MINDeSEA

Seabed Mineral Deposits in European Seas: Metallogeny and Geological Potential for Strategic and Critical Raw Materials

Deliverable 6.5: Report of the polymetallic nodules prospect evaluation for European waters based on data generated by this study

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6.5. Report of the polymetallic nodules prospect evaluation for European waters based on data generated by this study

Summary:

GeoERA is a Co-Fund ERA-NET action under Horizon 2020, towards "Establishing the European Geological Surveys Research Area to deliver a Geological Service for Europe". Its main objective is to contribute to the optimal use and management of the subsurface.

The project “Seabed Mineral Deposits in European Seas: Metallogeny and Geological Potential for Strategic and Critical Raw Materials” (MINDeSEA), materialized in the frame of the GeoERA Raw Materials Theme (Grant Agreement Nº 731166, project GeoE.171.001), resulted from the collaboration between eight GeoERA Partners and four Non-funded Organizations at various points of common interest for exploration and investigation on seafloor mineral deposits.

This document starts by giving a general and summary overview about the Polymetallic Nodules. Secondly, the identification and brief definitions concerning the critical and strategic elements are made. A summary of the results obtained during the MINDeSEA project is then addressed. The main part of this report is focused on the characterization of the Metallogenetic Areas defined, in this project, for the European Seas, with special emphasis on the evaluation, in terms of metal resource potential, of several critical and strategic elements. To better understand this characterization, critical and strategic element concentrations of Polymetallic Nodules, listed in the MINDeSEA database, in relation to their spatial distribution, are shown in various maps (Annex B). The Critical and strategic element concentrations of the Polymetallic Nodules, present in the distinct Metallogenetic Areas, defined in MINDeSEA project, are graphically compared among them and in relation to some reference marine regions, that have the most important and well characterized marine polymetallic nodules deposits (Cook Islands EEZ, Peru Basin, Clarion Clipperton Zone and Central Indian Ocean Basin)(Annex C). The generation of the Potentiality and Predictivity maps for the European Seas is further addressed and, at the end, the most significant conclusions are presented (a table summarizing the critical and strategic elements having significant potential as metal resources, in association to the correspon-

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1. Introduction

Polymetallic nodules are marine sedimentary mineral deposits, composed mostly of iron and manganese, that precipitate very slowly from seawater (hydrogenetic) or from bottom sediment pore waters (diagenetic), but most nodules are a combination of both (e.g. Kuhn et al, 2017). In Addition to these two elements, the polymetallic nodules contain numerous other elements, many of which are rare and valuable metals that can reach concentrations that are economically valuable, including some critical metals, and the rare earth elements (REE) (e.g. Hein & Koschinsky, 2014).

Polymetallic nodules can have different size, shape, and surface morphologies. Concerning the size parameter, nodules generally range from about 1 to 12 cm in their longest dimension. Some extremely large specimens of 21 cm have been found in the Peru Basin (von Stackelberg, 1997, 2000). but they can also have millimetric scale dimensions (micronodules). Typically, the most common dimensions are between 1-5 cm. In terms of shape, typically include spheroidal, ellipsoidal, elongated, discoidal, platy, botryoidal or irregular forms. Polymetallic nodules grow by mineral precipitation in concentric banded zones of micro-layers around a minute nucleus, that is commonly composed of a fragment of an older nodule, rock fragment, or even by biogenetic fragments (von Stackelberg & Beiersdorf, 1991).

The growth rate of hydrogenetic layers is typically in the range of 1–5 mm per million years, (Koschinsky & Hein, 2003), whereas diagenetic layers grow considerably faster (up to 250 mm per million years; von Stackelberg, 2000). Because most nodules form by both hydrogenetic and diagenetic precipitation, they grow at intermediate rates of several tens of mm/Ma years.

The chemical composition of nodules varies at different scales: on regional, local, inter- and intra-nodule. In general, the chemical composition of nodules is controlled by the ratio of the hydrogenetic to diagenetic components and whether the diagenetic component results from oxic or suboxic diagenesis (Glasby, 2000).

Hydrogenetic nodules have Mn/Fe ratios ≤5 (Halbach et al., 1989) and high contents of

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high field strength elements such as Ti, REE, Y, Zr, Nb, Ta, Hf as well as elements that can be oxidized on the surface of Mn oxides such as Co, Ce, and Te, low Y/Ho and high Th/U ratios (Koschinsky & Hein 2003; Hein et al. 2013). REEs in nodules are also of economic interest (Spickermann, 2012), but are generally two to six times lower than they are in Fe–Mn crusts (as an example, maximum total REEs plus Y in the Clarion Clipperton Zone - CCZ nodules are of about 0.08%).

Diagenetic nodules are characterized by much higher Mn concentrations than in hydrogenetic nodules, with Mn/Fe ratios >5 (Halbach et al., 1988). Traditionally, Ni and Cu have been the metals of greatest economic interest (with concentrations reaching more than 1 wt%), but this type of nodules are also significantly enriched in Ba, Zn, Mo, Li, and Ga (Lithium is especially high in diagenetic nodules, averaging 311 ppm in Peru Basin nodules (Hein & Koschinsky, 2014)). Other geochemical characteristics include: low Co and Ce concentrations, high Y/Ho ratios and low Th/U ratios (e.g. Wegorzewski, & Kuhn, 2014).

The most extensive deposits have been found in the Pacific Ocean, especially between the Clarion and Clipperton Fracture Zones (CCZ), the Peru Basin, and Penrhyn Basin (including the Cook Islands EEZ); a large nodule field also occurs in the Central Indian Ocean Basin (CIOB) (Fig.1).

2. Critical and Strategic elements

Critical Raw Materials (CRMs) are those raw materials which are economically and strategically important for the European economy, but have a high-risk associated with their supply due to the very-high import dependence and high level of concentration of set critical raw materials in particular countries. This risk is also coming from the fact that there is a lack of (viable) substitutes, due to the very unique and reliable properties of these materials for existing, as well as future applications. CRMs have a significant economic importance for key sectors in the European economy, such as consumer electronics, environmental technologies, automotive, aerospace, defence, health and steel. These materi-

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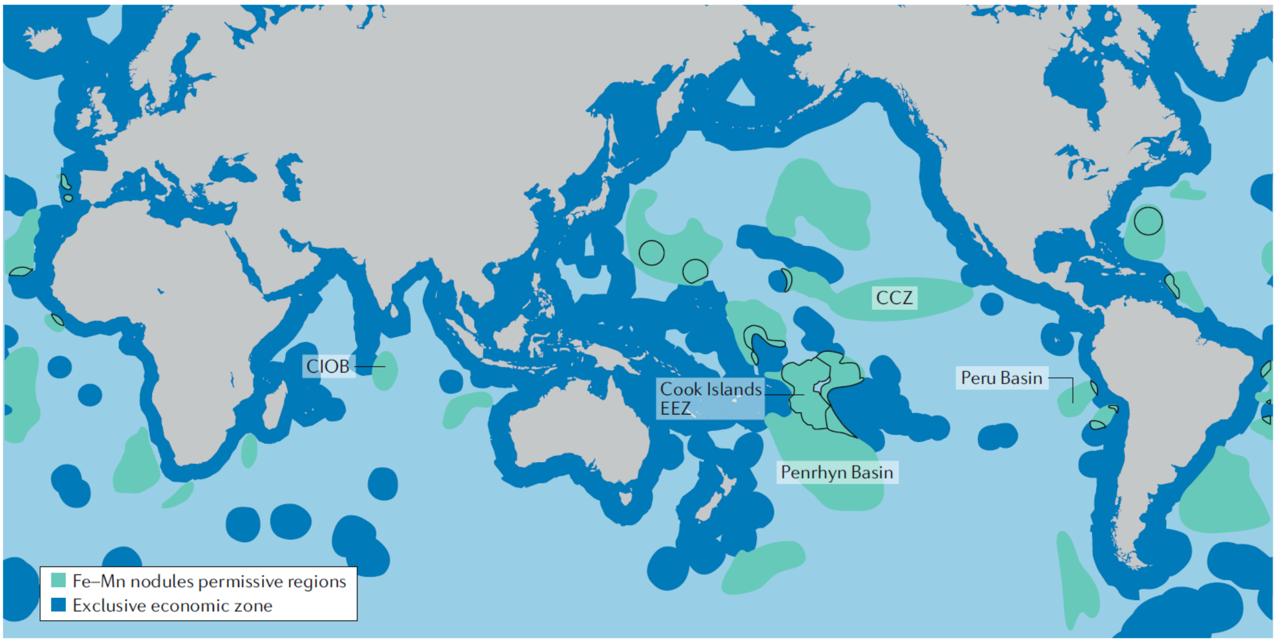

Figure 1. Global permissive areas for deep- ocean polymetallic nodule deposits. Global map showing the location of the polymetallic nodules permissive regions and the exclusive economic zones (EEZs). The four best known polymetallic nodule fields are the Clarion–Clipperton Zone (CCZ), Peru Basin, Penrhyn Basin (including the Cook Islands EEZ) and Central Indian Ocean Basin (CIOB). (Figure from Hein *et al.,* 2020)

als are not only 'critical' for key industry sectors and future applications, but also for the sustainable functioning of the European economy.

Critical Raw Materials (CRMs) are one of key subjects for the EU economy. The European Commission published the first list of CRMs for the EU in 2011, but due to the dynamic conditions controlling this criticality, such as the emergent technologies and economic factors, the list of these CRMs undergoes a triennial revision. In September 2020, Europe has considered 30 resources as Critical Raw Materials, three more than in 2017. In this latest list, four more resources were added, namely bauxite, lithium, titanium and strontium. Table 1 shows the 2020 critical raw materials. More detailed information concerning these CRMs may be obtained in https://ec.europa.eu/growth/sectors/raw-materials/areas-specific-interest/critical-raw-materials_en.

Important for this report is to identify, from this CRMs list, those elements that could be enriched in the polymetallic nodules. According to several authors (e.g. Hein et al., 2013,

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2020; Kuhn et al., 2017), these nodules can be strongly enriched in the following CRMs: Co, Heavy and Light Rare Earth elements, Nb, P, Sc, V, Li and Ti. All these CRMs were analyzed in detail through their concentrations listed in the MINDeSEA database, and compared among the marine regions from where they were collected. Additionally, all

2020 Critical Raw Materials (new as compared to 2017 in bold)		
Antimony	Hafnium	Phosphorus
Baryte	Heavy Rare Earth Elements	Scandium
Beryllium	Light Rare Earth Elements	Silicon metal
Bismuth	Indium	Tantalum
Borate	Magnesium	Tungsten
Cobalt	Natural Graphite	Vanadium
Coking Coal	Natural Rubber	**Bauxite**
Fluorspar	Niobium	**Lithium**
Gallium	Platinum Group Metals	**Titanium**
Germanium	Phosphate rock	**Strontium**

Table 1. EC 2020 Critical Raw Materials List

these elemental concentrations were compared with those existing in the four main marine mineral deposits for the polymetallic nodules: 1. Cook Islands EEZ; 2. Peru Basin; 3. Clarion Clipperton Zone (CCZ); 4. Central Indian Ocean Basin (CIOB).

Strategic elements are those that are usually defined as vital to modern technology, defense and industry and that have a high economic importance. They are also required for technological and industrial developments, but are in short supply and have no known substitutes. Comparatively to the CRMs, above defined, what distinguish the strategic elements from them is the fact that the supply risk is lower for the strategic elements. Both group of elements present high economic importance. Several strategic elements were identified has having the potential to be enriched in the polymetallic nodules, which include: Mn, Cu, Ni, Zn and Mo. In the same way as for the CRMs, these elemental concentrations were compared among the European marine regions from where they were collected and were also compared to the four reference marine regions identified above.

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3. MINDeSEA database: summary of its main features

Since the beginning of MINDeSEA project, from the beginning of the second semester of 2018, a compilation of polymetallic nodules occurrences in European seas was made from a range of resources, which included various scientific journals, that were searched through various databases, reports and thesis as well as cruise reports. National databases from the various geological surveys in Europe were explored and data library for polymetallic nodules was collected.

MINDeSEA database has listed a total of 296 polymetallic nodules occurrences. Their distribution is shown in Maps A1 and A2 (Annex A).

In Map A1, the spatial distribution of polymetallic nodules occurrences is made in relation to the MSFD (Marine Strategy Framework Directive) marine regions/sub-regions. The majority is located in the Baltic Sea (184 occurrences, corresponding to 62% of the total)

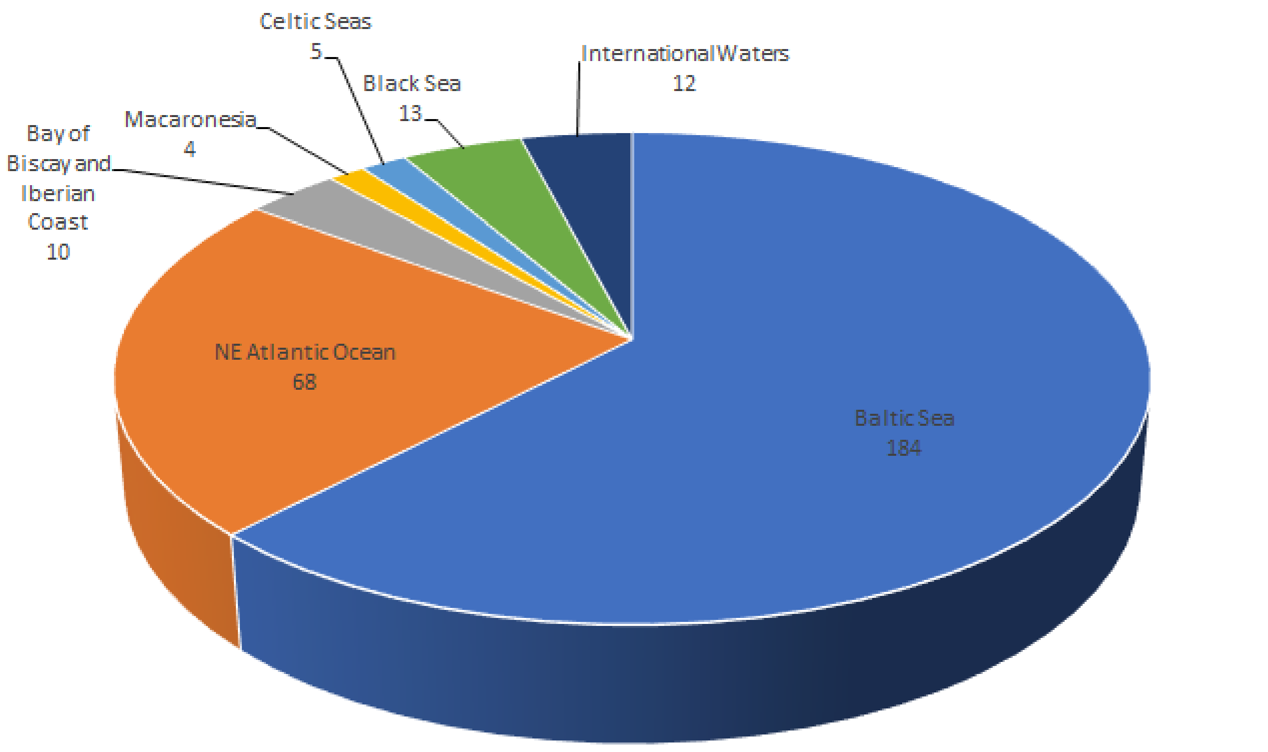

Figure 2. Distribution of the total Polymetallic Nodules occurrences listed in MINDeSEA (n=296) among the various MSFD regions/subregions and International Waters.

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followed by North East Atlantic Ocean (including some data points that are geographically inside the Arctic Ocean), having 68 recorded occurrences (23% of the total). The remaining occurrences (15%) are distributed through other five marine regions/sub regions. A graphical representation of this distribution is shown in Fig. 2.

In Map A2, the spatial distribution of the recorded occurrences is made in relation to the Economic Exclusive Zones. Sweden (99 occurrences, 33% of the data) and Russia (51 occurrences, 17% of the data) together have half of the total recorded occurrences.

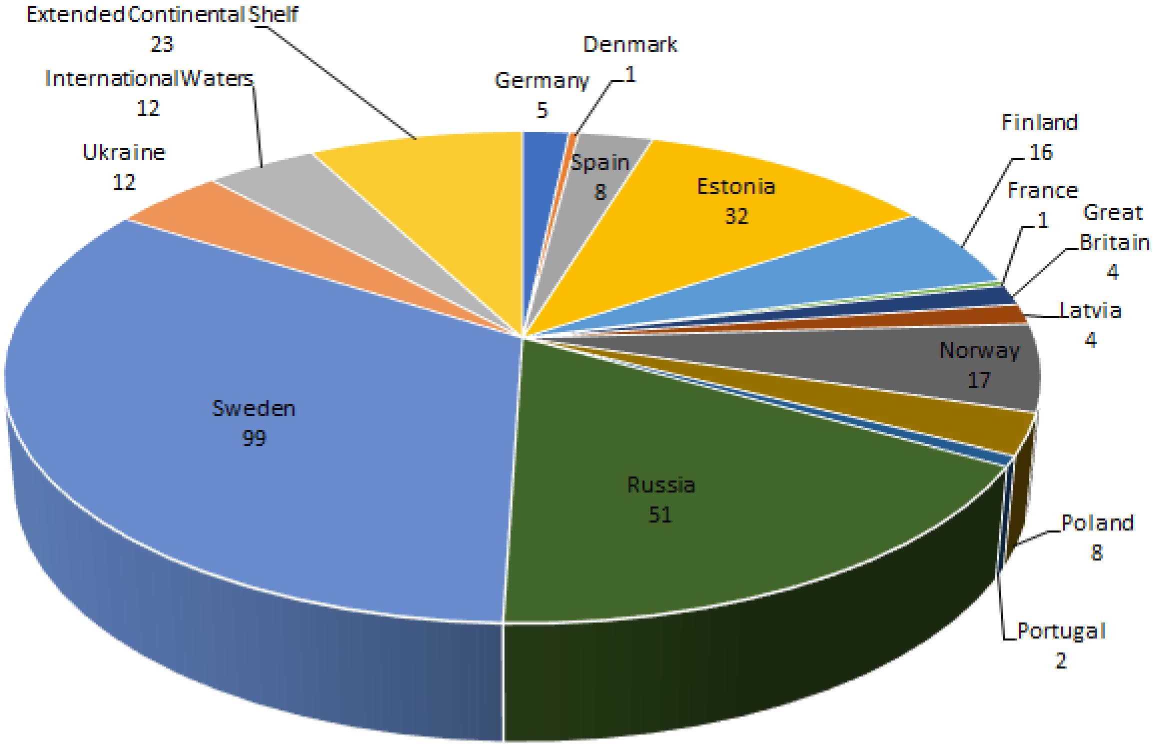

Figure 3. Distribution of the total Polymetallic Nodules occurrences (n=296) among the various EEZ's, also including the International Waters and Extended Continental Shelf.

About 4% of the data (corresponding to 12 occurrences) are located in International Waters, whereas 23 recorded occurrences (~8%) are inside extended continental shelfs of various European countries. A graphical representation of this distribution is shown in Fig. 3.

From the MINDeSEA database, defined along the project development, it is possible to

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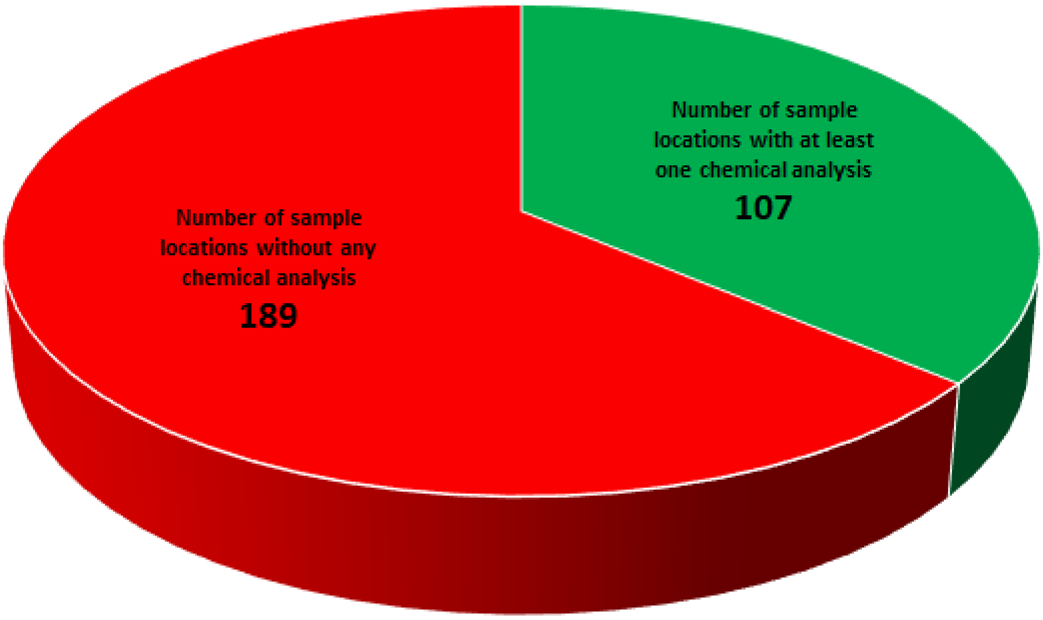

Figure 4. Graph showing the number of occurrences in polymetallic nodules with and without associated chemical analysis, listed in the MINDeSEA database

quantify the number of polymetallic occurrences that have chemical data associated. Thus, in Fig.4, it is possible to confirm that 64% of the total recorded occurrences (corresponding to a total of 189) don't have any chemical data information. Conversely, 107 sampling locations (36% of total data) provided chemical data for the polymetallic nodules collected in those locations. Some of these sampling locations have more than one nodule chemically characterized. Although the majority (72% of the data, corresponding to 77 nodules) of the sampling sites have just one nodule chemically characterized, some nodules occurrences (in a total of 28%) have more than one sample associated with chemical data (Fig. 5; Map A3). In this case, the MINDeSEA data base presents the corresponding average values. Due to their significant number of polymetallic nodules, chemically characterized, for a single sampling site, it is worth mentioning the following occurrences: I.D. 12 (in the Gulf of Cadiz, 38 nodules analysed); I.D. 25 (40 nodules analysed; Kiel Bay, Baltic Sea); I.D. 57 and I.D. 60 (40 and 30 nodules analysed, respectively; Baltic Sea proper); I.D. 78 and I.D. 84 (68 and 65 nodules analysed, Gulf of Finland, Baltic Sea, respectively)(Map A3). Taking into account all the chemical data compiled, the MINDeSEA database shows a total of 493 chemical analyses for polymetallic nodules sampled in 107 distinct locations. The six sampling sites (nodules occurrences) referred

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above contribute with 57% of all listed chemical analyses.

4. Metallogenetic Areas

Taking into account the spatial and density distribution of the polymetallic nodules compiled inside the MINDeSEA project, five metallogenetic areas were defined (Map A4), which include:

Metallogenetic Area 1 - Barents Sea (defining two separate areas, next to each other: Metallogenetic Areas 1a and 1b)

Metallogenetic Area 2 - White Sea

Metallogenetic Area 3 – Baltic Sea, divided into three sub-areas:

- Metallogenetic Area 3a - Gulf of Bothnia
- Metallogenetic Area 3b - Gulfs of Finland and Riga
- Metallogenetic Area 3c - Baltic Sea proper (including Kiel Bay)

Metallogenetic Area 4 - Black Sea

Metallogenetic Area 5 - Biscay Bay and Iberian Margin

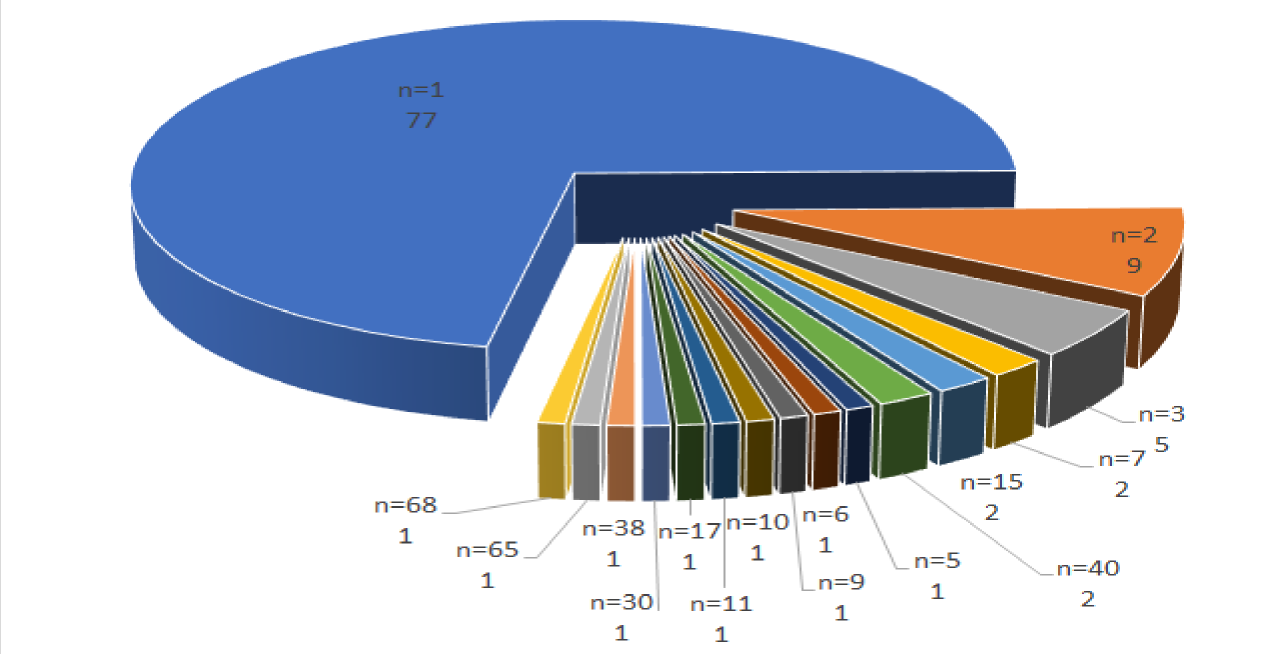

Figure 5. Graph showing the number of polymetallic nodules occurrences with a specific number (n) of analysed samples. Eg., n=1/ 77 means that there are 77 occurrences with1 nodule analysed.

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4.1. Metallogenetic Area 1 - Barents Sea

A group of 46 polymetallic nodule occurrences forms a first cluster of data points located North of Norway and East of Svalbard (Metallogenetic Area 1a; Map A4)

Chemical data are available just for six nodules, for the following elements of interest: Mn, Fe, P and REE (no data available for Y). The nodules are significantly more enriched in Fe (8.6-17.5 wt%), comparatively to Mn (0.02-0.14 wt%), giving very low Mn/Fe ratios (0.001-0.008), which are typical of polymetallic nodules of hydrogenetic origin. However, when considering the genetic diagram of Bau et al., (2014), using the available REE, where Ce_{Sn}/Ce_{Sn}^* is plotted against Nd, four data points are located in the diagenetic field, whereas two points have greater affinity with hydrothermal origin, showing that using distinct elements produce distinct origins for these nodules. Considering the abundances of strategic elements and elements including in the 2020 critical raw materials list, P concentrations vary between 1.2 and 1.8 wt%, which are higher than those presented by the nodules of the Cook islands region, Peru basin, CCZ and CIOB (all with P average concentrations <0.32 wt%) (Annex C). Total REE concentrations range between 0.004 and 0.014 wt%, which are well below the corresponding concentrations (0.040 – 0.168 wt%) found in those four iconic areas referred above (Annex C). Mn, as described above, have very low concentrations (0.02-0.14 wt%).

A second group of polymetallic nodules, with a total number of 15 occurrences, are identified immediately E to the first cluster described above (Metallogenetic Area 1b; Map A4). The density of the occurrences are smaller and chemical data is restricted to one sample. Using distinct chemical elements to assess the genetic origin, different results are obtained: low Mn/Fe ratios are indicative of a hydrogenetic origin; relationship between Ce_{Sn}/Ce_{Sn}^* and Nd indicates a diagenetic origin; Ce_{Sn}/Ce_{Sn}^* versus Y_{Sn}/Ho_{Sn} and the ternary diagram of Bonatti et al., (1972) (Co+Cu+Ni – Fe – Mn ternary diagram), both indicate a hydrothermal affinity. Comparatively to the four reference regions above referred, only interesting concentrations of P (3.05 wt%) and V (820 ppm) are found (Annex C).

Concerning the two metallogenetic areas, three major geomorphological settings are identified: Marine (37%), Continental Shelf (22%) and Coastal Plain (4%). The

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MINDeSEA database has only 16 entries on water depth, ranging from 148 to 385 m. About 59% of the Barents Sea occurrences have associated data for the host rock type, and the majority are clay (70%), but sand and silt are also found as substrate sediment. The age of the host rocks spans from Triassic to Paleocene (Cenozoic). The most frequent age (corresponding to 42% of the available age data for this sea) is from the lower Cretaceous (Albian). The data information concerning the morphology and texture of the nodules are very scarce, corresponding to less than 5% of the occurrences listed.

Since the morphotectonic setting of the Barents sea, which encloses the two metallogenetic areas above addressed, corresponds to a shallow basin, the environmental parameters favouring the formation of polymetallic nodules, defined for the most known and studied areas of Indian and Pacific oceans (CCZ, Peru basin, Cook islands region), whereas the morphological settings are totally distinct, cannot be applied. In this way, and taking into account the geographical area where these polymetallic nodules were identified, we assumed that the area involving the two metallogenetic areas, described above, forming a morphotectonic structure corresponding to a shallow basin, constitute an Area of Exploration Potential for this region (Map A5). The higher density of the nodules occurrences in the western cluster of this region, could only be the result of the higher number of samples collected, and not be directly linked to the higher probability to find such submarine deposits there – no information is available to confirm this. Even so, and based on the density of the occurrences, five smaller regions were individualized and considered as Area with High Potential of Discovery (Map A5).

Considering the potentiality of the area to supply critical and strategic elements, only P and V present interesting concentrations (higher than those in the four reference regions) (Annex C).

4.2. Metallogenetic Area 2 - White Sea

In this area only four occurrences are identified and none of them have any chemical data associated (Map A4). The samples were collected at shallow depths (42-226 mbsl), the host rocks are siltstones and sandstones and the rock ages are Tonian and Ediacaran.

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The white sea is formed by a shallow basin that connects to the Barents Sea. Due to the existence of these occurrences, together with the same shallow basin morphotectonic structure, it was decided to connect both areas and consider them as "Areas of Exploration Potential". To the area involving the four occurrences in the White Sea, was attributed the designation of "Area with High Potential of Discovery", where several nodules occurrences are found (Map A5)

4.3. Metallogenetic Area 3 - Baltic Sea

About 62% of all polymetallic nodules occurrences listed in the MINDeSEA database are from the Baltic Sea. From these, about half of the occurrences (99 in total) are from Gulf of Bothnia (Metallogenetic Area 3a), 57 occurrences are from Gulfs of Finland and Riga (Metallogenetic Area 3b); the rest, 27 occurrences, are from the Baltic Sea proper, including Kiel Bay (Metallogenetic Area 3c).

Concerning the genetic origin of the nodules from this region, distinct results are obtained:

a) When applying the ternary diagram of Bonatti et al., 1972 ((Co+Cu+Ni)*10 – Fe – Mn), the majority of the data points are distributed in the hydrothermal field, located at the base of the diagram. Some sample, inclusively, enter into the field of diagenetic origin. No distinction is observed among the three sub-areas.

b) When Ce_{Sn}/Ce_{Sn}^* is plotted against Nd (Bau et al., 2014), almost the entire population of the corresponding data points are plotted inside the diagenetic field. No distinction is observed among the three sub-areas.

c) When Ce_{Sn}/Ce_{Sn}^* is plotted versus Y_{Sn}/Ho_{Sn} , almost all data points are included in the hydrothermal field. No distinction is observed among the three sub-areas.

Hence, the are no differences in the genetic origins among the three sub-areas, all showing concise results; however, distinct origins are obtained depending on the discriminant diagrams employed.

In respect to the potentiality of these metallogenetic areas for some critical elements, and

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based on the graphs presented in Annex C, we can refer:

a) Concerning Co concentrations, no differences are observed among the three sub-areas, and all show very low concentrations (global variation between 0.001 – 0.024 wt%). These values are well below CIOB, CCZ and Cook islands EEZ (all ranging between 0.111 and 0.411 wt%; Peru basin shows much lower Co concentrations, but even so, above those observed for the Baltic Sea). No potentiality in Co for this region.

b) With the exception of the nodules from the Gulf of Finland, the highest REY total concentrations of the Baltic sea, only obtained for two samples (0.056 and 0.072 wt%), have higher values when compared to Peru basin (with average concentrations of 0.040 wt%). However, the rest of all REY concentrations are below 0.037 wt%. The three other reference regions have a REY variation between 0.081 and 0.168 wt%, well above those observed in the Baltic sea nodules.

c) Ti concentrations among the three sub-areas are similar, with a tendency for lower values in the nodules from Gulf of Finland. The entire variation in the Baltic sea is in the range observed in Peru Basin, CCZ and CIOB (altogether ranging from 0.16 to 0.42 wt%), but lower than the average value for Cook islands EEZ (1.2 wt%). Globally no potentiality of this metallogenetic region for Ti.

d) The number of available Li concentrations (8 occurrences with chemical data for this element) and the very low absolute values determined (4-10 ppm), makes this zone not to be considered as having any potential for this element. For comparison the global variation among the four reference regions are 51 to 311 ppm.

e) Considering the P concentrations, identical values are obtained for Bothnia Gulf and Baltic Sea proper; however one sample from this last sub-area has the highest P concentration (~10 wt%) of all nodules samples listed in the MINDeSEA database. The nodules from Gulf of Finland show a general increase in their P concentrations (1.09 – 4.93 wt%). Globally, the P concentrations are above those observed for the reference regions (0.15-0.34 wt%).

f) Nb concentrations are not available for the nodules from Bothnia Gulf. For the two

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other sub-areas of the Baltic Sea, the concentrations are identical and have very low values (<=12 ppm). The Nb concentrations for the reference regions are also low (< 100 ppm).

g) V concentrations for the Baltic sea metallogenetic area are lower than ~200 ppm. No significant differences mong the three sub-areas and all their concentrations are below the range defined for the reference regions (431 – 508 ppm).

h) Sc data are very scarce (only 14 occurrences have Sc chemical data associated). Only the nodules from Baltic Sea proper have Sc data, which concentrations vary 177 and 234 ppm (the highest Sc concentrations listed in the MINDeSEA database). All these concentrations are much higher than those observed for the reference regions (7.6 – 25 ppm). However the absolute values in Sc for the Baltic Sea are low, and no potentiality for this area is considered.

In summary, only P and REY elements have interesting concentrations.

Considering the strategic elements, the subsequent observations can be made:

a) The highest Mn concentrations are found in the Baltic Sea proper, which can reach values of 32.35 wt%. The maximum concentrations in the other two sub-areas are lower (Botnhia Gulf ~17 wt%; Finland Gulf ~25 wt%). In all three sub-areas, there is a serial variation in Mn concentrations up to the lowest concentrations. When compared to the reference regions, only Peru basin shows higher Mn concentrations than Baltic Sea.

b) Very low concentrations are observed for Cu in the Baltic Sea. Globally, the highest value (0.019 wt%) is well below the variation observed for the reference regions.

c) Similarly to Cu, Ni concentrations are very low in the Baltic Sea area (the highest concentration is found in Gulf of Bothnia – 0.074 wt%). All values well below those for the reference regions.

d) Only in the Bothnia Gulf sub-area high Zn concentrations are found – two nodules

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samples are completely out of the general variation for this element observed in the other metallogenetic areas or reference regions, showing concentrations of ~7000 and 10 000 ppm. These are the richest Zn concentrations found in the MINDeSEA nodules database. When considering the Zn values in the Baltic sea proper sub-area, it can be observed that they are in the range found among the reference regions.

e) The highest Mo concentration (~450 ppm) is found in the Gulf of Finland; this value is lower than the average concentrations observed in Peru basin, CCZ and CIOB (547-600 ppm for all three regions). Almost the entire variation observed in the Baltic sea area is between 1 and 300 ppm.

In summary, Baltic sea only shows a high potentiality for Mn and Zn. The areas surrounding the most dense occurrences sites were classified as “Areas with high potential of discovery”, whereas all the area of Baltic Sea was considered an “Area of Exploration potential”, basically because all the environmental parameters present in the areas where nodules occurrences were found are the same in all the Baltic Sea (Map A5).

Baltic Sea occupies a relatively small shallow water and confined area, whereas the strong inputs of river water, the topographic succession of sills and basins, and the narrow entrance to the North Sea are the main features that determine the hydrography of the Baltic Sea (Schneider & Müller, 2018). Relatively high nodule abundances between 10 and 40 kg/m^2 occur in the gulfs of Riga, Finland, and Bothnia in water depths between a few tens of meters and about 250 m, covering areas of a few hundred km^2. This abundance is related to the large input of Mn- and Fe-rich suspended matter through rivers in the northeast and east (Gulf of Bothnia, Gulf of Finland) and the formation of an oxidized layer in the upper 2–15 cm of the sediment column (Glasby et al. 1997). Fast-growing Fe-Mn concretions are mainly found in the western Baltic Sea, their formation being related to the development of summer anoxia and the diagenetic mobilization of Mn.

The chemical characteristics of the underlying sediment are likely also important to the formation and metal content of nodules (Zhamoida et al., 2017). These data are unfortunately less common. The most abundant Mn-rich spheroidal nodule fields are found with-

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in areas where Precambrian crystalline rocks (Baltic Shield) form the basement since these rocks are rich in Mn and Fe. Glacial till formed from fragments of these rocks has been proposed as also a source of Mn and Fe in these concretions. However, many observations of nodules are related to finer grained sediments clay/silt sediments, both older (glaciolacustrine) and newer (postglacial) deposited. Glacial clay is often covered by a residual of thin layers (2-30 cm) of mixed sediments (mainly silt, sand, gravel and cobbles). From observations, there are also some qualitative indications that larger nodule occurrences on the seabed are associated with older sediments and an environment that is neither to erosive and mobile or accumulating. This may result from the fact that this is an environment that have permitted the nodules to form and grow during longer time. In general, the abundance of nodules and the Mn contents increase within increasing water depth as a result of more active processes of Mn mobilization and migration in the anoxic environments encountered at the deepest basins of the eastern Gulf of Finland (Zhamoida et al., 2017). In the model run by Kaikkonen et al., 2019 depth was found to be the most influential predictor in the concretion models, followed by total phosphorus, occasional hypoxia, frequent hypoxia, and depth-attenuated wave exposure. Salinity, chlorophyll a, nitrogen, and ruggedness had smaller influences on the occurrence of nodules throughout the models.

4.4. Metallogenetic Area 4 - Black Sea

A total of 13 nodule occurrences are found in this region, but only 12 have chemical data associated. The nodules have high Fe concentrations (18.36 – 37.77 wt%) and low Mn concentrations (1.02 – 10.17 wt%). The Fe/Mn ratios have low values (0.05 – 0.49), which are indicative of a hydrogenetic origin. This genetic process is, in part, confirmed by the relationship among Co+Ni+Cu, Fe and Mn, in the ternary diagram from Bonatti et al. (1972), with some samples located in the hydrogenetic field defined there. However, other samples show an hydrothermal imput. The available REY data, when plotted in the Bau et al. (2014) diagram, are plotted in the diagenetic field. Concerning the concentrations of critic elements, the following observations can be made (see Annex C):

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a) Co concentrations (0.081-0.250 wt%; 4 occurrences) are only below those defined for Cook islands EEZ (showing a average value of 0.411 wt%), being in the range observed for the other three reference regions.

b) Very low REY concentrations (0.002-0.007 wt%; 7 occurrences), well below of the four reference regions.

c) Ti values are in the range of 0.01 – 0.10 wt% (data for 4 occurrences), slight lower than the concentrations observed in Peru basin, but clearly lower than the rest of the reference regions.

d) No Li data are available for these occurrences in the Black Sea.

e) Concerning P concentrations, they have values in the range of 1.75 – 2.57 wt%, which are significantly higher than those presented by the four reference regions (ranging between 0.15 and 0.34 wt%).

f) No Nb concentrations available for the nodules occurrences in the Black Sea.

g) Concentrations of V are between 160 and 300 ppm, and are lower than any of the reference regions (431 – 508 ppm).

h) No Sc data are available for this metallogenetic region.

With respect to the strategic elements, it is worth mention the following notes (see Annex C):

a) Mn concentrations are between 1.02 – 10.17 wt%, and are clearly lower than those observed in all four reference regions (16.1 – 21.4 wt%). Its range of variation is identical to the lowest ranges of variation observed in the other defined metallogenetic regions.

b) The Cu concentrations are low (0.037 – 0.092 wt%), and distinctly below those presented by the four reference regions (ranging from 0.226 to 1.071 wt%).

c) Similar to Cu, Ni concentrations (0.172 – 0.557 wt%) are below than those presented by three of reference regions (Peru basin, CCZ, CIOB, with 1.101 – 1.301

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wt%). The most Ni enriched nodules occurrences are only overcame by the nodules sampled in the Rockall Plateau, whereas the Ni content reaches ~0.9 wt% (Ni highest concentration listed in MINDeSEA database). However, the absolute Ni concentrations for this area are low and, thus, it results in a low potentiality for this element.

d) Very low Zn concentrations (108 – 256 ppm) and below the corresponding concentrations observed in the reference region having the lowest Zn contents (Cook islands EEZ, 545 ppm).

e) Mo concentrations are the lowest observed in the MINDeSEA database (5-14 ppm)

From the observations made above, it is worth to mention that for this metallogenetic area of Black Sea, only the critical element P has concentrations that stand out from the MINDeSEA listed chemical data and, additionally, are higher than those observed for the four reference marine regions. Considering the genetic origin for these polymetallic nodules, the results are dubious once different conclusions are obtained from the different relationships among the elements that were considered.

All of the polymetallic nodules occurrences are found in the north western continental shelf of the Black Sea, between the Crimean peninsula and the Danube Delta. The Black Sea shelf represents about 30% of the sea area (Ross et al., 197 4; Panin, Jipa, 1998), and it is mostly shallower than 100 m, but in the northwestern area the shelf break reaches 140 to 170 mbsl water depth (Popescu, 2002). Small areas involving the occurrences of polymetallic nodules in the north western continental shelf of the Black Sea were defined as “Areas with high potential of discovery”, whereas all the continental shelf, with the maintenance of the environmental parameters found in the areas above referred, is considered an ”Area of exploration Potential” (Map A5).

4.5. Metallogenetic Area 5 - Biscay Bay and Iberian Margin

This metallogenetic area includes the southern part of the Biscay Abyssal Plain and the eastern part of the Iberia Abyssal Plain. A total number of 8 occurrences are found in this

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area; only 5 occurrences are chemically characterized, and these were collected in the Galicia Bank. Two distinct types of chemistry are found: 3 occurrences have nodules enriched in Fe (~39 - 67 wt%) and with low Mn concentrations (~0.1 – 5 wt%), producing low Mn/Fe ratios (< 0.13). Conversely, in the other two occurrences, the nodules are very enriched in Mn (~39 – 43 wt%), have very low Fe contents (~0.3 – 1 wt%) and the corresponding Mn/Fe ratios have high values (~40 and 120). This chemical dichotomy is equally well expressed in the ternary diagram relating (Co+Ni+Co), Fe and Mn (Bonatti et al., 1972), with the first group of samples positioned in the hydrothermal field, whereas the second group of nodules are located in the diagenetic field. The relationship between Ce_{Sn}/Ce_{Sn}^* and Nd (diagram of Bau et al., 2014) show identical origins for both groups, but the correlation between Ce_{Sn}/Ce_{Sn}^* versus Y_{Sn}/Ho_{Sn} show an hydrogenetic origin for the samples enriched in Fe, and a diagenetic origin for those with the highest Mn contents.

The geographic, oceanographic and tectonic settings present in the Bay of Biscay and the Iberian Coast are quite distinct; in this region some (few) nodules occurrences are identified in the western Galicia continental margin. The metallogenetic information available have allowed to assume that their generation is controlled by local conditions. In the western Galicia continental margin, the polymetallic nodules occur together with extensive phosphorite pavements on seamounts and banks existent there (González et al., 2014, 2016). The formation of these nodules and phosphorites is at least partly related to the activity of deep-reaching faults and the geothermal mobilization of metals from a deeper crustal reservoir (González et al., 2014, 2016).

Taking into account the potentiality of this metallogenetic area for the critical and strategic elements, various observations can be emphasized (see Annex C):

a) Co concentrations (1.801 and 1.123 wt%) are very high in the samples having the highest Mn/Fe ratios, and are the two sampled occurrences with the highest Co concentration from the entire MINDeSEA database. These values are inclusively well above those shown by the four reference regions (with concentrations ranging from 0.111 to 0.411 wt%). Conversely, the other iron rich polymetallic nodules

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show the lowest concentrations (0.001 – 0.018 wt%) for this element, listed in the MINDesea database.

b) The range of REY concentrations (0.002 – 0.026 wt%) are all below those shown by the four reference regions (0.040 – 0.168), and are in the range observed for the other metallogenetic areas, defined in this MINDeSEA project; thus, the occurrences of this metallogenetic area have low potentiality for REY.

c) Very low and consistent Ti concentrations (all < 0.10 wt%); nodules from Peru basin are those that show the most similar Ti concentrations (average 0.16 wt%).

d) Li data are observed for 4 nodules occurrences, no systematic difference is observed between those having low and high Mn/Fe ratios and, altogether, present low concentrations (4 – 26 ppm) and smaller than those of the four reference regions (51 to 311 ppm). Low potentiality for this element.

e) Taking into consideration all the data for the metallogenetic areas here defined, P concentrations in Biscay Bay and Iberian Margin are those showing the lowest values of all (0.10 – 0.61 wt%). However the highest concentrations here observed are greater than those observed for all the reference regions. In this way, a low P potentiality is attributed to this area.

f) Nb concentrations are observed for 3 samples, with values of 5, 11 and 34 ppm. The last value is the highest Nb concentration in MINDeSEA database, but the entire number of Nb analyses is very restricted (n=12); for comparison, this Nb concentration is higher than those observed in Peru basin and CCZ (13 and 22 ppm, respectively), but lower than those shown by Cook islands EEZ and CIOB (90 and 98 ppm, respectively).

g) Two of the occurrences in this region (those having the highest Mn/Fe ratios) present the highest V concentrations of the entire list of registered polymetallic nodules occurrences, defined in MINDeSEA database (1740 and 2988 ppm). However, the lowest V value of the MINDeSEA database is also observed in this region (14 ppm), which makes it the metallogenetic zone with the greatest dispersion in

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the concentrations of this critical element. For comparison, the average concentrations observed in the four reference marine regions ranges from 431 to 508 ppm.

h) Sc concentrations are low, between 2 and 13 ppm, and are in the range observed for Cook islands EEZ, CCZ ans Peru basin (7.6 – 12 ppm). No potentiality for Sc in this area.

i) This metallogenetic area presents the two highest Mn concentrations of all the nodules occurrences listed in MINDeSEA database: 38,65 and 43,10 wt%. Additionally, both values are higher than those observed in Peru basin (average of 34.2 wt%), which is the reference region with the highest Mn contents. The other three samples from Galicia Bank, those enriched in Fe, have very low Mn concentrations (0.09 – 5.31 wt%).

j) Like Co and V, Cu concentrations are higher in polymetallic nodules having the highest Mn/Fe: 0.155 and 0.180 wt%. In relation to the other metallogenetic regions, here defined, the Cu concentrations are only exceeded by those observed in Rockall Plateau, Madeira Abyssal Plain, Kane FZ and in the Lochs Goil and Striven (which as a whole range from 0.190 to 0.247 wt%). However, this range of Cu variation for Galicia Bank is lower than any observed in the four reference regions (showing a compositional interval between 0.226 and 1.701 wt%).

k) Ni concentrations ranges from 0.001 to 0.233 wt% and are lower than the Ni average contents observed in any of the four reference regions (Ni concentrations interval between 0.38 to 1.301 wt%). The most Ni enriched samples from this Bank are those having the highest Mn/Fe ratios.

l) Very low Zn concentrations (9 – 205 ppm) which, as a whole, constitute the lowest Zn contents of the MINDeSEA listed occurrences.

m) Mo is enriched (220 and 291 ppm) in the nodules having higher Mn/Fe, comparatively to the Fe enriched samples (37 and 54 ppm). All Mo values from Galicia Bank are below those observed in any of the four reference regions (with Mo contents between 295 and 600 ppm). Low potential for this element.

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The only chemical data obtained for polymetallic nodules sampled in this metallogenetic area, comes from Galicia Bank. The most interesting metal concentrations – Co, Mn, V, Cu and Ni – are obtained in the samples having the highest Mn/Fe ratios. Special emphasis should be given to the high potentiality of this area for Co, V and Mn - these three elements have in this area the highest concentrations of all the occurrences listed in MINDeSEA database. Two small areas (one corresponding to the Galicia Bank and the other in the Biscay Abyssal plain) are considered "Areas of high potential of discovery", but the area involving both is considered an "Area of Exploration Potential" (Map A5).

4.6. Other areas with potentiality for critical and strategic elements

Beyond the regions that were considered Metallogenetic Areas, based on the higher density of polymetallic nodules occurrences, there are other areas that have also have nodules occurrences that can be evaluated for their potentiality in some of the critical and strategic elements. Here, it is identified those areas for the elements above discussed (see Annexes B and C)

Co

The distribution of Co concentrations (see Annex B, Polymetalic Nodules Co Map), firstly shows, for the lowest values, a continuous variation from 0.001 to 0.121 wt% (class 1). The defined Class 2, also shows a gradual increase in Co concentrations from 0.250 to 0.430 wt%. Some of these concentrations are found in areas not included in the metallogenetic areas above referred, such as Kane FZ, Rockall Plateau and Madeira abyssal plain. The class with high concentrations, beyond the two samples from Galicia Bank, includes one sample from Atlantis FZ (0.790 wt% of Co).

REY^T

There is a continuous variation from the smallest concentration listed in the MINDeSEA database (0.002 wt%) until 0.037 wt% of REY. Based on the median value, two classes were established (I and 2). The third class is the one showing the highest REE values, and should be mentioned the highest value listed in MINDeSEA for Madeira Abyssal

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Plain (0.235 wt%) – a value well above all those presented by the four reference regions. Also significant the REY concentrations found in the Rockall Plateau (0.079 wt%).

Ti

The highest concentrations of Ti are found in the regions not included in the Metallogenetic areas above referred. Loch Fyne occurrence shows the highest concentration of all (4.7 wt%), but high values are also found in Rockall Plateau and Canary basin (~1 to 1.5 wt%).

Li

It is only worth to mention the nodule occurrence in Madeira Abyssal Plain, having Li concentrations of ~72 ppm.

P

The highest P concentrations are all found in the metallogenetic areas described above. No interesting concentrations are found in other regions.

Nb

No Nb data for polymetallic nodules are available outside the the metallogenetic areas already described.

V

Two occurrences have nodules with V concentrations higher than 500 ppm: Madeira Abyssal Plain and Loch Fyne.

Sc

No important Sc concentrations are found outside the metallogenetic areas defined.

Mn

The only significant Mn concentrations important to refer outside the metallogenetic areas are are those found in the Lochs Fyne, Goyle and Striven (~16 – 19 wt%)

Cu

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Important Cu concentrations are found in the occurrences of Rockall Plateau, Madeira Abyssal Plain, Kane FZ and Loch Goil and Loch Striven (~0.200 to 0.250 wt%)

Ni

Important Cu concentrations are found in the occurrences of Rockall Plateau, Madeira Abyssal Plain and Kane FZ, with special emphasis for the highest Ni concentration listed in the MINDeSEA database for Rockall Plateau (0.880 wt%)

Zn

The only occurrence outside the metallogenetic areas worth to be mentioned is the one located in the Madeira Abyssal Plain (610 ppm).

Mo

Occurrences found in Kane FZ and Madeira Abyssal Plain (240 – 380 ppm) are the most important found outside the metallogenetic areas.

Taking into account the observations above referred, two areas present high potentiality for a number of metals: Madeira Abyssal Plain and Rockall Plateau. These areas were considered as “Areas with high potential od discover” (Map A5).

5. Polymetallic nodules prospect evaluation parameters and predictive areas

Nodule formation is favoured by a range of environmental factors, which are also used as a basis for defining prospective areas for seafloor exploration. The associated permissive areas do not necessarily contain economically minable nodule deposits but simply represent regions where their formation is feasible.

The most important polymetallic nodules ‘fields are located in the abyssal plains of the deep ocean, in about 4000–6000 m water depth in all major oceans, meaning that depth is an important favouring factor for their generation. Besides the probable control of water depth in the formation of the nodules, this parameter could also control the concentrations of their constituent metals.

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Another factor that could control the formation of the nodules is their geographic location. This factor is related with biological primary productivity (production of organic matter by phytoplankton that will supply organic carbon to the diverse heterotrophs organisms, particularly the zooplankton) in surface waters, which in turn controls the calcite compensation depth (CCD -the depth at which calcite dissolves quicker than it can accumulate) (Hein & Koschinsky, 2014). Nodules are associated with low surface primary productivity, typically found in the open ocean. The highest-grade nodules form near, but generally below, the CCD (ISA, 2010).

Another very important factor that strongly influence the formation of polymetallic nodules is the very low long-term sedimentation rates of < 0.5-1.0 cm per 1,000 years, typically found on abyssal plains removed from regions of high surface productivity and terrigenous input, that is in areas distant from continents (e.g. Hein & Koschinsky, 2014). Such slow rates are associated with deep-sea pelagic clays, the bulk of which are allogenic (Fütterer, 2006; Glasby, 2006).

Bottom-water oxygen concentration is also an important factor controlling the nodules generation. Nodules are preferentially associated with moderately high oxygen values (between 150 and 210 mmol/m3), which primarily occur in the northern and central Pacific and Indian Oceans, in the southern Pacific Ocean, and in the South Atlantic Ocean. Oxidizing conditions are critical for the formation of metallic oxides that comprise polymetallic nodules (e.g. Glasby, 1997). General absence of nodules in regions of very high and low dissolved oxygen concentration is linked to high rates of sedimentation in these areas, which are proximal to continental margins (Dutkiewicz et al, 2019).

Another important variable is seafloor lithology. The nodules show a strong preference for clay, followed by calcareous ooze in the vicinity of clay regions, with the remaining sediment types being relatively unimportant globally for nodule formation (Dutkiewicz et al, 2019).

Seafloor megafaunal biomass constitutes another factor controlling the generation of the

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polymetallic nodules, which typically occur in seafloor regions of moderate to low biomass concentration and low total organic carbon content (Dutkiewicz et al, 2019). Both of these variables are closely linked to surface productivity. Low surface productivity results in low flux of particulate organic carbon to the seafloor, which maintains benthic biomass adapted to low energy availability (Ramirez-Llodra et al., 2010).

Bottom currents can remove fine sediments that could cover the polymetallic nodules and, at the same time, oxygenate the abyssal plain where they grow (Lusty & Murton, 2018). Nodules are preferentially associated with very low bottom current speeds of <5 cm/s, which pervade all major nodule fields including the CCZ. These speeds are considerably lower than speeds of >10–15 cm/s that are required to erode fine silt and clay in the deep sea (Dutkiewicz et al, 2019).

Also crucial to the nodules growing is the availability of a source material for nucleation, which can include minute fragments of rock debris, indurated sediment, micro-nodule fragments or even biogenetic micro-parts. Furthermore, numerous seamounts and fault zones can provide a high amount of nucleus material to initiate nodule growth (Hein & Koschinsky, 2014).

Small- scale ocean floor topography could also determine whether nodule abundances reach values that are high enough to become economically interesting (Kuhn et al., 2017), and the presence of semi-liquid sediments that enhance the amount of pore water and diagenetic input to nodule growth can also be considered an environmental factor controlling the polymetallic nodules generation on the sea bottom.

The environmental parameters above identified, from published studies, are considered to promote the formation of polymetallic nodules, and could be used as a basis for defining prospective areas for seafloor exploration. However, all these parameters were based on studies made in the most important and well-known polymetallic nodule fields that are located in the Pacific and Indian oceans. A comparison of the geographic, submarine and geotectonic settings between those that occur in well-known Pacific and Indian ocean

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nodules fields and those that characterize the regions where the nodules occurrences were identified in this MINDeSEA project, allows us to identify two regions having “similar” settings – the abyssal plains located inside Macaronesia and those, contiguous but outside this region, found in International Waters (Canary islands and Madeira Abyssal Plains).

However, once depth is a very important favouring factor for the generation of polymetallic nodules, whereas the most significant polymetallic nodules ‘fields are located in the abyssal plains of the deep ocean, the predictivity areas for the existence of polymetallic nodules in European waters also include the major abyssal plains found there.

The limited number of nodules occurrences in Macaronesia and in International Waters and the insufficient metallogenetic data available only allow us to assume that prospect evaluation parameters would be identical to those defined for the nodules fields in the Pacific and Indian oceans. Additionally, through the identification of polymetallic nodules covering vast areas of the summit of the Tropic Seamount (about 30 samples of polymetallic nodules were collected during the James Cook cruise in 2016 (Murton et al., 2017), but no mineralogical or geochemical data are yet available) it is reasonable to assume that they growth under the same main hydrogenetic origin with some influence of diagenetic and/or hydrothermal processes during their formation (Marino, 2020).

6. Conclusions

a) The MINDeSEA database has listed a total of 296 polymetallic nodules occurrences.

b) About 85% of all listed occurrences (n= 252) are located in only two regions: 1) the majority, 184 occurrences (62% of the total number) are in the Baltic Sea; 2) 68 occurrences (23% of the total number) are in the Barents Sea.

c) From all the listed occurrences, only 107 (36% of the total number) have chemical

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data associated. A total number of 493 analysed polymetallic nodules are listed in the MINDeSEA data base.

d) Thirty occurrences have more than one polymetallic nodule analysed, and the chemical data listed in the MINeSEA database, for these occurrences, are the resulting average values.

e) Based on the spatial density of nodules occurrences, 5 major metallogenetic areas were defined:

- Metallogenetic Area 1 - Barents Sea, defining two separate areas next to each other: Metallogenetic Areas 1a and 1b)

- Metallogenetic Area 2 - White Sea

- Metallogenetic Area 3 – Baltic Sea, divided into three sub-areas:

  - Metallogenetic Area 3a - Gulf of Bothnia

  - Metallogenetic Area 3b - Gulfs of Finland and Riga

  - Metallogenetic Area 3c - Baltic Sea proper (including Kiel Bay)

- Metallogenetic Area 4 - Black Sea

- Metallogenetic Area 5 - Biscay Bay and Iberian Margin

f) The “Areas of Exploration Potential” where defined based on the spatial position of the metallogenetic areas, including other areas having similar and favourable morphostructures (MAP A4).

g) The “Areas with High Potential of Discovery” are related to the highest density of occurrences sites, having the same associated morphostructure (Map A4).

h) Taking into account that most important fields of polymetallic nodules are located in the abyssal plains of the deep ocean, in about 4000–6000 mbsl, depth is generally considered an important favouring factor for the generation of the nodules. Hence, all the major Abyssal plains present in the European Seas were considered as “Predictivity Areas”. Additionally, other marine areas with morphotectonic struc-

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Critical and Strategic elements	European marine areas
P, V	Metalogenetic Area 1 (Barents Sea)
P, REY	Metalogenetic area 3 (3a, 3b, 3c) (Baltic Sea)
P	Metallogenetic Area 4 (Black Sea)
Co, Mn, V, Cu, Ni	Metallogenetic Area 4 (Biscay Bay and Iberian Margin)
Co, Ti, Cu, REY, Ni	Rockall Plateau
Co, REY, V, Cu, Ni	Madeira Abyssal Plain

Table 2. Critical and strategic elements having significant potential as metal resources associated to

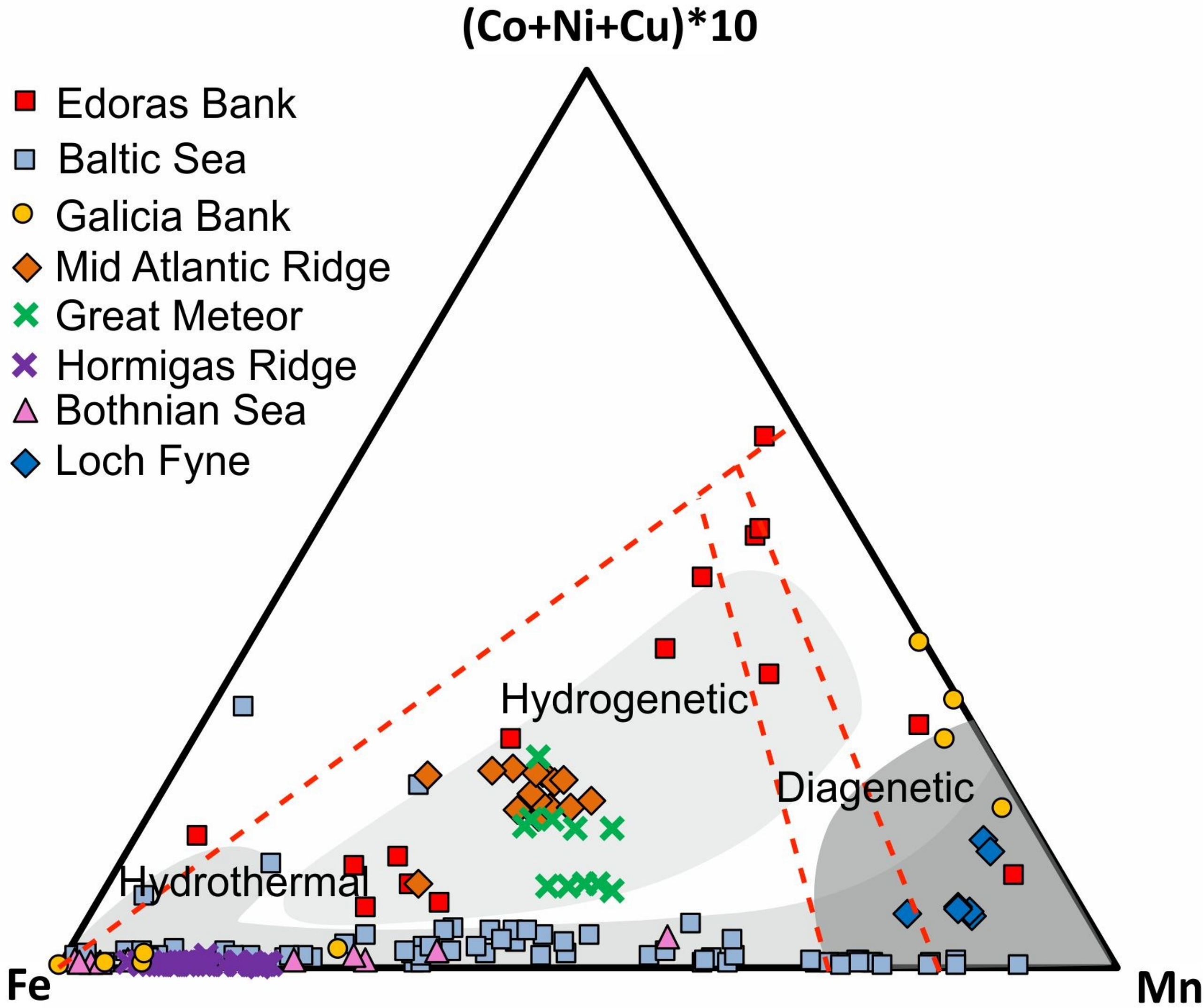

Figure 6. Relationship among Co+Ni+Cu, Fe and Mn, for polymetallic nodules registered in MINDeSEA database (ternary diagram from Bonatti *et al.,* 1972).

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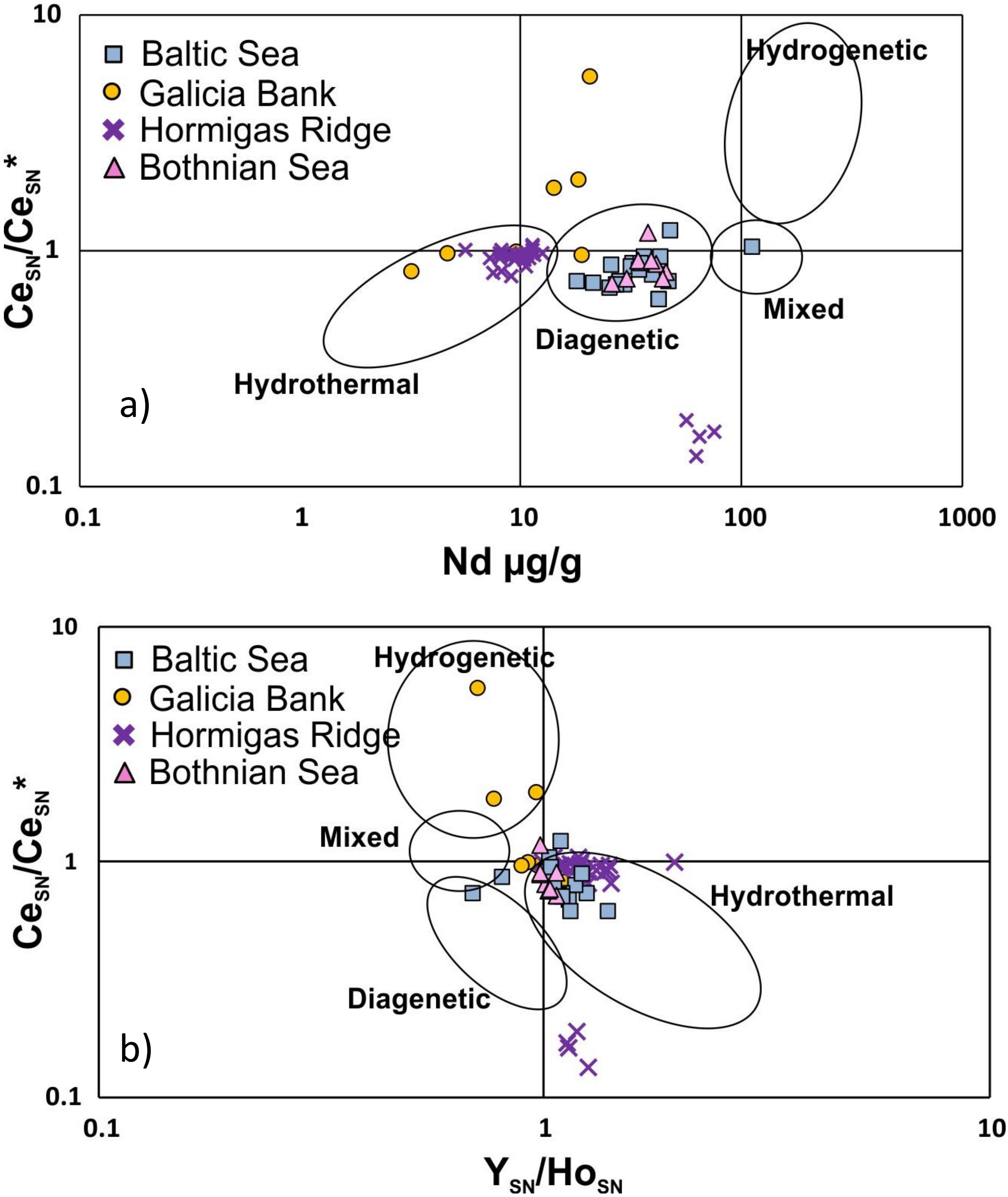

Figure 7. Relationship among some of the rare earth elements (Ce, Nd and Ho) and Y, for polymetallic nodules registered in MINDeSEA database. a) Ce_{SN} /Ce^*_{SN} *vs* Nd and b) Ce_{SN}/Ce^*_{SN} *vs* Y_{SN}/Ho_{SN} (Bau *et al.,* 2014). Subscript SN stands for Post-Archean shale normalized concentrations.

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