain boundary migration (GBM) (Stipp et al. 2002; Krutilová and Přikryl 2017). According to Passchier and Trouw (2005), these microstructural characteristics indicate deformation and recrystallization under middle temperature regimes and

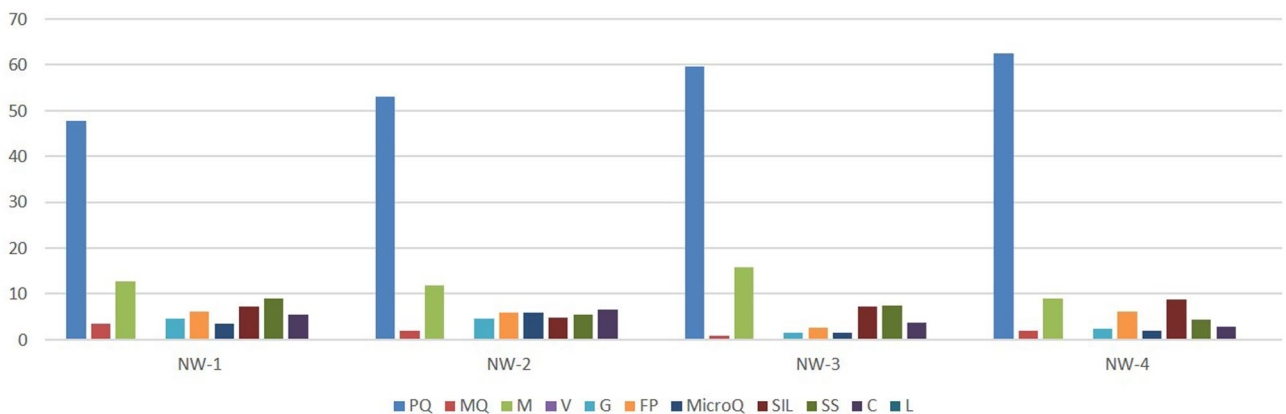


Fig. 7 Modal composition of 2–4-mm fine pebble fractions of the NW Hungarian quarries based on microscopic petrographic examination. Abbreviations: PQ, polycrystalline quartz; MQ, monocrystalline

quartz; M, metamorphic rock; V, volcanic rock; G, granitoid rock; FP, feldspar; MicroQ, microcrystalline quartz; SIL, siliciclastite and radiolarite; SS, claystone-siltstone-sandstone; C, carbonatic rock; L, loess

Table 1 ASR-related petrographic characterization of quartz in alluvial sandy gravel formations of Danube in Hungary (strain I.: strain lamellae in quartz; ASR categories: class I: very unlikely to be alkali-reactive; class II: alkali-reactivity uncertain; class III: very likely to be alkali-reactive with aggregates containing opal or opaline silica)

| Aggregate | Total quartz content (%) | Potentially reactive silica forms (%) | Reactivity class (RILEM AAR-1.1) | Undulatory extinction angle | Deformation rate (LNEC E415) |
|-----------|--------------------------|---|----------------------------------|-----------------------------|------------------------------|
| MD-1 | 65.0 | <100 μm : 25 Strain I.: 13 Sum: 38 | Class II | 4–10° | Weak |
| MD-2 | 68.9 | <100 μm : 28 Strain I.: 14 Sum: 42 | Class II | 6–10° | Weak |
| MD-3 | 69.2 | <100 μm : 24 Strain I.: 2 Sum: 26 | Class II | 7–12° | Weak |
| MD-4 | 71.7 | <100 μm : 25 Strain I.: 6 Sum: 31 | Class II | 9–12° | Weak |
| NW-1 | 70.7 | <100 μm : 27 Strain I.: 3 Sum: 30 | Class II | 5–7° | Weak |
| NW-2 | 73.9 | <100 μm : 25 Strain I.: 2 Sum: 27 | Class II | 5–8° | Weak |
| NW-3 | 77.7 | <100 μm : 30 Strain I.: 5 Sum: 35 | Class II | 4–7° | Weak |
| NW-4 | 80.9 | <100 μm : 25 Strain I.: 1 Sum: 26 | Class II | 7–10° | Weak |

increasing dislocation density. It can be connected to relevant ASR potential (Wigum 1995; Wenk et al. 2008; Šachlová et al. 2017). In addition, micro-cryptocrystalline quartz or chalcedony/moganite of siliceous limestone and radiolarite (Fig. 8) (similarly to cherts), and micro-cryptocrystalline matrix of fine-grained clastic sedimentary rock fragments (e.g. sandstone-siltstone-greywacke) also requires attention (e.g. Katayama 2004, 2010a; Marinoni et al. 2012; Šachlová et al. 2017). The potentially ASR

reactive forms of silica in subordinate amount (>6%) of volcanic rock fragments are glassy or microcrystalline groundmass (Fig. 9). The neutral (silica) chemistry of andesitic rocks assumes an intermediate sensitivity to ASR (Katayama et al. 1989, Katayama 2010b; Medeiros et al. 2020). Due to their limited abundance in the grain mixture, siliceous limestone, radiolarite and volcanic lithofragments might act as detached (local) deterioration centers during ASR processes.

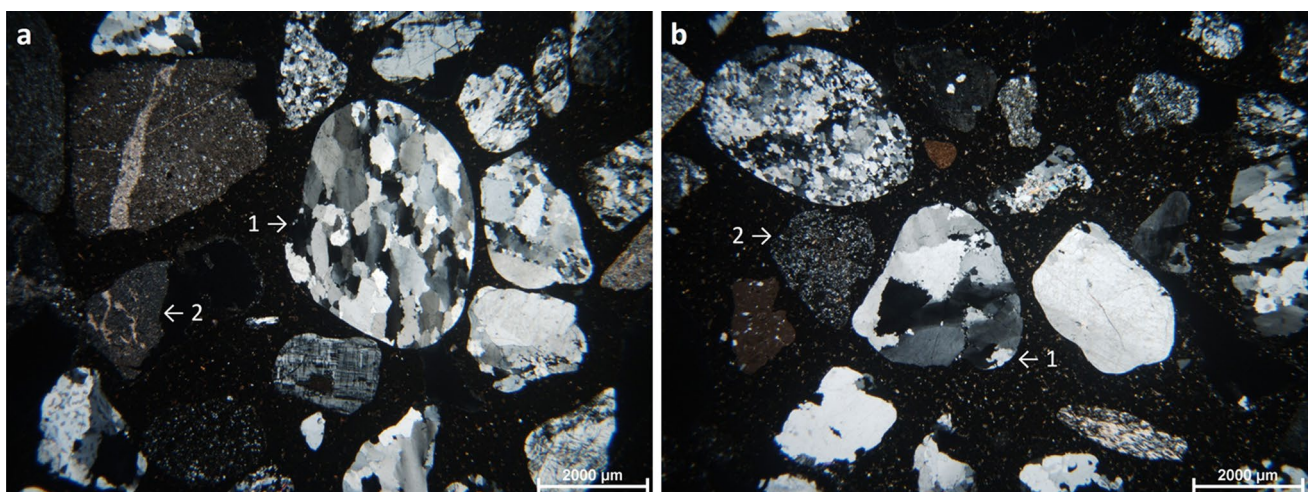


Fig. 8 Examples for ASR-reactive silica-rich grain types (1, crystalline quartzite; 2, microcrystalline siliceous limestone and radiolarite) in the 2–4-mm fraction of aggregates exploited at **a** NW-2 and

b MD-4. Note the quartzite grains with strained crystals of sutured boundaries and subgraining

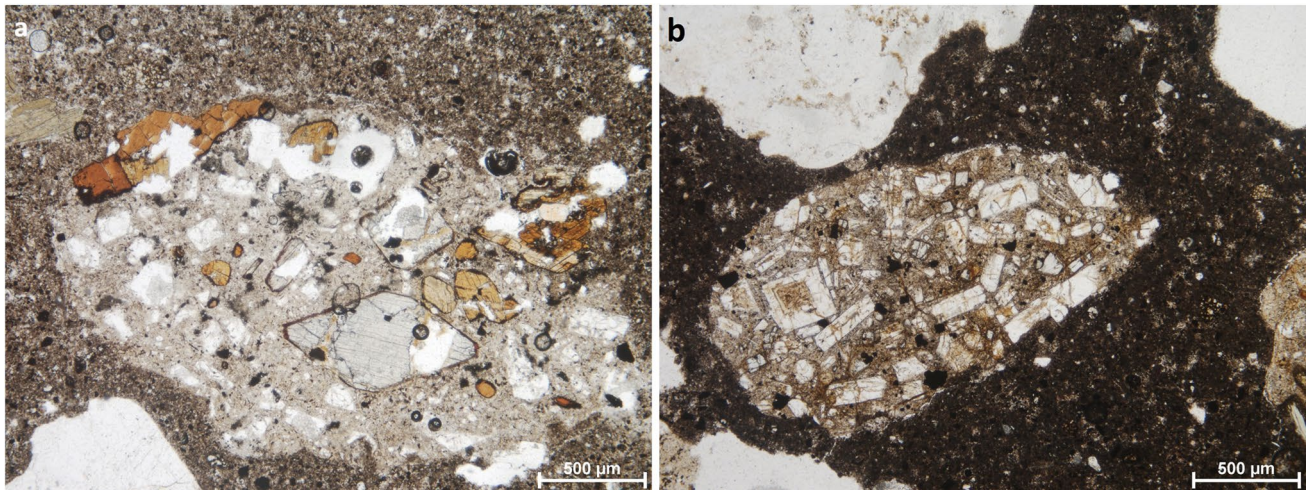


Fig. 9 Examples for ASR-reactive amorphous or microcrystalline groundmass in volcanic rock fragments in the 2–4-mm fraction of aggregates exploited at **a** MD-3 and **b** MD-4

Due to the variation of the characteristics of silica-rich aggregates inside a single quarry, the general specifications are conservative and require verification by expansion tests (e.g. accelerated mortar bar, concrete prism test). In addition, detailed SEM-PIA investigations (Šachlová et al. 2016, 2017) and CL microscopy (Krutilová and Příkryl 2017) would improve the knowledge on the ASR potential of each lithotype in the oligomictic natural mixtures.

Heavy mineral analysis of the 63–125- and 125–250-µm fractions

The results of the heavy mineral investigations are illustrated in Fig. 10 and plotted in Fig. 11. The two size fractions consist of very similar mineral assemblage; thus, they are described together, but indicating their ratios within parentheses, first for fraction 125–250 µm and then for fraction 63–125 µm.

Heavy mineral fractions of the Mid-Danubian region can be characterized by a variable mineral association containing rather simple monomineralic grains. The predominant garnet (23–55 and 27–33%) is associated with abundant opaque minerals (10 and 17%), green amphibole (8–13 and 16–18%), epidote-zoisite (5–18 and 10–18%), brown amphibole (2–12 and 8–13%) and orthopyroxenes (6–12 and 4–12%).

Oxi amphibole is a low quantity but characteristic phase (0–2% and 1–10%) (Fig. 11). The majority of the grains of these minerals are weakly rounded. The garnet is transparent and colorless (rarely rose). It is fragmented, xenomorphic/anhedral, sometimes contains inclusions. Pyroxenes are fresh, hypidiomorphic/subhedral forms with characteristic serrated edges. Orthopyroxenes are olive green-brownish green pleochroic (with abundant opaque inclusions), while clinopyroxenes are almost colorless with weak pale red-pale green pleochroism. Opaque minerals are well-rounded, isometric

grains. Hornblendes are dark green-bluish green-green-light green-yellow pleochroic, medium rounded, hypidiomorphic/subhedral crystals. Zoisite-clinozoisite grains are colorless, medium rounded, porous structured and hypidiomorphic/subhedral. Additional (below 3%) heavy mineral phases are siderite, chlorite, actinolite, limonite, tourmaline, staurolite, rutile, chloritoid, apatite, zircon, calcite, titanite, sillimanite, sericite, nontronite, tremolite, kyanite and muscovite. Complex lithic grains are low- to medium-grade metamorphic lithofragments (with zoisite, actinolite, green amphibole), micritic or sparry limestone.

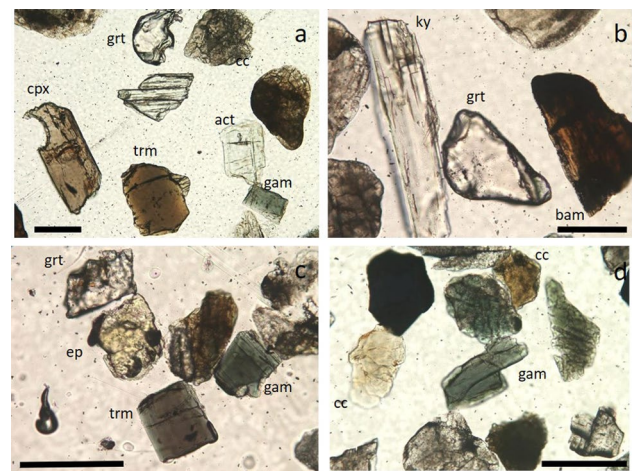


Fig. 10 Typical heavy mineral assemblages of sediments in the Mid-Danubian (**a**, **b**) and NW Hungarian (**c**, **d**) regions: **a** MD-2, 125–250 µm fraction; **b** MD-4, 125–250 µm fraction; **c** NW-4, 63–125 µm fraction; **d** NW-3, 63–125 µm fraction. The microscopic photographs were taken in plane polarized light, the scale is 100 µm. Abbreviations: grt, garnet; cpx, clinopyroxene; trm, tourmaline; act, actinolite; gam, green amphibole; ky, kyanite; ep, epidote; cc, calcareous lithofragment

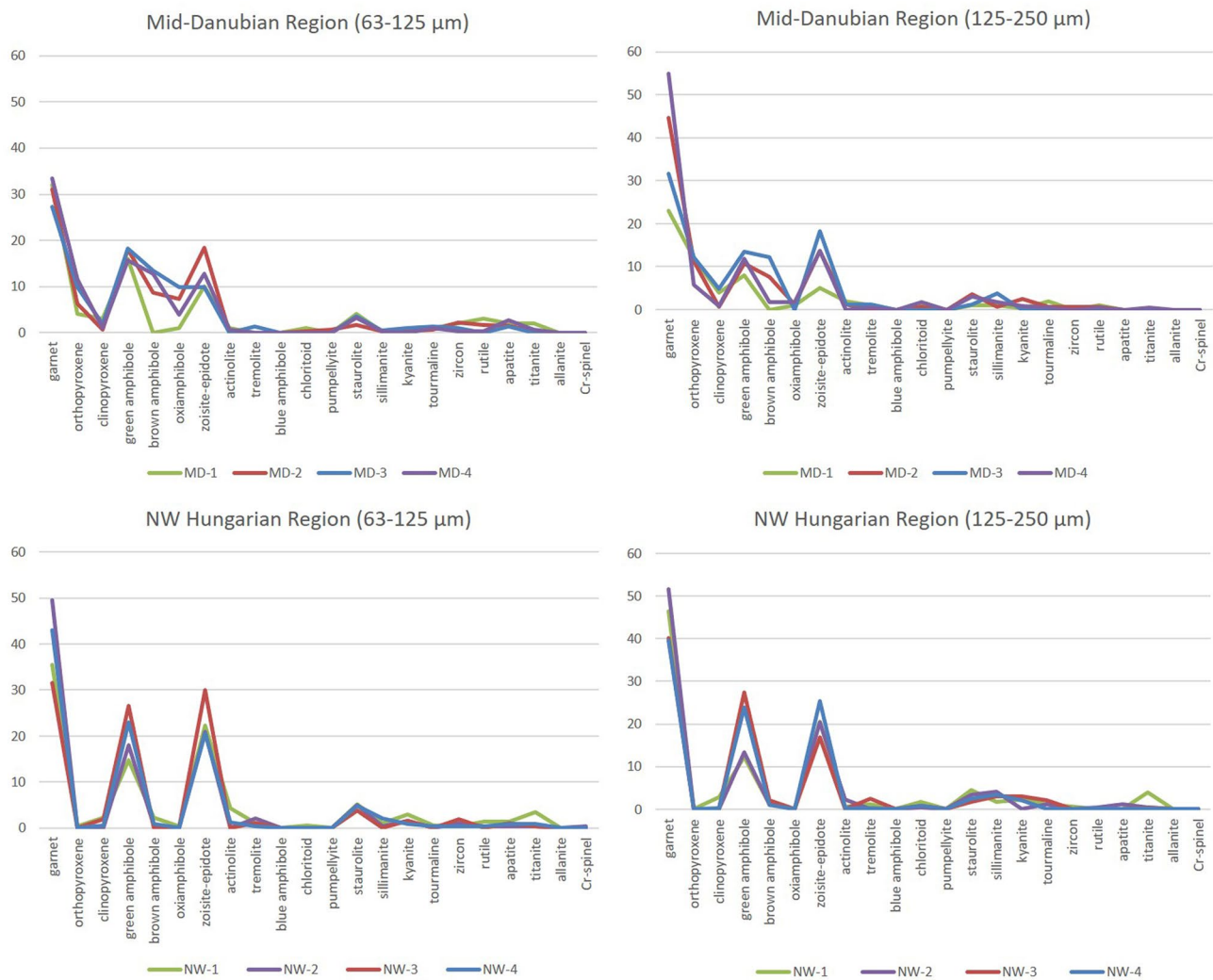


Fig. 11 Abundance (%) of translucent heavy mineral species in the 63–125- and 125–250- μm fractions of sediments of the Mid-Danubian and NW Hungarian regions

The association of the NW Hungarian samples is also varied, containing dominantly monomineralic grains and subordinate amount (2–5%) of metamorphic lithofragments (Fig. 11). The garnet is predominant (40–52% and 32–50%) together with the calcareous lithoclasts (both sparry and micritic, 18 and 33%), and those are associated with abundant green amphibole (12–27% and 15–27%), zoisite-epidote (17–25% and 21–30%) and opaque minerals. Characteristic features are the low quantity of brown amphibole (1–2% and 0–2%) and the absence of orthopyroxene and oxiamphibole (Fig. 11). Majority of the grains of these minerals are weakly rounded, except for opaque minerals and micritic grains. The garnet is transparent and colorless (rarely rose) with fragmented, xenomorphic/anedral shapes. Green amphiboles are clear, medium to dark green, pleochroic, hypidiomorphic/subhedral, columnar forms with thin cleavage planes. Zoisite-clinozoisites are colorless, hypidiomorphic/subhedral, prismatic or isometric crystals. Epidotes are colorless-lemon yellow,

pleochroic, hypidiomorphic/subhedral, prismatic or isometric crystals with dense fine-grained inclusions. Opaque minerals are rounded, isometric grains. Additional heavy mineral phases are chloritoid, staurolite, actinolite, kyanite, brown amphibole, clinopyroxene, rutile, apatite, sillimanite, tremolite, tourmaline and zircon. Complex lithic grains are metamorphic (greenschist, mica schist, graphitic-chloritic-sericitic phyllites, amphibolite) and calcareous lithofragments (sparry or micritic).

The heavy mineral assemblages of the two investigated regions show slight differences (Fig. 11). The main difference is the lithofragment content which is determined by the high limestone clast ratio at the NW Hungarian localities, while other (metamorphic) rock types are represented by similar abundances in the two regions.

The rich heavy mineral assemblage of the Mid-Danubian region has multi-source origin. They are partly derived from a metamorphic terrain (both the Bohemian Crystalline

Massif and the Alps; Thamóné Bozsó 2002; Józsa et al. 2020; Arató et al. 2021): garnets with inclusions, green amphibole, brown amphibole, epidote-zoisite-clinozoisite, metamorphic lithofragments, chlorite, actinolite, tremolite, tourmaline, rutile, sillimanite, kyanite, staurolite and chloritoid. The other important part of the assemblage can be connected to younger volcanic sources (e.g. Miocene andesite-dacite of the Börzsöny-Visegrád Mts., Józsa et al. 2020): clear garnet, orthopyroxenes, opaque minerals, oxiamphibole and green hornblende. Siderite and limonite can have a local origin, as authigenic weathering products of the basal layers of the Pleistocene gravel formation. The limestone fragments and calcites can be connected to the Mesozoic or Miocene calcareous sedimentary formations of the surrounding mountains (Gerecse, Pilis, Buda Mts.; Thamóné Bozsó 2002).

The heavy mineral assemblage of the NW Hungarian region shows a narrower range with dominantly metamorphic originated garnet, green amphibole and epidote-zoisite-clinozoisite (tremolite, chlorite, actinolite, tourmaline, rutile, sillimanite, kyanite, staurolite, zircon) of the Bohemian Crystalline Massif,

the Alps (Penninicum, Central Alps) and the NW Carpathians (Tatricum, Fatricum, Veporicum) (Thamóné Bozsó 2002; Józsa et al. 2020, Arató et al. 2021). Absence (e.g. orthopyroxene, oxiamphibole) or a very small amount (brown amphibole) of volcanic components indicates the upper course position of this region along the Danube flow without the contributions of the younger volcanic sources. Another feature is the significant amount of calcareous lithofragments (fine- to coarse-crystalline limestone) which can derive from the (Northern, Eastern or Central) Alps, the Leitha mountains or the NW Carpathians (Thamóné Bozsó 2002; Józsa et al. 2020, Arató et al. 2021).

Chemical characterization of the 0–4-, 4–8-, 8–16- and 16–32-mm fractions

Major, minor- and trace-element distributions of the Danube terrace sediments of the two studied sections are represented in Figs. 12 and 13. According to the conventions in geochemistry, the element distributions are normalized to geological reference materials to gain more comparable

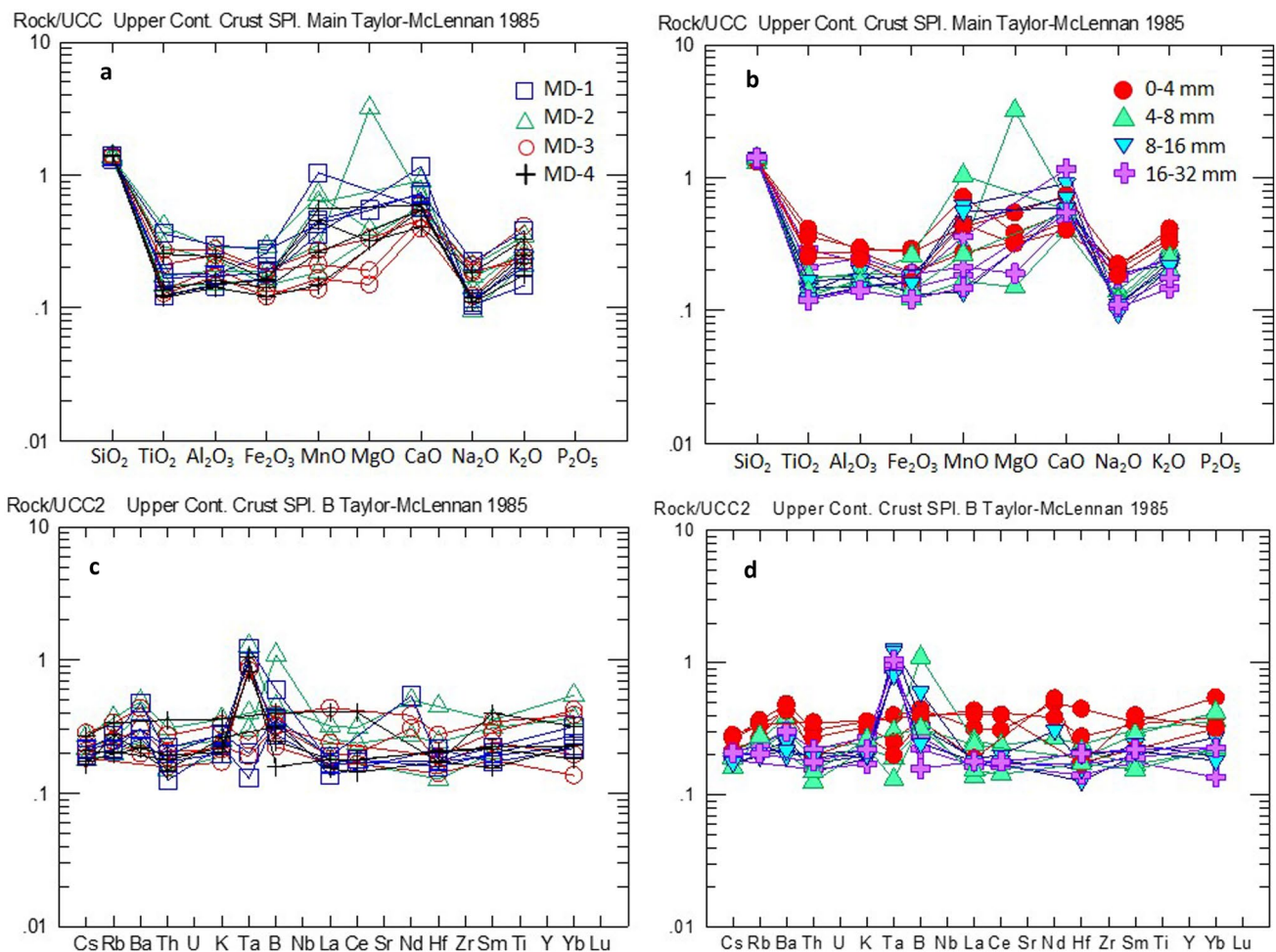


Fig. 12 Upper continental crust (UCC; Taylor and McLennan 1985) normalized major-minor and trace element distributions of 16–32-mm, 8–16-mm, 4–8-mm and 0–4-mm fractions of the Mid-Danubian sandy gravel quarries by localities and by grain sizes

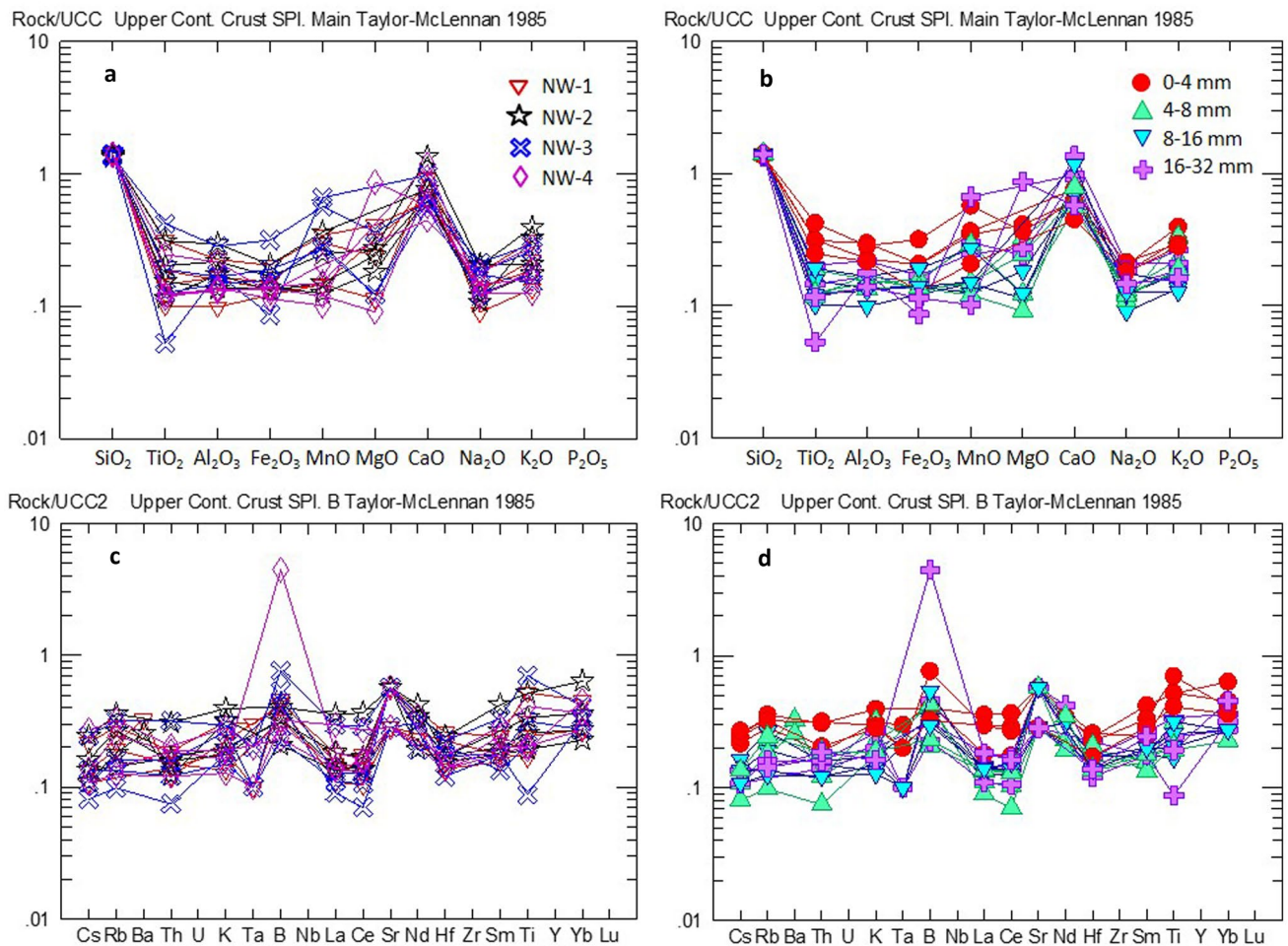


Fig. 13 Upper Continental Crust (UCC; Taylor and McLennan 1985) normalized major-minor and trace element distributions of 16–32-mm, 8–16-mm, 4–8-mm and 0–4-mm fractions of the NW Hungarian sandy gravel quarries by localities and by grain sizes

data. In the case of clastic materials (sands, gravels), the upper continental crust (UCC) is an accepted reference average composition (Taylor and McLennan 1985; McLennan 2001). After normalizing our measured concentration data with this reference composition, difference from the unity expresses (negative) depletion or (positive) enrichment of certain elements compared to average clastic material composition. These normalized concentration profiles describe the geochemical behavior of the observed sediment. Major and minor elements (abundance in the Earth's crust is above 1 w% and 0.1 w%, respectively) are presented in oxide form, while trace elements are in elemental form. The elements are sorted according to the geochemical conventions (considering the mobility and incompatibility of elements).

Quarried gravelly sediments of the Mid-Danubian region are characterized by 84–93 w% SiO₂; depletion in Na₂O and MgO (the latter is often below the detection limit of the method); moderate depletion in TiO₂, Al₂O₃, Fe₂O₃ and K₂O; and weak enrichment of CaO (1.6–4.9 w%), while by

varied concentrations (with relatively high values) for MnO (Fig. 12). Sediments at MD-1 have small concentrations of Fe₂O₃, MnO and CaO, while the gravels of MD-3 are the most enriched in these oxides. A common moderate depletion in all trace elements compared to the average reference composition can be observed with small discrepancies in boron and tantalum content (Fig. 12). The trace element contents are relatively consistent; there is no trend in the rare earth element (La-Lu) distribution. Based on the granulometry, a clear trend can be outlined: the finer the sediment the richer in minor and trace elements counter to SiO₂ and CaO. This enrichment is the most expressed for the 0–4-mm fraction of the sediments, while it is less clear in the coarser fractions. Tantalum enrichment in the coarsest fractions can be detected.

Sediments of the NW Hungarian region have similar major element distribution to the Mid-Danubian region with 84–94 w% SiO₂ and depletion of elements except for CaO (1.8–4.9 w%). The only difference is the wider

variation range of TiO_2 , Fe_2O_3 and MnO concentrations. The trace elements also show a comparable pattern to the other region but with more expressed depletion (Fig. 13).

The common geochemical feature of the two regions is the CaO-enrichment and depletion in MgO, while a very even, moderately depleted pattern was found for all trace elements. These are characteristic geochemical ‘fingerprints’ of the Danube sediments (comparing to clastic sediments of other gravel quarries in Hungary based on our own investigations). Based on these element profiles, the claimed origin of an industrial raw material can be validated on a regional scale.

Discussion

The comprehensive petrographic and geochemical investigation of alluvial sandy gravel formations of the middle course of the Danube shows that the main river of the Carpathian Basin has its peculiar petro-geochemical characteristics, distinguishable from other Hungarian gravel sources, and this determines its applicability to specific construction purposes.

It is common knowledge in concrete industry that the shape and texture of sand/gravel clasts influence the mechanical and fracture properties of concrete (e.g. Belhadj et al. 2014; Aissoun et al. 2015) and on the transport properties which can positively influence durability potentialities of the composite (Hu and Stroeve 2006). Particle geometry influences abrasivity in petrographically homogeneous rock types (Hofer et al. 2013). The form of coarse aggregate (angularity is preferred) has a significant effect on the stability, rheology and compressive strength of concrete. Angular forms and spheric shapes improve the resistance of the concrete to crack propagation. On the contrary, there is a different requirement of the aggregate shapes in the asphalt industry. The morphological characteristics of fine aggregates can be correlated with the high-temperature viscoelastic properties of asphalt mixes (Wang et al. 2018), while it is less expressed for coarse aggregates. Crushed materials and manufactured gravels (with low roundness values) have more complex morphological characteristics, and those can effectively improve the deformation recovery, reduce the deformation of asphalt mixes, provide larger failure stress but lower failure strain. Another geotechnical parameter is granulometry which determines the fineness index.

Rock fabric characteristics (especially size descriptors) have significant influence on the engineering properties in general. In the case of oligomictic sandy gravels, the overall morphology of clasts is determined by the lithotypes. Although, grain-shape analysis of the Hungarian clastic sediments was not done, based on the macroscopic observations

those have relatively homogeneous, well rounded and medium spheric character (see also Pécsiné Donát 1958). The grain populations are characterized by the balanced distribution of quartz-rich lithofragments to the metamorphic rock fragments with foliated texture and siliciclastic sedimentary rocks with a layered texture. The quartzose clasts have more isometric shapes while the foliated-layered rocks show more elongated forms. In addition, crushing the rock fragments with mica containing planes results in flaky shapes. The subordinate volcanic materials (in Mid-Danubian sediments) have well rounded and isometric shapes. The morphology of the pebble populations provides wide applicability of the sandy gravel raw materials. Tiny differences in the ratio of elongated clasts in the assemblages can impose some limitations to certain forms of use. The more elongated shapes obtained from crushing (due to their smaller mechanical resistance) imply that the crushed aggregates are less appropriate for use in road materials, coarse concrete aggregate and ballast for railways (Wenk et al. 2013). In addition, the observed Hungarian river-derived clastic raw materials are less appropriate for asphalt manufacturing due to their better roundness. For such purposes, the crushing of sandy gravel assemblages would be beneficial. The granulometry of the Hungarian clastic sediments was not observed in this study, but previous researches provided data. Burján (2002) evidenced that the sandy gravel sediments of the 1st and 2nd terraces in the Mid-Danubian region have similar granulometry (well sorted sediments with two-maxima distribution curves).

Mechanical adhesion between the aggregate and the cement paste is influenced by (1) the morphology of the aggregates, (2) chemical bonding and (3) chemistry of the cement. Both the (1) and (2) are affected by the composition of the aggregate. Silica-rich aggregates usually provide low adhesion to the binder in asphalt mixtures which results in weaker durability of the pavement surface (Anastasio 2015). Metamorphic rock aggregates (due to their weathering) can have weaker mechanical properties but better surface properties when used as concrete aggregate (Danielsen and Rueslten 1984). The nature of chemical bonding can be characterized on different scales (Miller et al. 2011; Miller 2010; Lyne et al. 2013; Jones 1993). Micritic and dolomitic limestone, siliceous gravel and marble show significantly weaker chemical bonding (lower affinity to bond) with asphalt paste than other aggregates. Most other aggregates (metabasalt, quartzite and sandstone) exhibit stronger bonding affinity during asphalt- and asphaltic-concrete production (Miller et al. 2011; Hefer and Little 2005; Brand and Roesler 2017a, b). On the contrary, the chemical bonding functions in concrete mixtures in a more complex way (more reactive binder than asphalt) (Bentz et al. 2017). In concrete production, high-absorption limestones and marble performed very well (Shannon et al. 2015, 2017) and

outperformed other aggregate types (e.g. metabasalt) in most cases, while siliceous gravel and granitic gneiss mixtures proved to be the weakest material (compressive, splitting and flexural strength). Matching the mechanical properties of the aggregate-paste couple can further improve the mechanical performance of concrete (Beushausen et al. 2015; Bremner et al. 1986; Aitcin et al. 1990; Neville 1997; Sengul et al. 2002). Constituents with similar elastic modulus or coefficient of thermal expansion (volumetric changes during hydration) can result in lower-stress concentration (i.e. microcrack generation) at the interface of the two phases during loading. The incompatibility of properties of the aggregate-paste couple likely promotes the development of interfacial stresses, weakening the bond between the two phases and lowering the measured concrete strength.

The geotechnical properties of aggregates are determined by their petrographic-mineralogical composition. Textural features, grain size and grain boundary characteristics influence the uniaxial compressive, point load and tensile strengths, porosity, water absorption, specific gravity, the abrasivity, aggregate crushing and impact value and dynamic (P-wave velocity) characteristics (e.g. Pang et al. 2010; Rigopoulous et al. 2013; Sajid and Arif 2015; Er and Tuğrul 2016; Aligholi et al. 2019). In igneous rock system, fine-grained rocks with basic chemistry have higher engineering quality and lower abrasiveness potential than acidic and coarse-grained ones (Aligholi et al. 2019). However, contrary results were also published (Sajid and Arif 2015) where average coarse-grained texture with multi-modal distribution resulted in stronger matrix with lower porosity and water absorption values. Abrasivity is proved to correlate with quartz content and size rather than the grain shape or the chemical composition (Erichsen et al. 2011; Hofer et al. 2013; Er and Tuğrul 2016; Krutilová and Přikryl 2017). In long-term performance, resistivity to wear is also dependent on the petro-mineralogical composition and grain size (Gökalp et al. 2016)

Considering the above summarized observations on monomictic aggregates, some statements about multi-source aggregates and about the Hungarian alluvial sediments can be made. In the case of natural oligo-polymictic sandy gravel mixtures, the contrasting hardness of rock-forming minerals (i.e. the average mineralogical composition of the lithotypes) is determinative (Krutilová and Přikryl 2017). It means that the predominance of the quartz-rich lithotypes (approx. 60–70% in MD and 40–60% in NW samples) improves the resistivity to physical effect (abrasion, fragmentation). However, subordinate lithotypes (volcanites, sedimentary and low-quartz metamorphic rocks) will partly modify these values due to their heterogeneous texture (e.g. phenocryst content in volcanites; Krutilová and Přikryl 2017). Textural features (evolved interlocking of grain boundaries, recrystallized grain assemblages) of predominant quartzitic rock

types in the Hungarian sandy gravels positively influence mechanical properties (e.g. compressive strength). The additional, non-quartzitic rock fragments can positively affect this when considering recrystallized textured volcanic (Török and Czinder 2017; Czinder and Török 2019, 2020) or fine-grained sedimentary lithotypes. The above-detailed studies demonstrate that the selection of an optimum aggregate for a specific concrete application is essential. In the case of the Hungarian alluvial clastic sediments, it is not possible to characterize the assemblages with a common material property since these have oligo- or polymictic composition with different rock types. Although the quartzite and quartz-rich lithotypes are predominant, at least 20% of the grain populations have different lithology which modifies the physical properties. Based on these preliminary observations, a detailed laboratory testing (e.g. micro-Deval, UCS, PSV tests) is planned to provide quantitative data for asphalt fillers or road surfacing concrete applications.

Based on the analytical and petrological results, all sand and gravel samples have SiO_2 content over 80 w%. It is present in different silica forms, i.e. macro-microcrystalline quartzite, cryptocrystalline radiolarite and macrocrystalline quartz content of granitoid rocks, volcanites or sandstones. In addition, SiO_2 is also a major component of the silicate minerals (feldspar, amphibole, mica, clay minerals) of metamorphic and igneous rocks. The major constituents besides SiO_2 , are Al_2O_3 , Fe_2O_3 , K_2O and Na_2O . Aluminium is mainly related to the alumo-silicates, especially clay minerals. The alkalis and the iron are also major components of the silicate minerals. The CaO content is relatively high in both regions (1.6–4.9 w%) which is almost entirely derived from the calcareous (mainly limestone) lithofragments. Chemical composition analyses of the samples rated as a function of particle size revealed that elemental variability increases with decreasing grain size, i.e. the coarser pebble fractions contain more quartz-quartzite. Trace element distribution of sand and gravel samples show moderate depletion relative to UCC which indicates the relatively mature character of these sediments. All the trace element concentrations decrease with increasing particle size, except for Cr, which has a larger concentration in gravels than in sands. Gravels of the Mid-Hungarian MD-1 and 2 quarries have the lowest trace element contents.

The pebble petrography indicates relatively high maturity with quartz-dominated metamorphic-metagranitoid rock fragments, as a weathering product of crystalline rocks. The subordinate role of sedimentary and volcanic rocks is related to local sources (input from tributaries). Fine pebble petrography showed similar compositions to the macro-scale data but with diagnostic differences between the two regions. All the sampled sediments are dominated by metamorphic quartzite clasts, while low-quartz-content metamorphics, igneous and siliciclastic sedimentary rock fragments are

subordinate. In the Mid-Hungarian region, volcanic rocks are more significant; on the contrary, in the NW Hungarian region, limestone is characteristic. The heavy mineral analysis also confirms this observation. In addition, the Mid-Danubian region has alluvial sediments with a multi-source origin (metamorphics of both the Bohemian Massif and the Alps; younger volcanites of Börzsöny-Visegrád Mts.; limestones of Gerecse/Pilis/Buda Mts.), while the NW Hungarian Danube sediments are less varied (metamorphics of both the Bohemian Massif, the Alps and the NW Carpathians; limestones of Alps/Leitha Mts./NW Carpathians) (Thamóné Bozsó 2002; Bradák et al. 2014; Szeberényi et al. 2014; Józsa et al. 2020; Arató et al. 2021). Due to the high silicate content (i.e. a high water-affinity), the observed alluvial sediments are relatively susceptible to stripping (incompatible to asphalt), so their use as raw materials in asphalt industry is less favorable.

Bulk chemical composition of monomictic aggregates usually is considered during prediction of ASR potential (e.g. volcanites, Medeiros et al. 2020; Krutilová and Přikryl 2017), especially when the petrographic modal analysis is impossible due to the very fine-grained nature of the studied rocks. Another application of major element composition is for evaluation of abrasion resistivity (Er and Tuğrul 2016). In our research, we aim to apply the minor and trace element compositions to predict the activation risk of aggregates in a radiation environment, e.g. in the shielding concrete of nuclear facilities (power plants, research infrastructures). Concrete structures in such facilities are exposed to high doses of ionizing radiation and therefore can suffer mechanical degradation, and radioactivity might be induced during their service period, depending on their isotope composition (Ichikawa and Koizumi 2002; Denisov et al. 2012; Saouma and Hariri-Ardebili 2014; Pignatelli et al. 2016; Rosseel et al. 2016). Long-lived radioisotopes remain radioactive long after the shutdown of the facility (Islam et al. 2016) which requires special attention during the decommissioning. To produce radiation-resistant concretes with low activation susceptibility it is essential to select the proper ingredients, e.g. aggregates that contain the isotopes forming later the long-lived radionuclides only in low quantities. Heavy mineral studies have been done to complement and support the results of elemental analysis. It is known from the literature that the trace elements responsible for the long-lived radioactivity are enriched in heavy minerals of the sand and gravel (Islam et al. 2016). The chemical compositional study revealed significant differences among trace element abundances of sandy gravels from the NW Hungarian and the Mid-Danubian regions. In the case of Hungarian clastic sedimentary formations, the problematic isotopes are found to be accumulated in mineral species like garnet (Shimizu 1975; Smith et al. 2004; Ranjbar et al. 2016; Warren et al. 2018), amphibole (Parker and Fleischer 1968; Li et al. 2017), pyroxene (Shimizu 1975; Smith et al. 2004), rutile (Meyer et al. 2011) or zircon (Belousova et al. 2002; Claiborne et al. 2010).

It can be highlighted that there are some aggregates with a remarkably lower or higher ‘impurity profile’, originating from different mines within the Hungarian alluvial sediments. Impurity has a different context when concretes of open-air supporting or inside structures subjected to heat or other intensive radiation are considered. For outdoor concrete walls or blocks, the harmful constituents that are sensitive to chemical attack (e.g. sulphate or ASR) are the micro-cryptocrystalline SiO₂ varieties, iron sulfides, ferric and ferrous oxides, free lime and free magnesia (with alkali-, oxidation-, hydration- or carbonatization-reactive chemical properties). For indoor concretes suffering continuous thermal, electromagnetic or particle radiation, the activation potential of the constituents is an additional fundamental parameter; this is determined by the chemical isotope contents carried in by the mineral phases. In the observed samples the sole harmful component is the high silica content in varied forms. However, its high concentration anticipates the possibility of the pessimum behavior (Stanton 1940; Hobbs 1988) without the danger of ASR processes. In addition, ASR assessment needs to consider the cumulative amount of coarse and fine aggregates, in order to evaluate the behavior of the material. During the investigation of British river, glacial, beach and marine gravel deposits of aggregates, Rayment et al. (1990) found that the most ASR-reactive constituents are the cherts (due to microchalcedony content), siliceous mudstone-limestone (due to crypto-microcrystalline silica and chalcedony), metaquartzite and slightly metamorphosed sandstone (due to strained quartz). For Scottish sand and gravel deposits, any harmful constituents owned only a small proportion due to the very varied polymictic composition. These results indicate that the multi-source composition of alluvial to marine deposits can significantly affect the behavior of the aggregate. Well-known reactive minerals are reactive silica (micro- or cryptocrystalline quartz, submicroscopic or disordered quartz, strained quartz, opal, calcedony, tridymite, cristobalite, devitrified glass) and fine-grained greywacke (e.g. French 1991; Beck et al. 2000; Danielsen and Nebdal Svendsen 1991, Anastasio et al. 2016) and may be present in aggregates in small amounts (2 w% opal, 20 w% greywacke).

Alkali-reactivity of some rock types commercially used in Hungary was assessed (Fernandes et al. 2018). Felsic (sandstone, granite) and intermediate rocks (andesite) proved to have a higher potential to alkali-silica reactivity than basic rocks (diabase) due to their higher silica content. However, further studies, recommendations or guidelines are not provided on a domestic scale. A more detailed study on commercially used and available aggregate types was published and recommended by Fernandes et al. (2016). ASR sensitivity of granitic and quartzite rocks was also investigated (Kerrick and Hooton 1992; Monnin et al. 2006; Hagelia and Fernandes 2012; Ramos

et al. 2011, 2016). Dolar-Mantuani (1983) and West (1991, 1994) pointed out the undulatory extinction angle as a possible indicator. According to Ramos et al. (2016), there is a positive — but moderate — correlation between the amount of microcrystalline quartz, the undulatory extinction angle and the ASR potential (especially in the lower value region of the plot). However, other authors claimed that there is a direct correspondence between the undulatory extinction angle of quartz and ASR sensitivity (Andersen and Thaulow 1989; Ramachandran and Beaudoin 2000). They related the ASR potential of aggregates to their micro-cryptocrystalline silica content. According to the Concrete Society (1987), aggregates containing >30% highly strained quartz (av. UEA>25° for min. 20 quartz crystals) are regarded as potentially alkali-silica reactive.

In the observed Hungarian alluvial sandy gravel formations, the predominant strained quartz component (>60%) belongs to the weakly deformed type (UEA<14°). However, about one-third of this silica (20–25%) has an average grain size below 100 µm which has to be considered as deleterious component (though only a subordinate part can be attributed to chalcedony). This amount determines the sediments to be potentially reactive (Class II) to ASR. In addition, the textural characteristics of the dominant quartzite lithotype implies that ASR microcracks might rather appear along aggregate grains, while intra-grain cracking would be rare (Šachlová et al. 2016, 2017).

Further lithotypes with potential ASR (schist-metagranitoid, granitoid, andesite, siliceous limestone, radiolarite) can show alkali content which proved to be an inert feature in the alkali access of ASR processes (Drolet et al. 2017). Studies on monomictic volcanic aggregates (e.g. basalts; Korkanç and Tuğrul 2005; Medeiros et al. 2020) indicated ASR-related microcracking along the aggregate grains; though, this phenomenon was affected by the glass content of volcanites. In case of the Hungarian oligomictic formations, neutral volcanites (mainly andesite) contain fresh or recrystallized matrix carrying the ASR sensitivity.

Alkali-silica reactivity is a deleterious process especially in open-air concrete structures (concrete roads, bridges) where those are directly exposed to humidity. It can also have potential in large and thick concrete structures (e.g. some meters thick constructions) where real dry state of the body can never be reached. Besides, high-temperature environment can also accelerate the initiation of ASR (e.g. heating systems, radiation-induced heating load). These application fields of concrete can be impacted by ASR damage if the investigated Hungarian aggregates are used.

Conclusion

This paper evaluates the applicability of Hungarian oligomictic alluvial gravel formations for concrete and asphalt manufacturing. These specific sediments of the Danube, one

of the main rivers of Europe, bear the traces of a complex formation history with multiple sources. This makes these aggregate types less strictly classifiable than monomictic rock aggregates exploited in open-air pits. Multi-source clastic sediments cannot be determined by single values of physical properties but can behave more flexible and less predictable than monomictic aggregates. A detailed petrographic and textural characterization is even more important than for monomictic materials and can provide a suitable complement to reliably predict engineering properties of oligo-polymictic sedimentary rocks. Complementary chemical and heavy mineral investigations proved to be useful in evaluation of the activation potential of aggregates for application in radiation shielding concrete structures.

It was an aim to link the petrological characteristics of the gravel formations and their possible applications in the concrete and asphalt industry field. These so far missing or undervalued parameters are decisive for many civil engineering properties and can improve the dialogue between specialists and the producers. A future geotechnical assessment will be necessary to reveal aggregates that are satisfactory for use in specific construction fields.

The two regions on the middle course of the Danube — Mid-Danubian and NW Hungarian regions — have similar sandy gravel formations on the bases of macro-microscopic petrographic, heavy mineral and geochemical investigations. However, moderate differences deriving from the distinct geological positions on the river section could be revealed. Due to their chemical and petrographic compositions, the investigated gravel formations from Hungary are appropriate aggregates for conventional applications of concrete (e.g. house walls, basements), while their application to open-air constructions (e.g. bridges, concrete covering of roads) might be problematic due to their possible alkali-silica reactivity. For such purposes, crushed clastic materials of monomictic composition (mined rock types) are the better choice in case of low ASR properties. Another option is the use of supplementary cementitious materials or blended cements which can control ASR or limit the alkali content of the concrete. The direct application of these Hungarian alluvial gravel formations for road construction purposes is more appropriate than for asphalt manufacturing or railways ballast for which utilization crushed gravel would be beneficial.

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interpreted those data and revised the manuscript. HI participated in the fieldwork, measured and evaluated the PGAA data. SzL revised the manuscript and managed the related research grant.

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Declarations

Conflict of interest The authors declare no competing interests.

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