Limonite, a hydrogeological phenomenon and the raw material for the Fennoscandian bloomery industry involving "*jernvinna*"

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This manuscript was written during their last research on the bog-iron ore phenomena. Arne Espelund died in August 2019 and Olav M. Skulberg died in January 2021. This article marks the end of the authors scientific endeavor to reveal important insights into this early industry.

Sammendrag

Myrmalm, et hydrogeologisk fenomen og råmateriale for den blomstrende industrivirksomheten knyttet til jernvinna. Boreale torvmyrer i hellende terreng, hvor grunnvannssig medfører anrikning av myrmalm (*limonite*), er det sentrale temaet som drøftes i denne artikkelen. Geokjemiske og geomikrobiologiske fenomener på slike lokaliteter dannet grunnlaget for jernvinna i den fennoskandiske region av Europa. Det var naturgitte forutsetninger knyttet til landskap med dystrofe vannforekomster, jern- og manganholdig grunnvann, og et kildemangfold, som skapte mulighetene til blesterteknologien. Den førte til smibart jern som ga øks og plog. Forskningsarbeidet som behandles i artikkelen, er basert på de omfattende undersøkelsene til Arne Espelund knyttet til jernvinna i Norge og naboland. På utvalgte lokaliteter, hvor jernframstilling av myrmalm hadde foregått, utførte vi feltarbeid med innsamling av prøver (jord, plante- og dyreliv). Dette materialet bidrar til informasjon om virksomhetene knyttet til den gamle og primitive jernvinna. Erfaringene av feltundersøkelsene og laboratoriearbeidet blir behandlet og kommentert i artikkelen vurdert i lys av synspunkter i relevant hydrobiologisk, geokjemisk og geomikrobiologisk faglitteratur.

Summary

This article provides observations and facts related to the peatland ecosystems in boreal landscapes where compounds of iron and manganese are enriched via geochemical and geomicrobiological activity generating bog-iron ore (limnonite; $Fe_2O_3H_2O$). These resources within our geographic region fostered the bloomery industry in the Bronz- Iron Age. Our research work was initiated and based on the essential investigations performed by Arne Espelund on the ancient bloomery ironmaking in Norway. Together we have carried out field work on selected localities with furnace sites belonging to the period with expanding bog-iron technology. The results and experience obtained from the survey and laboratory investigations are discussed in relation to information and viewpoints in current professional literature. Boreal landscapes with peatbog ecosystems promoting deposits of bog-iron ores made the foundation possible for

the initial Fennoscandian exploitation of metallic iron, being based upon the natural resources including subterranean waters and springs with their chemical and biological activity. The *conditio sine qua non* for the bloomery industry,in the geographical Weichselian glaciated regions of the Northern Europe, realized via the human ancestral culture with its ironmaking success.

Introduction

Groundwater discharges pouring out in springs of gently sloping mires involve specific compounds of iron and managnese may be enriched as ochre via chemical and microbiological processes. The generated deposits consist of bog-iron ore (limonite - Fe_2O_3 H_2O). This peatland resource fostered the functional and cultural stage in our Bronze and Iron Age in Fennoscandia. In Norway the word "*jernvinna*" is the name of the primary technology resulting in malleable iron; axe and plow (Brøgger 1925).

This article aimss to provide av "digest" of the prevailing first hand professional knowledge on the considered matter. The pristine landscapes of Fennoscandia, with their special peatland ecosystems (Moore & Bellamy 1974),were in a way designated via nature to become the cornerstone for the prolific bog-iron technology and its human expanse.

In connection with the regional investigations that Arne Espelund performed during his research on "*jernvinna*" (reports 1999, 2004, 2013) field work was carried out on selected localities with ancient furnace sites. In these places samples also were collected to be used in microscopical search for biological objects and fragments associated with the bog-iron ore operations. The discussion is mainly based on results and experience from these laboratory studies and relevant literature.

The formation of bog-iron ore is described geological and geographical by Holtedahl (1953), Kuznetsov (1970) and Espelund (2004). The different types of localities include a manifold in environmental conditions giving rise to ferrigenous deposits, and to the ore qualities. When considering the microorganisms participating in the generation of this peculiar hydrogeochemical product, it turns out that they belong to taxonomic groups with important ecological role in peatbog environments, i.e. iron bacteria (sheathed bacteria, *Chlamydobacteriaceae*) and diatoms (species of pennate *Bacillariophyceae*).

A supplement with photographs and comments is included hindmost in the paper, based on studies of a favoured area with spring manifold. The intention is to visualize the bog-iron phenomenon by seaking evidence in a forest landscape with moorland sites in Rendalen, Hedmark. Bloomery iron making was performed here, as widely in the northern part of Østerdalen until the end of the 18th century (Evenstad 1790, Bull 1916, Brøgger 1941).

Siderotrophy; a premise for the bog-iron ores

"The water has a similar nature
as the ground it is streaming through".

Gaius Plinius (AD 23 - 79)

The geological Baltic shield extends across Fennoscandia, covering the Kola peninsula, Finland, parts of Sweden and Norway. In this territory the basement is primarily composed of gneiss, granite and quartzite (Rudberg 1961). The Precambrium bedrocks are hard (erosion resistant) and acid (poor in calcium). Podsolic soils are generally predominant in outlying fields and may include about 80 % of the woodland areas. Peat formation is widespread on mires where the water table of groundwater in the underlying geological sediments is high, bringing up vegetation producing plant biomass which exceeds its decomposition rate. The peatlands are geologically a consequence of the Weichselian glaciation (Holtedahl 1953, Moen 1999).

The peatbog ecosystem prevalent in bog-iron ore localities is mainly characterized by oligotrophic-dystrophic conditions (Brønmark & Hansson 2006). Humification processes ensure the drainage water to become typically rich on organic substances. The predominant boreal climatic factors in combination with ferrigenous deposits generate siderotrophy (Naumann 1932). This gives rise to suited conditions for microbial energetics and metabolism utilizing the content of available iron and manganese. Cycling of these elements via biogeochemical activity is performed by an array of adapted species of microorganisms (Kuznetsov et al. 1983).

Compounds of iron and manganese are entering the aquifer mainly in a soluble form, but partly as suspensions. In lakes and wells these metallic elements are oxidized and will precipitate to the bottom. Here they can accumulate, transformed as ochre. Or, it may change into a reduced soluble form, and then again diffuse into the free water mass (Gorlenko et al. 1983).

Landscapes characterized by siderotrophic environments with peatbogs supported the natural conditions for the bloomery industry. They came to be the foundation for the technological and cultural stage in the Bronze and Iron Age period in Fennoscandia (Magnus & Myhre 1976, Espelund 2013).

Microorganisms; participants in the bloomery industry

Iron bacteria

The main ores of iron in the Baltic shield are the oxides haematite (Fe_2O_3), magnetite (Fe_3O_4), and the carbonate siderite ($FeCO_3$), (Kukal 1971). The ability of these minerals to weather and release iron in solution is strengthen by acidity and the presence of humic substances (Gjessing 1976). The conditions in dystrophic waters, mires and soils are in such a way favourable for the formation of limonite,an impure hydrated ironoxide, via chemical and biological activities (Kjensmo 1967, Ehrlich 1988).

A number of selected microorganisms are capable of depositing iron and manganese on their cell surfaces (Gorlenko et al. 1983). Investigations of this accumulation of ferric hydroxide on the sheaths of iron bacteria (e.g. *Leptothrix ochracea*) resulted in the discovery that these organisms can oxidize ferrous iron (Brock et al. 1994). This process, together with the ready non-biological oxidation of iron within the pH domain 5 - 8, at which the bog organisms perform good growth, generates the bog-iron ore. The primary conditions for technological bloomery exploitation are in this way established.

Iron- and manganese-oxidizing eubacteria belong to the most important organisms being able to lay down ochre under watery conditions (Thunmark 1942, Ehrlich 1983). They are widely distributed, and have great ecological significance in freshwater and soils, in mass development visible to the naked eye. *Leptothrix ochracea* is probably the most distributed iron-storing, sheathed bacteria in Norway. This is an obvious species on all the localities investigated in this study. Their cells are inside an enriched iron-impregnated sheath (measure 0.8 - 2 µm), equipped with one flagell. During mass development they form flocculent material, in reddish-brown multitude (Häusler 1982). It also causes the "rusty" appearance of different natural water bodies.

Studies of the genus *Leptothrix* played an important part during the advancement of scientific microbiology, as earlier described in detail (Cholodny 1926).

Transformation of iron

The microbiological processes resulting in bog-iron ore depend on the physiological metabolism of filamentous, sheath-forming bacteria. The active species of bacteria can grow autotrophically, using ferrous iron as inorganic electron donors. They are termed as chemolithotrophic organisms.

The groundwater fostering wells in siderotrophic regions are rich with dissolved ferrous iron. Only a small quantity of energy is available via the oxidation of iron from the ferrous to the ferric state. For this reason, the bacteria must oxidize large amounts of ferrous iron to achieve necessary growth.

Iron exist in three oxidation states - Fe^{0}, Fe^{2+} and Fe^{3+}. At pH-values less than 5, the ferrous state Fe^{2+} is prevailing during anaerobic conditions in the well water. Before the microbiological oxidation, the ferrous iron is adhesive to organic compounds on the surface of the sheath-forming bacteria to increase its availability. The chemical output is limonite: $Fe_2O_3.H_2O$, or the brown iron ore. In Norwegian it is named "*myrmalm*" or "*brunjernstein*".

Diatoms and silica; their role in the bloomery iron making

Silicon, unlike carbon, is not found in the free condition. It constitutes more than one-quarter of the crust of the earth. The oxide (SiO_2) and its compounds are components of sand and many minerals. Also, the bog-iron ore (ochre) consists of an abundant fraction of silica (~ 20%) (Espelund 2004) and diatoms are usually significant contributors.

The major source of silica in fresh water originates from the decomposition of alumino-silicate minerals (e.g. clay) in the drainage basin. In free water percolating igneous rocks the delivered silica content is less compared with water draining geological quaternary deposits. However, the silica content of inland water is less variable than many of its other major inorganic constituents. The silica concentrations in surface water in East-Norway are generally in the range 1 - 8 mg SiO_2 per liter. Much of the variation is due to the utilization of SiO_2 in the diatom metabolism.

All cells of diatoms, unicellular photosynthetic microorganisms (Graham & Wilcox 2000), have a silica wall (frustule), in which silicic acid exists dehydrated and polymerized to form silica particles (opaline silica). The diatoms are among the most extensive and numerous eukaryotic organisms of waters and soils. They can produce sediments consisting of almost merely frustules (kieselguhr, diatomaceous earth), or as varying shares in the dy-sediments (a *gyttja* mixed with unsaturated humic-colloids of littoral and allochthonous origin (Hansen 1959).

When siderotrophic conditions are prevailing, the special sediment type termed *diatom ochre* can be generated. In its typical composition the sediment constitutes a mix between kieselguhr and iron ochre, with a content of iron varying between 15 - 30 % of the total substance of sediment. With increasing shares of diatoms, the sediment gradually changes to *diatom gyttja*.

The microfossils of diatoms in the sediments and water samples investigated in our regional study were mainly frustules and frustule-fragments of species from pennate genera (e.g. *Pinnularia*, *Cymbella*, *Frustulia*, *Surirella* and *Eunotia)*. Diatoms can in mass occurrence be a significant component of bog-iron ore. The content of silica in the ore may cause different practical and qualitative consequences during the bloomery processes (Espelund 2004, 2013).

Supplementary information

This supplement presents illustrations and explanations of phenomena observed on the catchment area around the timberline of River Mistra in Rendalen, former Hedmark county. The purpose is to inform about the peculiar nature with the former peatland iron-bloomery localities. Attention is centered on the geochemical, hydrobiological and microbiological objects and processes associated with the formation of bog-iron ore (limonite). Photographs by Vidar M. Skulberg.

The peatland environment in question, Misterkjølen, Rendalen

River Mistra is a 60 km long watercourse in the Østerdal Valley with a catchment area of about 540 km^2, being an effluent to the river Glomma via the watersystem Lake Storsjøen - River Rena. During centuries the peasant culture in this geographical area has been based upon agriculture, the forest and mountain dairy coupled with a supplementary "economy". In Rendalen the bloomery ironmaking may, during historical periods, have been such an important secondary income. This livelihood on farms continued from the Middle Ages (300 B.C. to 1500 A.D.) and at intervals up to the end of the 18th century.

The bog-iron ore excavation was primarily performed on the marshy highlands around 500 - 800 m.a.s.l. Misterkjølen is the name of this wide eastern plateau with boreal pine and birch forest on ground with acid and strongly base-

Figure 1. Looking out over the landscape situated southeast of River Mistra, a hilly, undulating plateau with treeline forest on ground varying in humidity and drainage. The scenery is seen against the lifting bluish skyline in east to northeast, with the conspicuous mountain ridge Rendalssølen (highest top 1755 m.a.s.l.).

Figure 2. In this spring the groundwater comes out at the bottom of the basin, and the overflow forms the beginning of a brook entering the minerogenic moorland, and running downhill to River Mistra.

deficient, boggy soils (Figure 1). Here were localities with supplies of raw material for the early production of malleable iron.

On the topographical maps of Misterkjølen (The Norwegian Mapping Authority, 2005) the relevant bloomery sites still have the place-names written as the ancestors used them at the time when blast furnace activity was done. Examples: Blæsterbekken, "*the brook near the place with furnaces for smelting the bog-iron ore.*" Jønsbuåsen, "*the hill nearby the hut where the iron workers had their lodging*".

Some characteristic species in the vegetation on these expanses at the tree line may be mentioned. They include reindeer moss (*Cladonia* spp.), cowberry (*Vaccinium vitis-idaea*), bilberry (*V. myrtillus*), crowberry (*Empetrum nigrum*), bearberry (*Arctostaphylos uva-ursi*), together with

Table 1. Chemical composition of the water

	Gitlaugmyrkilden*	Mineral spring**	Blæsterbekkmyra***	
Calcium	0.81	8.49	0.982	mg/l
Chloride	0.28	0.35	0.31	mg/l
Sulphate	0.86	0.38	0.24	mg/l
Conductivity temp.	22.6	22.8	22.8	°C
Conductivity	1.25	7.85	1.07	mS/m
Iron	5.3	467	245	µg/l
Manganese	1.54	518	91.4	µg/l
Silicon	3.68	6.26	1.21	mg/l
Total nitrogen	<50	105	225	µg/l
Total phosphorus	24	92	16	µg P/l

* NIVA - Analyseoppdrag 651-6697, 09.07.2018

** NIVA - Analyseoppdrag 651-4622, 30.06.2017

*** NIVA - Analyseoppdrag 651-4622, 02.07 2017

cloudberry (*Rubus chamaemorus*), bog-moss (*Sphagnum* spp.), cotton sedge (*Eriophorum vaginatum*) and dwarf birch (*Betula nana*).

Gitlaugmyrkilden (a limnokren), Fuggdalen nature Reserve

In the spring surroundings (Figure 2) the bog vegetation includes numerous species spread over a great range of humidities. The marshy surface is generally covered by bog-mosses (*Sphagnum* spp.). On this bryophyte carpet scattered grassy and sedgy plants are developing (e.g. bog cotton, *Eriophorum vaginatum*, and deer-sedge, *Scirpus caespitosus*). Along with these are specimens of heather (*Calluna vulgaris*), dwarf birch (*Betula nana*) and willows (e.g. *Salix lapponum*).

The temperature of the spring water shows little variations during the years, varying near around 4 °C. The quality of the water has also an unchanging regular character (Table 1).

Hydrobiological characteristics

The water in the spring basin has a phytoplankton with species typically for clear-water springs. Diatoms are the numerous group among the microalgae. Frequent pennate species e.g. *Fragilaria virescens*, *Pinnularia viridis*, *Eunotia arcus* and *Tabellaria flocculosa* are prominent.

A multitude of benthic algae are developing in the *Sphagnum*-overgrown water margin of the spring basin. Filamentous species of several genera - including *Microspora*, *Draparnaldia*, *Oedogonium*, *Klebshormidium* - grow vigorously here. An interesting species can be mentioned in this connection. The chlorophyte *Microthamnion strictissimum* is regularly observed in the benthic algal community.

Mineral spring (a rheokren), Fuggdalen nature Reserve

Groundwater discharge from the aquifer is pouring out in the mineral spring brook where compounds of iron and manganese are precipitated via chemical and microbiological processes. This ochre consists of the chemical substance limonite (Figure 3A).

The aquifer is of sedimentary origin. Quaternary deposits cover the valley slopes of Mistra. Vertically their thickness is of varying magnitudes between 50 - 100 m or more. The geological material is mainly composed of consolidated gravel, sand and silt. Accordingly, the subterranean water reaching the mineral spring has constant physical and chemical characteristics. The temperature in the water during the year is less than the mean air temperature of the surrounding discharge area. In the mineral spring the me-

Figure 3A. In this spring the aquifer water flows away from its mouth following the terrain gradient, forming a brook downwards to the River Mistra. In Norwegian the key word "ochre well" is named "raudvelle".

arured temperature fluctuates between 3- 5 °C, the average is 4.1.°C. The water quality in the mineral spring is similarly showing little variations through the year (Table 1).

Hydrobiological characteristics

The mineral spring community with its different levels in the food web, is the basis to produce the ochre, the pre-phase of bog-iron ore. The organisms include both aerobic and anaerobic species, with an interesting diversity for detailed scientific investigations. However, so far only introductory studies have been undertaken in the Fuggdalen mineral spring. Protozoa and other invertebrates are co-operative animal participants in the community. So far, they have not been examined.

Knowledge is therefore rudimentary of even the basic elements in the local complex of biodiversity in this aquatic environment. Iron bacteria occupies a first stage in the overgrowth of the mineral spring wet perimeter with benthic vegetation. They are developing a copious ochre, coating the sides and bottom of the stream, and forming fluffy fragments of filaments being the cause of the "rusty" appearance of the water. The shape and dimensions of cells, their sheaths and colony structure, have been used as taxonomic diagnostic characters.

The cells of *L. ochracea,* developing in one single chain, are constantly leaving their sheaths, and forming new capsular coverings (Figure 3B). When looked at in the microscope, the floating flocculants in the spring water are to a great extent consisting of empty iron impregnated structures.

L. ochracea is a ubiquitous, important species of sheathed iron bacteria. It is an active factor in the production of ochre deposits (limonite), in all parts of the world. The genus *Leptothrix* includes five species described in the prevalent systematic taxonomy of bacteria. The investiga-

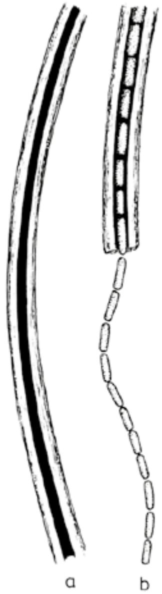

Figure 3B. A drawing of the species Leptothrix ochracea *developing in the Fuggdalen mineral spring. (a) showing an empty bacteria sheath, an ironincrusted capsule with diameter about 2 µm. (b) the cell chain of the iron bacteria leaving its filamentous sheath. (Original , Häusler 1982).*

tion of the diatom flora in the mineral spring resulted in the identification of more than fifty pennate diatom species living in the community of microalgae (Figure 3C).

Not to be surprised that a significant content of SiO_2 in the bog ore used for the bloomery ironmaking, descended from the biomass of diatoms living in the peatland habitats where the raw material was harvested.

The Blæsterbekk-mire with sloping fens and helokrens

The prevailing natural conditions in the highland mires in Rendalen are as disposed for the hydrogeological formation of bog-iron ore. Their grounds worked out of moraine material, and covered with peatland vegetation fostered by helokren springs, adapt siderotrophy with the essentials for the biogeochemical processes involved (Figure 4A).

The Quaternary mineral remains forming the basis of the Blæsterbekk-mire, are unsorted, with a totality of components differing in size

Figure 3C. Cymbella aspera *(Ehr. Cl.), this is one of the large diatom species in the Fuggdalen mineral spring (measured frustule: length 250 µm, width 58 µm). Behind , a glance into the microbial diversity feeded by the nutrient rich water from the aquifer. An oasis is this habitat, shaped in the post-glacial mineral deposits of sparagmite origin. (Photo. J. Sanecki)*

Figure 4A. This spring is located uppermost on the bordering of a sloping fen, a minerotrophic mire type evolved on the subsoil of the terrain with its hydrology. The groundwater is oozing out wide from the spring upon the moraine surface in different directions on the marshy area.

Figure 4B. Cross section of a ground moraine on Misterkjølen being the basis for an overlying, gently sloping mire (named Blæsterbekkmyra*) with groundwater supply via helocrens.*

Figure 5. A collection of bloomery slag pieces found on different sites in the Østerdal Valley with interest for archaeologists and metallurgists. Slag is a byproduct of bloomery ironmaking. It gives significant information about the operation and process made use of by the successful ironmakers long ago. The photo exemplifies some different morphological types of bloomery slag.

from boulders down to clay particles and smaller (Figure 4B). Horizontal below in the bulk of matter a layer of rusty deposits appear with iron enriched substance (ochre), an evidence of the siderotrophic groundwater. This precipitated matter is not the type of metalliferous ore used for bloomery activity. But the included ochre is suited to prepare pigments.

The bloomery ironmakers preferred a coarse ore, usually collected from selected places found at the rim of the wet sloping fens described above. This bog-iron ore type consists of pellets and lumps, brownish in color and easily crumbling between fingers. This was the raw material used for ironmaking in marshy highlands also in the Østerdal Valley.

Seen in the forefront on the Figure 4B is a cavity presented. In this trench the drainage water from the mire system enters, forming a creek bed finding outlet from the Blæsterbekk-mire. The water is running northward via River Renåa, downstream to River Mistra. The chemical quality of the drainage water is indicated (Table 1).

Final reflections

Bog-iron ore (limonite) was the raw material for bloomery ironmaking in Norway during the period 300 BC to 1500 AD, and at some places until the 18th century. The traces of this activity left behind on bloomery sites include remains of furnaces and slag heaps. They are among the foremost research objects in archaeology concerning ancient ironmaking (Figure 5). Arne Espelund with his scientific profession metallurgy, had of cause special preference for these bloomery remains. His ideal was to show the validity of the natural sciences in studying history.

Because of the great natural changes during some thousand years, there is no close correlation between the present-day ecosystem of boreal peatlands and the marshy landscape at the time when the bog-iron ore was generated. The type of good mineralized ochre for bloomery purposes was created in a distant past, probably during the termination of the Weichselian glaciated period of Northern Europe.

It is interesting to mention that although bog-iron ore globally constitutes in quantity rather small geological deposits, they are to-day the only active natural iron-bearing sediments being created, taking place in the boreal region of the formerly glaciated northern hemisphere. A hydrogeological phenomenon deserving continual scientific attention.

The aquatic nature of biotopes similar with the Fuggdalen mineral spring type where the subterranean water, confined in darkness, becomes released, trans-shaped to an open streaming water meeting daylight, prepare the birthplace of a novel species rich community of organisms. A remarkable hydrogeological phenomenon of metamorphose in living conditions, capturing the curiosity of scientists, especially among ecologists and biologists.

Epilogue and acknowledgements

The iron bacteria and diatoms have become evolutionary adopted to the special ecological niches of lacustrine and peatland environments generating bog-iron ore. Their metabolitic activities make out an important factor in the geochemical cycling of the elements iron and silicon in nature, including their role in the accumulation of ore deposits.

During the Iron Age, distinct in the viking period 900 - 1100 A.D., the bog-iron ore was exploited all over Scandinavia. The bloomery industry was in Europe extensively taken in use (Phillips 1980), in Norway during the approximate period 300 B.C. to 1500 A.D., and at some places until the 18th century.

Present-day refinery experiments with furnaces similar in design to Iron Age ones reveal their amazing efficiency (Espelund 1999). The technological skill of the pioneers and their novel iron tools helped expanding the Fennoscandian peoples subsistence, output and culture.

The reverse of the medal is a copious theme. The archaeologists experience that the same geochemical and microbial reactions generating bog-iron ore have given the Iron Age artefacts in Fennoscandia short longevity due to rapid corrosion. And the problems caused by iron- and manganese bacteria make worldwide regularly control restorations necessary in water supplies and drainage systems (Beger 1952, Kristiansen 1981, Stenvik & Hilmo 2020).

A final critical comment is worth mentioning. In Norway the spring habitats has received far less scientific attention than lakes and rivers. They are still understudied. That is a challenge in front of our onward water research.

The principal team of scientists in the research included J. Sanecki (phycologist) at the Polish Academy of Science, and J. Kotai (chemist), at Norwegian Institute for Water Research (NIVA). Their support and advice were constructive and fruitful for the work. The chemical analysis of water quality has been made at the laboratory at NIVA. The late authors were grateful for all the goodwill supporting this work.

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