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Report 0: Bauxite deposits and bauxite residue accumulations in the ESEE region: What we learned from previous studies in Croatia, Hungary, Montenegro and Slovenia

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CONTENT

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1. INTRODUCTION

Bauxite deposits are by far renowned for their importance in the industry of aluminium, followed by the usage of bauxite raw materials in cement, chemical and industry of abrasives.

Along with world large resources of bauxite like those in Guinea, Australia, Vietnam, China or Jamaica, Europe's most significant ones are those in Circum-Mediterranian area, and thus East and Southeast Europe is Europe's most important bauxite production area.

This report is to summarize so far available date on bauxite deposits in the ESEE region focusing on Croatia, Hungary, Montenegro and Slovenia. Also, the report includes available data on red mud accumulations in these countries. All the data presented originate from previous research and prospecting work on bauxite deposits in the partner countries. Abundances of rare earth elements (REE) in bauxite resources are given in cases where available

The aim of the report is to give an insight and background for further assessment of potential of bauxite and bauxite residue for a recovery of rare earth elements in the ESEE region. Rare earth elements are much needed commodity in nowadays industry, with a European demand almost completely covered by imports made worldwide.

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2. COUNTRY OVERVIEW

2.1. Croatia

2.1.1. Bauxite deposits in Croatia and bauxite REE abundances

Bauxite deposits in Croatia vary in their origin, age, size and economical importance. They are widespread and confirmed at numerous locations in the country.

By type of formation the Croatian bauxites are divided into three groups (Marković, 2002):

1. ***Bauxite deposits including clayey bauxite, bauxitic clays and clays of Triassic, Jurassic and Neogene age.***
   Triassic deposits are found in the Slunj area and Lika. They were formed by bauxitic processes in situ during emersion between Ladinian and Carnian age, which was related to the beginning of the Alpine orogeny. The parent material was largely clayey (kaolinite) derived by weathering of the Ladinian clasts and pyroclasts, and then deposited in large relief depressions. After drying out, large deposits of clayey bauxite were formed by lateritization of kaolinite producing böhmite and diaspore. These are found in exchange with bauxitic and kaoline clays. The origin is inferred from spatial relationship of the bauxite deposits and clastic deposits as well as from similarity of accessory mineral associations in clastic deposits and bauxite.
   Jurassic clayey bauxites occur at numerous localities in Istria. They are similar to the Triassic ones by their formation, however, origin of clay materials has not been revealed yet. Since the Malmian mainland was characterized with a low relief, the material was unlikely to have been transported by rivers from distant areas but was rather sourced from weathering of the carbonate bedrock. An aeolian deposition has been suspected as a source of the parent material, too.
   Neogene bauxite deposits of this type were similarly formed by bauxitigenic processes in situ but from a more diverse source material. Partly this was a material derived by weathering of the carbonate bedrocks, but also from aeolian sediment derived by weathering of metamorphic rocks and tuff. The Neogene deposits are found in Karlovac County (Tounj) and Dalmatia (Peruča, Trilj).

2. ***"Terra rossa" type of bauxite*** – Lower Paleogene (Istria, Dalmatia and islands) and Upper Paleoegene (Dalmatia).
   Lower Paleogene bauxites were deposited after Laramian movements on paleorelief developed on the Upper Cretaceous limestones. They are preserved close or along the boundary with transgression sediments, so called Kozina deposits.

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Upper Paleogene bauxites were formed during emersion caused by Pyrenean orogenic phase in Middle Eocene, and were accumulated in the paleorelief of Upper Cretaceous and Lower Paleogene limestones, more rarely Kozina deposits. They are overlaid by transgressive Promina sediments, which were deposited during Middle and Upper Eocene as well as Lower Oligocene. Formation of both Lower and Upper Paleogene bauxites has been explained by formation of *terra rossa* due to a weathering of carbonate bedrocks and accumulation of insoluble fraction thereof. Some contribution of aeolian material is also possible.

3. ***Mechanically redeposited bauxite deposits***
   These deposits were formed by mechanical redeposition of already existing bauxite deposits. They are usually found within the Promina deposits and are not economically significant.

Economically the most important bauxite deposits exploited so far are those developed at the boundary of Upper Cretaceous limestones and Paleogene deposits as well as those in the contact of Upper Cretaceous/Lower Paleogene limestones with Middle Eocene/Lower Oligocene deposits (Promina). Minor quantities of Triassic (Karlovac County), Jurassic (Istria) and Neogene (Sinj) bauxites have been exploited for the industries other than aluminium production (cements, abrasives, bricks).

Mining of bauxite for aluminium industry in Croatia was continuous from 1914 till 1990. It has been evaluated that roughly 27.5 million tonnes of bauxite had been recovered in this period. After 1990 the bauxite mining gradually ceased, and today only smaller quantities are recovered for cement industry (Vujec, 1996)

Abundances of rare earth elements in bauxite deposits of various ages and genesis were determined by Crnički (1987) and more recently by prospecting study of Croatian Geological Survey at 190 locations of various bauxite ages. The results of the later study are summarized in Table 1.

Table 1. REE abundances in Croatian bauxites determined by the Croatian Geological Survey prospecting work at 190 locations (Database of the Croatian Geological Survey). The individual REE abundances are summarized as total REE, total LREE (La-Eu) and total HREE (Gd-Lu).

Deposit age	Bedrock age		Average (ppm)	Range (ppm)
Early Triassic (Carnian)	Early Triassic (Ladinian)	***Total REE***	314	109-569
		LREE	230	68-436
		HREE	84	34-235
Jurassic	Jurassic (Kimmeridgian)	***Total REE***	606	580-633
		LREE	465	446-485
		HREE	141	134-147

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Deposit age	Bedrock age		Average (ppm)	Range (ppm)
Lower Cretaceous	Not identified	***Total REE***	466	254-631
		LREE	374	211-521
		HREE	92	43-122
Upper Cretaceous	Upper Cretaceous	***Total REE***	487	432-566
		LREE	405	354-471
		HREE	82	75-94
Paleocene	Not identified	***Total REE***	539	142-1348
		LREE	415	95-1237
		HREE	124	47-403
Paleocene	Upper Cretaceous	***Total REE***	563	200-1409
		LREE	433	148-1157
		HREE	129	53-410
Promina (Middle Eocene-Oligocene	Upper Cretaceous	***Total REE***	791	406-5199
		LREE	632	313-4711
		HREE	159	93-488
Promina (Middle Eocene-Oligocene	Early/Middle Eocene	***Total REE***	1031	536-2082
		LREE	693	402-1456
		HREE	338	105-861
Promina (Middle Eocene-Oligocene	Not identified	***Total REE***	1038	515-3149
		LREE	790	398-2870
		HREE	248	94-651
Jelar (Middle Eocene)	Upper Cretaceous	***Total REE***	666	622-695
		LREE	497	468-514
		HREE	169	153-181
Neogene	Upper Cretaceous/Upper Eocene	***Total REE***	466	362-584
		LREE	359	294-457
		HREE	108	50-143
Quaternary	Upper Jurassic/Upper Cretaceous/Upper Eocene	***Total REE***	316	296-345
		LREE	244	226-264
		HREE	72	66-81

2.1.2. Red mud accumulations in Croatia

Production of aluminium ceased in early 1980s having left a red mud accumulation nearby former alumina factory in Obrovac, Zadar County, the former enterprise of the Jadral company alumina production. The estimated amount of the red mud is 1.3 million tonnes. The attempts to remediate two bauxite residue pools in the past decade ended by adding a substantial amount of construction material waste in the pool, thus making any attempt for future re-use of the bauxite residue for REE recovery most likely unviable.

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2.2. Hungary

2.2.1. Bauxite deposits in Hungary and bauxite REE abundances

Production of bauxite and alumina in Hungary has a long history of mining and processing operations, which have been accompanied by numerous professional and scientific work in the field.

Hungary's Transdanubian Range is well known for its Cretaceous-Tertiary bauxites. They all belong to the group of karst bauxites (overlying karstified carbonate rocks) and occur at major regional unconformities of Albian, Turonian/Senonian, early Eocene and Oligocene age. Additionally there is a fifth (mid-Miocene) likewise subaerial unconformity characterized by kaolinitic red soils and redeposited bauxite-pebbles, however, no real economy-grade deposits are associated with this youngest horizon. There are also some early Cretaceous (Berriasian) bauxite indications in South Hungary in the Villány hills. Their REE concentration is slightly higher than that of all the other Hungarian occurrences, however, they have never been exploited on the large scale and were not processed to alumina either.

1. ***<u>Transdanubian Range</u>***
   The Transdanubian Range (TR) belongs to the ALCAPA megatectonic unit. In Mesozoic times, it was part of the Southern shelf of Neotethys thus sharing the evolution of the depositional environment of the future Easter Alps. As a result of successive phases of the Eoalpine deformation, its Late Mesozoic carbonate sequences (including also the overlying early Paleogene rock-suite) are interrupted by several unconformities, four of which were accompanied by long-lasting subaerial exposure and bauxite formation. As a result of subaerial exposure in all cases typical karst relief was created and partially or completely filled with bauxites. More recently, based on an integrated study of bauxites, their associated bedrocks and the early diagenetic features of their cover, Mindszenty et al. (2000) attempted to incorporate bauxites into the currently available paleogeodynamic reconstructions. They concluded that the observed distribution of bauxites in the TR is in accordance with foreland-type geodynamics controlling Cretaceous and partly also Eocene deformation of the area, put forward by Tari (1994). In this context Cretaceous bauxites can be considered as weathering products formed and partly redeposited on the apex and the flanks of a migrating gentle Cretaceous forebulge, in the Senonian already involved in thrusting. In the Paleogene the geodynamic scenario seems to have changed inasmuch as the morphology of the deposits shows the imprints of large-scale strike-slip movements, probably related to the beginnings of the „escape" of the Transdanubian Range from its original East-Alpine position (Kázmér & Kovács 1985).

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Lithofacies and micromineralogy of the three bauxite horizons are different. Albian and Senonian bauxites, though both displaying distinct oolitic-pisolitic textures are different in terms of porosity (Albian: 6%, Senonian: 25 to 28 %). Eocene bauxites are either pelitomorphic or intraclastic to gravelly with pseudo-ooids only. Their micromineralogy substantially changes with time (Mindszenty et al, 1991). In the scarce (0.01%) acid-insoluble residue of Albian bauxites titanite, amphibole, kyanite and some calc-alkaline igneous rock-fragments were detected, whereas in the Senonian ones only the ultrastables (zircon, rutile, tourmaline), some calc-alkaline igneous and very few anchimetamorphic rock-fragments could be identified. Eocene bauxites are an order of magnitude richer in detrital minerals, in addition to the ultrastables they abound in higher metamorphic minerals and rock fragments (garnet, staurolite, sillimanite, kyanite), euhedral volcanogenic zircon and ilmenite grains and even some volcanic rock fragments of trachytic texture were identified in them. Zircon grains were fission-track dated as Eocene by Dunkl (1992) pointing to contemporaneous volcanic activity contributing to the pre-bauxitic material. Latest results of Kelemen et al. (2017) provided an insight not only into the nature of the parent rocks of the bauxitic weathering products but - by single crystal dating zircons from the micromineralogical residue of Cretaceous and Eocene bauxites – also into the whereabouts of the possible source terrains of bauxites having accumulated on the then exposed karst surface of the TR. They showed that at the time of the formation of the Cretaceous deposits only Jurassic and older rocks were exposed in the wider surroundings of the TR. Carboniferous and Permian siliciclastics, Ladinian „Pietra verde"-type volcanoclastics of probably South Alpine origin and perhaps also windborn tephra of some distant CAMP origin could be supposed as the source for the pre-bauxitic material. In the case of the Eocene bauxites the contribution of contemporaneous ashfalls were proved already by Dunkl (1990) and 1992 by fission track dating of euhedral zircon grains.

2. ***South Hungary/Villány Hills***

The Villány Mts belong to the Tisza megatectonic unit and - as it was shown first by Géczy (1973) and reinforced later on e.g. by Voros (2012) - unlike in the TR, Mesozoic formations of this lithospheric segment show clear northern Tethyan affinity. In early Mesozoic times the Tisza unit belonged to the passive northern margin of the Neo-Tethys Ocean and formed part of the European shelf. In the Jurassic, because of the accelerated Central Atlantic rifting, the Tisza „fragment" became gradually detached from paleo-Europe and developed into a „microcontinent". The Villány Zone was a central, relatively elevated sector of this microcontinent, where subsidence was apparently slower than in the adjacent areas. It was partially isolated by deeper-water sections from its surroundings. Apparently as a result of changing intra-plate stresses, in early Cretaceous times, the interior of this lithosphere segment became subject to

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gentle up-bulging bringing about the transient subaerial exposure of part of the previously full marine depositional environment (cf. with Cloetingh, 1988)

The result of the exposure is the Nagyharsány bauxite occurring within the Late Jurassic/Early Cretaceous shallow-water carbonate platform succession at an erosional unconformity between latest Jurassic (Tithonian) and Early Cretaceous (Berriasian to Valanginian) strata. As shown by biostratigraphy of the immediate bedrock and cover the age of the bauxite is probably intra-Berriasian, i.e. the subaerial episode was relatively brief (at least the minimum apparent stratigraphic gap occurring in it is much smaller than that in the case of any of the bauxite deposits of the TR). Unlike the TR bauxites, on the outcrop scale, no appreciable angular unconformity between bedrock and cover can be observed here. Also the mineralogy is different from the bauxites of the TR: major Al-minerals of the Nagyharsány bauxite are böhmite and diaspore (as opposed to the prevailingly gibbsitic-böhmitic of the TR bauxites. Though at places where bauxite fills meter-scale dolines, the major Fe-minerals are hematite and goethite (reddish colour, suggesting early diagenesis under vadose conditions!), at places where the underlying karst relief is shallower, the bauxite is pale-coloured and often contains chamosite, suggesting ambivalent redox conditions during early diagenesis. The lithofacies of the bedrock and the cover together with the concordant appearance of the bauxite-filled shallow-karst relief and the relatively short apparent stratigraphic gap all point to a temporarily exposed isolated pelagic platform as the depositional environment of the Nagyharsány bauxite. The same relative isolation is reflected by the rather poor detrital mineral assemblage in the micromineralogical residue.

The source of the pre-bauxitic material is not simply the dissolution residue of the karstified bedrock but probably there was an admixture of aeolian (partly perhaps also pyroclastic) contribution too, as was suggested already by Noszky in 1952, then more recently by Császár (2002). Noszky suggested that the contemporaneous diabase volcanism of the Mecsek Mts (currently situated to the North of Villány) could be blamed for the pyroclastics, while Császár insisted that rather the alkali (phonolitic) volcanism might have been the source of the windblown tephra supposedly deposited over the exposed carbonate platform of the Villány block. This seems to be supported by the results of Nagy (1989) according to whom in addition to a few heavily worn zircon and quartz grains, ilmenite was the only mineral which could be identified in the <0.063 and the 0,063-0,125 and 0,125-0,2 mm size-fraction of the bauxite.

No large-scale exploitation was ever undertaken in the Villány zone. During World War II between 1941 and 1944 a modest amount (cca 38 000 tonnes) of it excavated from former exploration adits, was shipped to the Ajka aluminium plant where it became mixed with Mid Cretaceous (Alsópere) and Eocene (Nyirád) bauxites, and was partly processed into aluminium,

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partly exported to Germany and allegedly also to Sweden (to be converted into alundum) (Gádori-Szepeshegyi, 1978; Nagy, 1989)

Along with some open-pits the majority of the workable reserves were exploited by underground mines. Important deposits subject to mining activity were Alsópere (Albian) Halimba and Iharkút (Santonian), Nyirád, Csabpuszta, Bakonyoszlop, Fenyőfő, Iszkaszentgyörgy, Gánt, Nagyegyháza, Csordakút, Mány (Eocene) and Óbarok (Oligocene). They have been under development since 1926 with a total annual production of cca 3 million metric tons in the late 1980-ties. Since then, for economic reasons, production has gradually declined until bauxite mining came to its end in Hungary in 2013 with Halimba-II SW as the last underground mining. Since in Hungary all mining companies are obliged to recultivation, most of the former open-pits are refilled by now.

Bauxite is accessible only in some of the old pits of Nyirád Darvastó, Iszka-Kincses, Gánt and Óbarok (Eocene), and some material is available also from the old deposits of Cretaceous bauxites of Alsópere and Iharkút. After final closure of the mines some selected core samples, originally stored by the Exploration Company, were transferred to the Geological Survey's Data Bank, and some of them are hosted by the Natural History Museum of Zirc (Transdanubia).

REE determination in Hungarian bauxites has been quite extensive. The first geological/geochemical overview of the REE concentration of Hungarian bauxites was published by Maksimović et al in 1991. They analysed 130 samples from 12 deposits with the aim of using the vertical distribution of REE's and other trace elements to prove or disprove the predominantly allochthonous vs autochthonous nature of the studied bauxites. Their working hypothesis was based on Maksimović 1988, namely that in deposits having been subject to long-lasting in situ development in the vadose zone mobile REE's and trace elements would be leached out from the upper part of the deposits and concentrated at the bedrock contact. Also they wanted to see whether – regarding the REE content - there was any difference between bauxites belonging to the different stratigraphic horizons. Y and La were analyzed only. Their concentration never surpassed a few tens or a few hundred ppm but there was a clear tendency of the South Hungarian (Nagyharsány) samples showing, on the average, higher concentrations than any one from the Transdanubian Range. In South Hungary also the expected downward enrichment of Y and La was obvious. Within the TR, Cretaceous bauxites showed somewhat higher values than the Eocene ones. Obviously allochthonous bauxite of Gánt did not show any characteristic REE-distribution pattern in the vertical profile.

In 1980, Stefániay, V. and his co-workers of HUNGALU's Engineering and Research Center (ALUTERV_FKI) has carried out a detailed geochemical study of bauxite samples representing six major deposits of the TR (North Bakony: Dudar and Bakonyoszlop (Eocene), Iharkút and Németbánya (Cretaceous), and South Bakony: Csabrendek (Cretaceous and Eocene). REE-

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concentrations of most analysed samples proved to be on the average, an order of magnitude higher than those analysed by Maksimović et al. (1991), and there was an order of magnitude difference between the extremes, as well.

In the National Research Center of Mining Geology and Engineering Cornides et al. (1985) carried out a representative study of bauxite samples from all important Hungarian deposits. By mass spectrometry and neutron-activation analysis they investigated a series of trace elements and Y, La, Ce, Nd, Gd, Yb and Hf, however the documentation stored at the Data Bank is by far not complete, with REE concentrations of the individual samples being not clear.

In 2013 in the frames of a project aimed at a general study of all potential REE enrichments of Hungarian igneous, metamorphic and sedimentary formations also some bauxites and red-muds were analyzed by Török et al. at the Hungarian Institute of Geology and Geophysics.

Total REE abundances of the most important Hungarian bauxite deposits are presented in Table 2.

Table 2. Total REE abundances determined in bauxite deposits in Hungary (Török et al. 2013; Stefániay, 1980-81; Data Bank of the Hungarian Geological and Mining Survey)

Locality	Bedrock age	Cover age	Average (ppm)	Range (ppm)
Csővár	Upper Triassic	Quaternary	123.5	90-227.9
Nézsa	Upper Triassic	Quaternary	291.6	190.6-423
Szendehely	Upper Triassic	Quaternary	120.8	16.9-274.1
Dudar	Upper Triassic	Quaternary	1729.3	862-2620
	Upper Triassic	Mid Eocene	1660.8	532-3509
	Upper Triassic	Oligocene	1015.7	557-1616
Bakonyoszlop	Upper Triassic	Mid Eocene	1085	1065-1102
Iharkút	Upper Triassic	Quaternary	3404.1	9384-648
	Upper Triassic	Oligocene-Miocene	1003.5	532-1616
	Upper Triassic	Upper Eocene	3445.4	756-6484
	Upper Triassic	Upper Cretaceous	3073.8	862-6654
Németbánya	Upper Triassic	Quaternary	2540.9	902-3879
	Upper Triassic	Upper Cretaceous	2869.4	1095-5388
Csabrendek	Upper Cretaceous	Early-Mid Eocene	3520	1032-10268
	Upper Triassic	Upper Miocene	3021.8	1398-4917
	Upper Triassic	Mid Eocene	3089.3	1277-6357
	Upper Triassic	Early-Mid Eocene	6861.3	4223-11856

2.2.2. Red mud accumulations in Hungary and REE abundances therein

The first small-scale Hungarian aluminium-producing capacity was established in the early ’30s at Magyaróvár (presently called Mosonmagyaróvár) and simultaneously a few thousand tonnes

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capacity aluminium smelter was set up by the Weiss Manfred Concern in Budapest/Csepel). At the same time the idea to establish another aluminium plant at Almásüzitő was also put forward (Várhegyi 1984). However, development of the future large scale vertically integrated aluminium industry of the country started in the outh Bakony area at Ajka-Tósokberénd where next to the newly discovered bauxite reserves of Halimba and Nyirád, a 20 thousand tonnes capacity aluminium plant and a 10 thousand tons smelter was set up in the early ’40s (Gádori-Szepeshegyi, 1987).

The Ajka site was particularly favourable not only because of the close vicinity of several bauxite deposits but also because the energy requirement of the plant could be met by the Ajka Coal Mines then providing 200 thousand t/year fuel to the industrial establishments. In those times, the alumina plant was fed by 50 thousand tonnes of bauxite annually. The Ajka aluminum industrial center has been active ever since then and processed bauxite arriving from both underground and open-cast mines of the Bakony area (Nyirád, Haimbva, Szőc, Iharkút, Németbánya, Fenyőfő, Dudar). In 1991 the smelter was shut down for economy reasons and from 2000 onwards, in addition to the domestic reserves, the aluminium plant began to use more and more bauxite imported from Bosnia and Montenegro. In 2006 they stopped producing smelter-grade alumina. The bauxite residue left over by the Bayer process between 1943 and 2011 (in all about 30 million tonnes) has been stored in ten red-mud cassettes. In 2011 because of the catastrophic dam failure and red-mud spill from Casette No.X., the production was switched over to the dry technology. By now all the older cassettes have been recultivated; even No.X is covered by a thin layer of soil and is already partially revegetated. The plant – currently operated by MAL Ltd (a private company being the legal assignee of the former state-owned Hungarian Aluminum Corporation) - is still in operation. Its current products are gallium, various kinds of special alumina, zeolites, and Al-alloys.

The Almásfüzítő Bayer plant was active between 1950 and 1997 with an initial capacity of 60 thousand t/year (raised to 360 thousand t/year in 1970). The processed bauxite was supplied mainly by the bauxite mines of the Vértes Hills (Gánt) and of the northernmost parts of the Bakony area (Iszkaszentgyörgy-Kincsesbánya). In 1994 also the Almásfüzítő Plant stopped producing smelter-grade alumina and for its last three years it switched over to special aluminium production. The bauxite residue produced at the Almásfüzítő Plant between 1950 and 1997 amounts to cca 17 million tonnes and is accommodated by eight cassettes. Cassettes Nos. I to VII storing all about 12 million tonnes of red mud, were built on the floodplain of the Danube within the administrative boundaries of the village, while cassette No. VIII. is situated slightly to the north, within the administrative boundaries of Neszmély. This last one was operated between 1986 and 1997 and it accommodates about 5 million tonnes of red-mud. Cassettes Nos. I to VI are completely recultivated (covered) by now. The only cassettes still partially open are Nos. VII and VIII.

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The Mosonmagyaróvár Alumina Plant has been almost continuously active since 1934. Currently it is owned by a private company called Motim Ltd. They stopped producing smelter-grade alumina in 2002 and currently their production is restricted to various refractories, alundum, other kinds of special alumina and various commercial goods which have nothing to do with the aluminium industry. The byproducts of the Bayer process (according to some estimates amounting to 8.76 million tons) have been stored in six cassettes.

Currently there are no active smelting capacities in Hungary.

REE concentrations in the analysed red-muds in Hungary so far, surprisingly enough, show no particular enrichment as compared to the bauxites. According to Ochsenkühn—Petropoulou et al. (1994) at the end of the Bayer Process there should be an at least twofold enrichment in the red-mud as compared to the processed bauxite. However among the analysed 72 Hungarian red mud samples there were 28 samples of which REE concentration has reached at least the order of magnitude of 103 (6 from Ajka's cassette No. IX; 19 from Almásfüzítő and 4 from Magyaróvár). Trying not to draw premature conclusions, we suggest only that this apparent anomaly may be assigned to two possible factors: (1) The aluminium plants were fed by a mixture of bauxites extracted from deposits or parts of deposits characterized by different REE contents and therefore eventual high REE concentrations may have been diluted and/or (2) In the case of the old storage depots, interaction of the red-mud with infiltrating rain-water and/or with stagnant or slowly moving groundwater may have resulted in mobilization and translocation of some of the REE's (this is why it would be imperative to collect systematic information about the total thickness of the storage cassettes as well).

An important conclusion of this preliminary evaluation is that the Ochsenkühn-Petropoulou principle cannot be applied mechanically to all red-mud storages. Estimation of the REE-potential of any particular red mud accumulation has to be undertaken with extreme caution and should be supported by thorough exploration of the history of the depot concerned (as reflected by the thickness, internal structure and preferably also by documents of the filling history).

Measured total REE abundances in the Hungarian red mud accumulations are presented in Table 3.

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Table 3. Total REE abundances in red mud accumulations in Hungary (Dobosi et al., 2011; Török et al. 2012; Data Bank of the Hungarian Geological and Mining Survey)

Accumulation locality		Average (ppm)	Range (ppm)
Ajka	***Total REE***	910.7	435.7-1231
	LREE	711.6	328.9-965
	HREE	199.1	106.8-268.3
Almásfűzítő VII	***Total REE***	998.5	345.5-1415
	LREE	776.4	267.3-1149
	HREE	212.1	78.1-287.6
Mosonmagyaróvár	***Total REE***	1444.7	1341.8-1572.2
	LREE	1110.5	1058.9-1169.7
	HREE	334.7	269.3-402.5
Neszmély	***Total REE***	1050	972.7-1127.4
	LREE	840.8	772.9-908.6
	HREE	209.2	199.7-218.8

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2.3. Montenegro

2.3.1. Bauxite deposits in Montenegro

Montenegro belongs to the area of the southeastern Dinarides. Bauxite deposits in Montenegro can be divided into: a) red karst bauxites of Triassic, Jurassic and Lower Paleogene age, and b) white karst bauxites of Lower Cretaceous age (Pajović, 2009).

Economically most significant bauxite deposits in Montenegro are Jurassic red bauxite deposits, which are widespread within structural-tectonic unit of High Karst. These were formed on the karstified paleorelief composed of Late Triassic, Early Jurassic and Middle Jurassic – Oxfordian carbonate sediments. The bauxite accumulation depressions are most prominent in Late Triassic limestones hosting the most important red bauxite deposits of Montenegro. These are discordantly overlaid by thick transgressive Late Kimeridgian and Tithonian limestones.

Previous studies indicate REE to be related to the various groups of minerals like bastnäsite which is a common autigenic mineral. Also, monazite-(La), monazite-(Nd) and goyazite are present in lesser amounts. Major mineral association of the bauxite Vojnik-Maganik and Prekornica region is composed of böhmite, kaolinite, gibbsite, hematite, goethite, anatase and calcite; and accessory minerals like zircon, ilmenite, magnetite, biotite, feldspar, mottramite, monazite, xenotime, and REE carbonates. The occurrence of residual and autigenic monazite and xenotime indicates a REE origin from primary sources, as the other REE phases are related to the bauxitization processes. REE tend to be concentrated in the lower part of the bauxite bodies emphasizing a strong vertical distribution, especially in bauxites formed on Late Triassic and Early Jurassic carbonates. The geochemical footprint indicates the Jurassic bauxites of Montenegro to be similar to the Jurrasic karstic bauxites from Turkey and Cretaceous karstic bauxite in Italy with shales, sandstones and intermediate igneous rocks as a parent material. The distribution of Ni and Cr groups the Jurassic Montenegrin bauxites together with other Jurassic and Cretaceous karstic bauxite of Turkey, Greece, Italy, Serbia and Slovenia, pointing the source of the alumosilicate material towards the rocks with mafic igneous composition. These are most likely ophiolites from the Western Vardar Zone, accounting also for a volcanic ash of the same composition and aeolian transportation followed by bauxitization processes in the conditions of tropical and subtropical climate.

Resources of the Jurassic karstic bauxites in Vojnik-Maganik and Prekornica ore region are estimated at 78 million tonnes. Average content of total REE abundances is presented in the Table 4.

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Table 4. Average total REE abundances in Montenegro deposits overlying bedrock of different ages (Radusinović et al., 2017)

Bedrock age	Average (ppm)	Range (ppm)
Upper Triassic	1053	518-7027
Lower Jurassic	1115	645-4145
Middle Jurassic – Oxfordian	1071	662-2069

2.3.2. Red mud accumulation in Montenegro

In Montenegro there is a red mud accumulation situated next to the today UNIPROM-KAP (former KAP) factory in Podgorica. Since alumina production ceased in 2009, the red mud basin is not filled with red mud any more. There is roughly 8 million tonnes of red mud accumulated there. No REE abundances have been determined therein so far.

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2.4. Slovenia

2.4.1. Bauxite deposits in Slovenia

There are several bauxite occurrences in Slovenia, however, since these are mostly smaller and irregular bauxite deposits, frequently with an unfavourable silica content, they are not exploited for aluminium industry. and there is no domestic bauxite mining. The most important deposit are those found in Bohinj-Rudnica, Hrast, Logarje, Kamniška Bistrica, Kokarje-Žifernik, Kopitov Grič, Kozina, Podlipa, Rjavi Grič and Vranja Peč (Budkovič, 2009). So far, there has been no determination of REE in the Slovenian bauxites.

2.4.2. Red mud accumulations in Slovenia

In Slovenia there is an abandoned disposal site for red mud from alumina production, which is situated near Kidričevo, a town that is located approximately 2 km west of an industrial zone in the southeast part of the country. The disposal site covers an area of 42 ha. The overall depth of the deposited red mud is not known, but has been estimated to be, on average, about 4 m. The landfill is located in a water protection area (Koren & Lapajne, 2017).

History

In 1942, the German company Vereinigte Aluminium Werke began to build a factory for the production at Kidričevo. The selected flat area of the Dravsko polje region, where hospitals and camps were located during World War I, was very suitable for the construction of a large industrial complex since it had already been partly communally equipped, and there was a railway nearby. The construction works were completed in 1946, when this area already belonged to Yugoslavia. In 1948 the factory was renamed as the "Boris Kidrič" Factory of Alumina and Aluminium, and designated as a state company of federal importance.

The regular production of alumina and aluminium started in 1955. The Bayer process was used to process alumina from bauxite ore. The capacity of the factory was 45.000 tons of alumina and 15.000 tons of aluminium per year. In 1957, a contract was concluded with the French company Pechiney, according to which the production capacity was increased to 80.000 tons of alumina per year. In 1963, the electrolytic hall B, with a production capacity of 42.000 tons of aluminium per year, was built. In 1991, the production of metallurgic alumina ceased. By then 3.4 M tonnes of metallurgic alumina had been produced, and approximately 1.6 M tonnes of red mud had been generated. The red mud was deposited directly on the existing soil, which consisted of a mixture of siliceous and carbonaceous gravel and sand, and only in a small area was the red mud landfilled into an abandoned gravel pit. The bottom of the gravel pit is in an area where there are high levels of groundwater, but it is not known to what depth the red mud reaches. Also, it is unknown whether it is in direct contact with groundwater nor whether the red mud

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has been deposited at groundwater level in an unsaturated area of an aquifer. Up until 1965, the red mud was transported to the landfill by wagons, whereas up until 1991 a pipeline was used for this transport, in which the red mud was in the form of a suspension. According to available historical data, the red mud was washed with water before disposal.

The activity which caused the greatest perturbation in the general public was a programme about the proposed ecological rehabilitation of the red mud landfill (Figure 1), which could be seen from the air as a large red lake from which red dust rose during dry weather. In 1992, a first tree was symbolically planted on the rehabilitated landfill, and it was officially closed in 2007. However nothing else was done except for the covering layer, so these activities cannot be considered as efficient rehabilitation of the landfill. In the same year, the factory was renamed as the company: Talum d.d. In 2004 a solar power collection plant was built on top of the landfill.

Figure 1: Bird's view of the landfill in 1994 (source: Surveying and Mapping Authority of the RS, digital ortophoto, 3.8.1994)

Hydrological situation

The landfilling of red mud is the responsibility of the management body of Talum d.d. It has no properly designed drainage system - neither for drainage waters nor for rainwater. These layers represent a hydro-dynamically permeable open aquifer. In the area of the landfill of red mud the groundwater is fed indirectly by rainwater from the western side of the Pohorje mountain. The results of the permeability measurements performed in observation wells showed that coefficient of permeability ranged between 5x10-5 m/s and 5x10-4 m/s.

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The direction of a groundwater flow in the area of red mud landfill is from the west towards the east. Effective porosity was estimated to be in the range of 0.1 – 0.2, whereas the groundwater flow rate was estimated to be 6.7 m/day. The groundwater is located at a depth of between 4.0 and 10.0 m beneath the surface of the red mud landfill (Koren & Lapajne, 2017).

No surface water occurs in the immediate vicinity of the landfill. The closest water sources are: towards the southwest, the river Reka (distanced approximately 2.5 km from red mud landfill), which flows into the river Polskava; on the eastern side is the Struga stream (which is distanced approximately 6 km from the red mud landfill), and flows in the direction of the groundwater stream. Further on there is the Central Drava Channel (which is distanced approximately 7 km from red mud landfill) (Koren & Lapajne, 2017).

The highest point of elevation of the red mud landfill is located in the landfill's western part and reaches a height of 252 m above sea level, whereas the lowest point of elevation is located in the landfill's eastern part and reaches a height of 242 m above sea level (Koren & Lapajne, 2017).

The chemical characteristics of red mud

The environmental impact of red mud was assessed, based on a modified Tessier's sequential extraction and speciation analysis procedure, in order to estimate the partitioning of elements between easily and sparingly soluble mud fractions (Milačič and el, 2012). Additionally, chemical speciation of Al and Cr was performed in the highly mobile water-soluble red mud fraction.

The pH of the water soluble fraction which has a critical influence on the Cr and Al speciation was 9, taking into account the fact that the red mud has already been partly neutralised. The results of a performed speciation analysis showed that, at a pH of 8, the Al present in the red mud exists mainly as an insoluble hydroxide, whereas the Cr species, at a pH of 9, are distributed between $Cr(OH)_3$ and Cr(VI). The total concentration of Al in the water-soluble red mud fraction was 17.3 ± 0.5 mg L^{-1}. The concentration of the $[Al(OH)4]^-$ species, which is highly mobile and extremely toxic towards living organisms in terrestrial and aquatic habitats, and significantly contributes to the hazardous environmental impact of red mud, was found to be 11.1 ± 0.3 mg Al L^{-1}.

So far the effects of trace elements have not been determined nor REE abundances measured.

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3. CONCLUSION – Some guidelines for further evaluation of REE potential in bauxite and red mud within REEBAUX project scope

Previous studies of the bauxite deposits in the focus countries revealed an extensive coverage of bauxite occurrences providing a detailed geology description and a display of models proposing bauxite genesis and origin of parent material, which have still not been resolved completely. REE analyses of the bauxite deposits in the countries have been sporadic through the decades, sometimes not covering determination of all individual elements of the REE group. This was frequently dependent on the analytical methods available at the times. More recently, as interest for REE increases in respect of genetic interpretations of bauxites as well as in a wider focus of industrial and technological requirements, more systematic approach in investigation of the bauxite REE abundances has been launched. On the other hand, REE abundances red mud accumulations, with exemption of Hungary, have attracted little interest so far, thus leaving an additional motivation for the activities within REEBAUX project.

Lessons learned from the previous studies point to the following guidelines to be considered in the forecoming project activities:

1. A common sampling methodology should be established for the prospecting in all the bauxite/red mud localities to be addressed within the project,
2. A common sample preparation and sample REE analysis procedure has to be defined among project partners,
3. A relation of REE abundances and bauxite deposits of certain age, occurrence and genetic type should be established in order to make assessment of REE potential more effectively,
4. A long period of bauxite investigation with more or less connection to the industrial sector requires a more elaborate approach in a study of bauxite-related resources for their utilization in the industries,
5. To make use of bauxite-related resources as a possible source for recovery of REE, a detailed understanding of their mode of occurrence in bauxite deposits and bauxite residue has to be clarified,
6. Bauxite residue, although being considered a valuable source of REE, with abundances usually at least twice as high as those in the bauxite raw material, seems not always to follow this trend (as evidenced by the previous studies of the Hungarian red mud); thus, factors like origin of bauxite material, technology of alumina production applied, ground

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water influence to the accumulation pools and mode of REE occurrence (mineralogy) in red mud have to be considered,

7. General decline of bauxite mining and related processing industry has been recorded in the region in past decades. In the focus countries the existing aluminium processing industry has been detached from the local mines which are either closed (example Hungary) or export the ore to the third parties (example Montenegro). Potential strategies for REE recovery from bauxite/red mud could bring local mining and processing industry to cooperate more closely again.

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ANNEX (overview of bauxite deposits localities in Austria with indication of historical mining)

Austrian bauxite-related resources were not planned for the report in 2018. However, an overview of the known bauxite deposits is presented (Table 5) with indication of the historical mining sites, since there is no more bauxite mining in the country. So far, no data on REE abundances in the Austrian bauxites have been reported.

Table 5. Overview of the bauxite deposits localities in Austria

Locality	Age	Ever exploited (yes/no)
Glanegg	Mid-Cretaceous	yes (closed)
Lugberg	Late Jurassic (?)	no
Grimming	Late Triassic	no
Unterlaussa - six deposits	Mid-Cretaceous	yes (closed)
Dreistätten	Mid-Cretaceous	yes (closed)
Hieflau	Mid-Cretaceous	no
Altenmarkt	Mid-Cretaceous	tests
Russbach	Mid-Cretaceous	no
Grossgmain	Mid-Cretaceous	yes (closed)
Widschschwenter Alm Kufstein	Mid-Cretaceous	tests
Brandenberg	Mid-Cretaceous	yes (closed)
Alland	Mid Cretaceous	no

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