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Exhalation of Rn-222 from polymetallic nodules – assessment of radiological risk during transportation and storage

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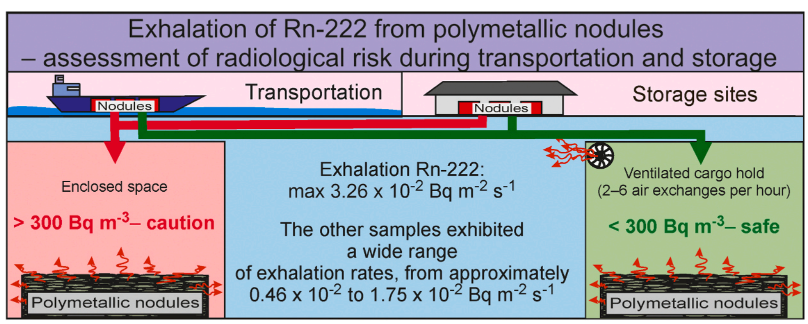
HIGHLIGHTS

- Rn-222 release from polymetallic nodules was quantified experimentally.
- Surface exhalation varied strongly between nodule samples.
- Sample geometry controlled the apparent Rn-222 release to air.
- Bulk-layer thickness limited effective Rn-222 transfer to the chamber.
- Ventilation reduced estimated indoor radon in a cargo-hold scenario.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to assess the potential radiological hazards associated with the presence and exhalation of Rn-222 from polymetallic nodules collected from the Pacific Ocean seabed within the Clarion–Clipperton Zone, with a focus on radon-related inhalation hazard under defined transport/storage ventilation scenarios. The research focused on measuring the surface exhalation rate of Rn-222 from both whole nodule and pulverized samples, using an AlphaGUARD monitor operating in flow mode. The highest recorded exhalation rate was 3.26×10^{-2} Bq m^{-2} s^{-1}, while the values for the remaining samples ranged from approximately 0.46×10^{-2} to 1.75×10^{-2} Bq m^{-2} s^{-1}. Based on the experimentally determined exhalation rates, a scenario-based analytical estimation of indoor Rn-222 activity concentration was carried out for an enclosed ship cargo hold with typical geometric dimensions. Under the assumed ventilation and effective exhalation-area conditions, predicted indoor Rn-222 activity concentration remains below the reference level of 300 Bq m^{-3} , as set out in the Council Directive 2013/59/EURATOM. The study also emphasised the importance of storage conditions, particularly the influence of moisture content in nodules on Rn-222 exhalation, as well as the need to implement basic radiological protection measures such as controlled ventilation and dust suppression. The presented results indicate that, with appropriate management and the application of simple technical measures, transportation and storage

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of polymetallic nodules do not pose a significant radiological hazard, in terms of the analysed Rn-222 inhalation scenario, and indoor Rn-222 activity concentration in the air can be controlled effectively.

1. Introduction

Polymetallic nodules (also referred to as ferromanganese or manganese nodules) were first recovered from the seafloor of the World Ocean in the second half of the 19th century. Belkin et al. (2021*)* [1] claim that the first nodules were collected in 1873 during a scientific expedition (1872–1876) aboard the British vessel *Challenger*. Current interest in polymetallic nodules stems from their relatively high content of valuable elements, including nickel, copper, cobalt, lithium, molybdenum, zirconium, and rare earth elements [2], which makes them a prospective source of raw materials for metal extraction [3]. Nodule deposits have been found in many areas of the World Ocean, including in particular the Clarion–Clipperton Zone in the north-eastern Pacific, where the total mass of nodules is estimated at 21 billion tonnes, the Peru Basin in the south-eastern Pacific, the Cook Islands region in the south-western Pacific, and certain parts of the Indian Ocean. Nodules have also been reported in other regions, such as the South China Sea [4] and the Baltic Sea [5].

Activities relating to seafloor exploration, assessment of environmental risks associated with nodule mining, and the development of mining technologies are conducted under national and international consortia and overseen by the International Seabed Authority (ISA). ISA is an autonomous international organisation established under the 1982 United Nations Convention on the Law of the Sea (UNCLOS) and the 1994 Agreement relating to the Implementation of Part XI of the Convention.

There are numerous publications on the formation of nodules on the ocean floor [6], their structure and chemical composition [7], proposed mining methods [8,9], ore processing technologies [10], including bioleaching [11], as well as the environmental consequences of nodules extraction and processing [12]. In the context of seafloor ecosystem protection, extensive research is underway [13], including large-scale studies conducted as part of international research projects such as Deep-Ocean Mining Environmental Studies and the Benthic Impact Experiment [2].

A lot of attention has recently been given to the potential health hazards from ionising radiation to personnel involved in the extraction, transportation, and processing of polymetallic nodules. This is due to the fact that nodules contain radioactive elements, including U-238, Th-230, Ra-226, Rn-222, Pa-231, Pb-210, and Th-232 [14–18]. Radium-226, having a half-life of $T_{1/2}$= 1600 years [19], together with its decay products, are classified among the most hazardous *Naturally Occurring Radioactive Materials* (NORM) to human health. As a result of Ra-226 decay, radioactive Rn-222 ($T_{1/2}$=3.8 days) is formed. Rn-222 emanates within the porous structure of nodules (or other radium-bearing materials), and only its fraction is released into the surrounding environment [20]. Under normal conditions, Rn-222 is a gas that can penetrate into the human respiratory system, where its radioactive decay products, including polonium-218 (Po-218), may deposit in the lungs [21]. The European Union has established a reference level of 300 Bq m^{-3} for annual average indoor radon concentration [22]. This is important when interpreting short-term transport or storage scenarios, because transient increases in radon concentration do not directly correspond to the annual-average exposure metric, especially when adequate ventilation is provided.

The aim of this study was to assess the likelihood of radiological hazards arising from exposure to Rn-222 during technological processes, particularly those associated with the transportation and storage of polymetallic nodules. It was hypothesised that accumulation of excessive masses of nodules in confined spaces, as may occur during transport and storage, could pose a health risk to humans. Furthermore, it was proposed that implementation of simple operational regimes such as limiting the exposure time, controlling the ratio of nodule mass to air volume in enclosed spaces, ensuring adequate ventilation, and employing protective barriers could effectively reduce this hazard. Unlike previous studies focused mainly on radionuclide content or chamber build-up observations, the present work combines standardized Rn-222 exhalation measurements with a practical transport/storage scenario analysis, enabling an application-oriented assessment of indoor radon risk in cargo-hold conditions based on experimentally determined exhalation parameters. The study does not aim to provide a complete radiological risk assessment covering all exposure pathways or all radionuclides, but rather a focused evaluation of a radon inhalation scenario under defined ventilation assumptions. This approach was intentionally adopted as a first-step engineering and radiological screening method for transport and storage conditions, using measured Rn-222 exhalation parameters as model input.

While the present study is based on a limited set of individual polymetallic nodules collected during research cruises, the dataset provides a well-defined basis for quantifying Rn-222 exhalation. However, more extensive sampling in the Clarion–Clipperton zone will be necessary to assess spatial variability and obtain representative values for the entire zone.

2. Materials and methods

2.1. Samples characteristics

The study utilised polymetallic nodules obtained from the repository of the Interoceanmetal Joint Organization, headquartered in Szczecin (Poland). The nodules were collected from the Pacific Ocean floor within the Clarion-Clipperton Zone, approximately 3000 kilometres west of the coast of Mexico. The samples were selected at random, with each sample originating from a different site located within the IOM claim area. The study involved two sets of samples. The first set consisted of five nodule samples (N = 5), each of them comprising several lumps of nodule material. The second set consisted of pulverized nodule samples (N = 15) prepared by separating a representative portion of each nodule either by breaking by hand or cutting with a diamond saw. Special care was exercised to ensure that the material included all structural zones of each individual nodule, spanning from the central core to the outer surface laminations. Depending on the size of each nodule, approximately one-quarter to one-half of its total volume was collected for analysis. The material was first air-dried at room temperature. Once dried, the individual samples were pre-ground in a ceramic mortar and subsequently milled into fine powder using a mechanical agate mill (model: Fritsch Pulverisette, 02.102). No sieve-based size classification was applied at this stage.

Details and characteristics of the analysed samples are summarised in Table 1. The dimensions (height and diameter) of the non-pulverised nodules were measured in accordance with the methodology proposed by Volz et al. [15]. The mass of these samples ranged from 0.037 to 0.063 kg. To calculate the specific surface area of the pulverized nodules, it was necessary to determine the particle size distribution. Measurements were performed using a PSA 1090 L particle size analyser equipped with a dispersion unit. The pulverized material consisted of particle sizes ranging from 0.04 µm to 500 µm, while the mean particle diameters of the analysed samples were between 12 µm and 23 µm. The pulverized sample masses varied from 0.008 kg to 0.033 kg. Before exhalation measurements, all samples were dried under laboratory conditions to ensure repeatability and comparability of results, because moisture can significantly affect radon emanation and exhalation.

Table 1
Nodule samples characteristics, mass and surface area.

Sample code	Description	Mass [kg]	Surface area [m^2]
1	pulverized sample	0.0158	2.43
2/1	pulverized sample	0.0199	2.39
2/2	4 intact nodules of the following dimensions: 0.034 × 0.04 m; 0.025 × 0.031 m; 0.027 × 0.028 m; 0.012 × 0.014 m	0.0514	0.01
3	pulverized sample	0.0143	2.14
4	pulverized sample	0.0212	2.55
5	pulverized sample	0.0089	1.22
6	pulverized sample	0.03097	5.8
7	pulverized sample	0.0226	3.93
8/1	pulverized sample	0.0218	4.03
8/2	pulverized sample	0.01105	2.12
9/1	pulverized sample	0.02975	4.51
9/2	6 intact nodules of the following dimensions: 0.03 × 0.04 m; 0.025 × 0.028 m; 0.015 × 0.025 m; 0.014 × 0.015 m; 0.005 × 0.01 m; 0.006 × 0.007 m	0.03738	0.014
10	pulverized sample	0.04272	6.99
11/1	pulverized sample	0.0405	6.74
11/2	pulverized sample	0.0312	5.2
11/3	6 intact nodules of the following dimensions: 0.031 × 0.038 m; 0.028 × 0.03 m; 0.019 × 0.038 m; 0.021 × 0.022 m; 0.019 × 0.025 m; 0.018 × 0.02 m	0.06293	0.013
12/1	pulverized sample	0.01954	3.52
12/2	pulverized sample	0.01246	1.94
12/3	5 nodules of the following dimensions: 0.02 × 0.024 m; 0.018 × 0.03 m; 0.024 × 0.025 m; 0.021 × 0.025 m; 0.017 × 0.024 m	0.03645	0.009
13/1	pulverized sample	0.01877	2.78
13/2	4 intact nodules of the following dimensions: 0.018 × 0.03 m; 0.022 × 0.025 m; 0.02 × 0.025 m; 0.018 × 0.02 m	0.03581	0.007
14	pulverized sample	0.026	3.31
15	pulverized sample	0.03314	3.59

2.2. Rn-222 exhalation measurement (EN ISO 11665–7-based accumulation method [23])

For the measurement of Rn-222 exhalation, an AlphaGUARD DF2000 monitor, commonly used in environmental studies, was employed [24,25]. The detector operates using an ionisation chamber, which enables direct measurement of gas ionisation effects caused by alpha particles emitted during Rn-222 decay and its long-lived progeny. Additionally, the AlphaGUARD DF2000 is equipped with integrated sensors for recording environmental parameters such as temperature, atmospheric pressure, and relative humidity. The active volume of the ionisation chamber is 0.56 dm^3 . The AlphaGUARD DF2000 monitor was within its valid calibration period during the measurements. Before each measurement series, the system was purged and instrument operation was verified under laboratory background conditions.

Rn-222 exhalation, understood as the release of this gas from the investigated material into the atmosphere, was assessed using a flow-through measurement setup. This operating mode requires the use of an exhalation chamber to confine the volume of the analysed air, and a pump to transfer the air to the ionisation chamber. The measuring device operated in a 10-minute flow-through mode, employing a specially designed exhalation chamber with a volume of 1.2 dm^3 . Before each measurement, the detector loop was purged to approach laboratory background conditions. A schematic diagram of the setup is presented in Fig. 1.

The exhalation measurement followed the accumulation approach described in EN ISO 11665–7 [23] (surface exhalation from materials),

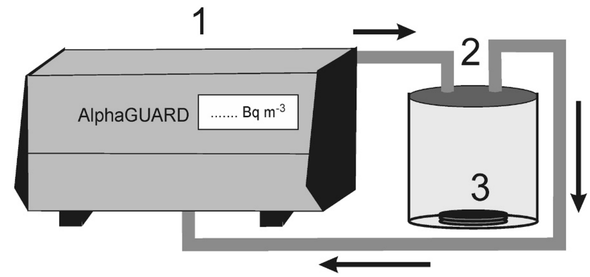

Fig. 1. Apparatus setup for the measurement of changes in Rn-222 activity: 1 –AlphaGUARD™ DF2000 detector, 2 – sample chamber, 3 – sample.

using the initial build-up of radon activity concentration in the closed measurement system to determine the exhalation parameter. In the early accumulation interval (short compared with the half-life of Rn-222), the increase in radon activity concentration in the chamber can be approximated as linear. The rate of increase was determined from the slope of the concentration-time relationship:

$$v_{\mathrm{Ex}} = \frac{\Delta C(t)}{\Delta t} = a \tag{1}$$

where: v_{Ex} – rate of increase in indoor Rn-222 activity concentration (Bq m^{-3} s^{-1}); *C(t)* – activity concentration in the exhalation chamber at time *t* (Bq m^{-3}); *t* – elapsed time from the start of exhalation (s); *a* – the slope obtained from linear regression (Bq m^{-3} s^{-1}).

In this study, the interval used for slope determination was standardized to the first 3 h of accumulation, consistent with practical application of EN ISO 11665–7 [23] for estimating the initial build-up rate. This standardized early-time window was selected to provide a robust and comparable estimate of the initial source term, while minimizing the influence of longer-time system- and material-dependent processes (leakage, back-diffusion, and retention effects) that become increasingly relevant as accumulation proceeds. Linear regression was performed as $C(t)=at+b$ without forcing the intercept to zero. This is important because the laboratory background radon concentration was non-zero (typically 20–40 Bq m^{-3}, mean ~27 Bq m^{-3}). Due to the 10 min recording interval, the first measured point was obtained after sealing and not exactly at $t = 0$. Therefore, the fitted intercept *b* is a regression parameter and should not be interpreted directly as the ambient background concentration at the moment of sealing. Although the exponential accumulation model provides the more general physical description of build-up in a closed system, applying full-curve source--term fitting would require explicit treatment of non-decay losses. In this study, such effects are addressed through an effective loss constant (λ_{eff}) when interpreting long-duration curves, whereas the exhalation coefficient itself is determined from the standardized early-time interval. The surface exhalation rate *E* was calculated from the slope *a*, the effective gas volume of the measurement system V_{eff}, and the effective surface area *S* [23]:

$$E = a\frac{V_{eff}}{S} \tag{2}$$

where: *E* – surface exhalation rate (Bq m^{-2} s^{-1}); V_{eff} – effective gas volume (m^3); *S* – effective surface area of the sample (m^2).

For comparison between samples of different mass and geometry, the mass exhalation rate of Rn-222 (E_M) was also calculated from the surface exhalation rate:

$$E_M = E\frac{S}{m} \tag{3}$$

where: E_M – mass exhalation rate (Bq kg^{-1} s^{-1}); m – the sample mass (kg)

The effective accumulation volume V_{eff} was defined as the free gas volume of the exhalation chamber plus the connected AlphaGUARD loop (including the ionization chamber and tubing), corrected for the sample volume. In the present setup, the effective system volume was approximately 1.8 dm^3. The largest sample volume was approximately 0.063 dm^3, corresponding to ~3.5% of V_{eff}, i.e., below 10%, which satisfies the usual accumulation-method requirement used in EN ISO 11665–7-based practice [23].

In addition to the ISO-based early-time evaluation of the surface exhalation coefficient, we estimated the radon emanation factor (dimensionless) for selected intact nodule samples using long-duration measurements in the same closed-loop system. For a closed system, the radon activity concentration in the gas phase approaches a steady state C_{eq} when production balances losses. Following the closed-chamber approach recommended for NORM materials [20,26], the emanation factor ε was calculated as:

$$\varepsilon = \frac{V_{eff} C_{eq}}{m C_{Ra-226}} \tag{4}$$

where V_{eff} – the effective gas volume of the measurement system (m^3), C_{eq} – the steady-state Rn-222 activity concentration in the closed loop (Bq m^{-3}), m – the sample mass (kg), and C_{Ra} – the Ra-226 activity concentration in the sample (Bq kg^{-1}).

Because reaching full equilibrium may require several days in an ideal leak-free system, C_{eq} was obtained from nonlinear fitting of the long-duration build-up curves using an exponential approach-to-equilibrium model:

$$C(t) = C_{eq} - \left(C_{eq} - C_0\right) e^{-\lambda_{eff} t} \tag{5}$$

where C_0 is the activity concentration at sealing (Bq m^{-3}) and λ_{eff} (s^{-1}) is an effective build-up constant representing the combined effect of radioactive decay and non-decay losses processes in the closed loop (e.g., leakage and back-diffusion effects treated jointly).

The raw AlphaGUARD time-series exports used for slope estimation and uncertainty analysis are deposited in the University of Opole Research Data Repository (version 1.0) [27]

Measurements were performed for both the unprocessed polymetallic nodule samples and the pulverised nodule material. In the case of the unprocessed nodules, the effective surface area was calculated according to the formula applied by Volz et al. [15]:

$$S = 0.5\pi d^2 + 0.25\pi \frac{h^2}{e} \ln\left(\frac{1+e}{1-e}\right) \tag{6}$$

where: d – diameter, (m); h – height, (m); e – eccentricity, determined as in Eq. (7), (-);

$$e = \sqrt{1 - \frac{h^2}{d^2}} \tag{7}$$

In the case of the pulverised samples, knowing the particle size distribution and the mass fraction of each size class, the specific surface area S_w was calculated using the following equation:

$$S_w = \frac{6}{\rho_S} \sum \frac{f_i}{d_i} \tag{8}$$

where: S_w – specific surface area (m^2 kg^{-1}); ρ_S – density (kg m^{-3}); d – diameter of the i-th particle size fraction (m); f_i – mass fraction of the i-th particle size class (-).

Next, in order to obtain the total surface area of the pulverised material, the specific surface area was multiplied by the mass of the analysed sample:

$$S = S_w m \tag{9}$$

where: S – effective surface area of the powdered material (m^2); m – mass of the analysed sample (kg).

In the standardized 3 h interval used to determine the slope, radioactive decay and additional loss and retention processes (e.g., leakage, back-diffusion) were treated as negligible for practical estimation of the initial growth rate, in accordance with the early-time approximation. This does not imply that these processes are physically absent; their influence becomes more relevant for longer accumulation times.

2.3. Indoor Rn-222 activity concentration

To interpret long-term build-up behaviour in the closed system, the increase in indoor Rn-222 activity concentration can be described by an exponential accumulation model [23]:

$$C(t) = C_0 + \frac{ES}{V\lambda}\left(1 - e^{-\lambda_{eff} t}\right) \tag{10}$$

$$\lambda_{eff} = \lambda_{Rn} + \lambda_B + \lambda_V \tag{11}$$

where: C_0 – is the initial radon activity concentration in the system at sealing (Bq m^{-3}); λ_{Rn} – decay constant of Rn-222 (s^{-1}); λ_B – back-diffusion rate constant (s^{-1}); λ_V – leakage rate constant of the system (s^{-1}); t – time elapsed from the start of the exhalation measurement (s).

In the present study, this exponential form was used primarily for interpretation of long-duration build-up curves, while the exhalation coefficient itself was determined from the standardized 3 h linear interval described above, according to EN ISO 11665–7 [23].

For practical risk assessment, measured exhalation coefficients were then used to estimate indoor Rn-222 activity concentration in air (e.g., hypothetical cargo-hold scenario) using a steady-state room model that includes air exchange:

$$C = \frac{ES}{V t_p} \tag{12}$$

where: C – indoor Rn-222 activity concentration (Bq m^{-3}); E – surface exhalation rate (Bq m^{-2} s^{-1}); S – effective surface area (m^2); t_p – air exchange rate (s^{-1}); V – volume of the room (m^3).

This Eq. (12) follows previous applications for indoor radon estimation cited in this manuscripts: Duenas et al. [28], Mahur et al. [29], and Kowalczk and Froelich [30]. In the practical scenario analysis presented later in this study, Eq. (12) was applied to estimate Rn-222 concentration in a model ship cargo hold. The calculations should be interpreted as a screening-level engineering approximation based on measured exhalation coefficients and simplified source-term assumptions, rather than as a full predictive transport model. In particular, the model assumes that the effective source term can be represented by the exhalation coefficient measured in laboratory conditions and an assumed effective exhalation surface area exposed to the ventilated air volume.

3. Results and discussion

To date, the only scientific studies presenting experimental data on the exhalation of Rn-222 from polymetallic nodules have been those by Volz et al. [15] and Kunze et al. [17]. These works constitute an important point of reference for further analyses; however, their scope was limited both in terms of the number of analysed samples and the morphological diversity of the studied nodules. In contrast, the present study is based on a broader experimental dataset (whole and pulverised nodule samples) and uses these measurements as input for a scenario-based estimation of indoor Rn-222 activity concentration in cargo-hold conditions.

In the present study, Rn-222 surface exhalation rate was measured in samples of polymetallic nodules of diverse characteristics (Table 1). The obtained results enabled a simulation aimed at assessing the potential risk of exceeding the permissible indoor Rn-222 activity concentrations in closed spaces where polymetallic nodules may be stored when their industrial-scale exploitation starts in future. As part of this analysis, a hypothetical ship cargo hold was used as a model example of such space, corresponding to the actual parameters of planned transport vessels designed for nodule carriage.

3.1. Rn-222 exhalation

In their study, Volz et al. [15] used individual nodules having diameters of 2.5 cm, 4 cm, 7 cm, and 10 cm sourced from four different geographical locations. The samples were purposefully selected so as to establish whether the size of a nodule affects its capacity for Rn-222 exhalation. Measurements were carried out using a commercial Rn-222 detector (Rad7, Durridge Company), which enables continuous monitoring of indoor Rn-222 activity concentration in a closed test chamber. The duration of each measurement was set at 6 h, sufficient to capture the initial phase of indoor Rn-222 activity concentration increase in the chamber.

By contrast, the study by Kunze et al. [17] applied a different experimental approach. Nodules of various shapes – spherical, irregular, and flattened – and differing sizes were randomly selected from ten distinct sampling sites. This allowed a broader representation of the material's natural variability and enabled an assessment of how geometric parameters influence Rn-222 exhalation parameters. However, only four dry samples were ultimately selected for exhalation measurements: three small, ellipsoidal nodules with lateral dimensions of approximately 2 cm, and one irregularly shaped nodule with lateral dimensions of around 5 cm. Only one measurement was performed for each of the samples. Similarly, single measurements were taken for seven wet nodules collected from different locations. The measurements were performed in continuous mode using a RadonScout Professional Rn-222 monitor (Sarad GmbH), which recorded indoor Rn-222 activity concentrations with high temporal resolution. The average exposure time in the measurement chamber was approximately 10 h.

For the sake of comparison, the results obtained by Volz et al. [15] are contrasted with our results of indoor Rn-222 activity concentration measurements in non-pulverised nodules (Fig. 2). Fit quality metrics for pulverized samples) are summarized in Table 2.

The observed differences in the slopes of the regression lines, which correspond to the rate of indoor Rn-222 activity concentration increase over time, indicate substantial variability in the Rn-222 exhalation rate across individual nodules. The recorded exhalation rates ranged from 131 $\pm$ 14.3–578 $\pm$ 24 Bq m^{-3} h^{-1} for the samples analysed in the present study, and from 16 to 781 Bq m^{-3} h^{-1} for the nodules examined by Volz et al. [15]. This demonstrates that the rate of Rn-222 exhalation is strongly dependent on the specific properties of each sample. Comparable rates were reported by Kunze et al. [17]: 63 Bq m^{-3} h^{-1} for dry and 138 Bq m^{-3} h^{-1} for wet nodules.

Such huge differences between the regression slopes may result from differences in the internal structure of the nodules, their porosity, radionuclide content (mainly Ra-226) and the radon emanation factor, as well as from the degree of nodule surface area development and the presence of microfractures facilitating migration of Rn-222 to the exterior. A significant role is also played by geometric characteristics such as the size and shape of the sample, as they can affect gas pressure distribution and diffusion pathways.

Importantly, the samples analysed in both studies were collected in different geographical locations, which might be one of the major reasons for the observed differences as radionuclide content in nodules is site specific in, which directly affects their Rn-222 exhalation capacity. As a result, even when identical measurement methodologies are applied, samples from different sites may have significantly different exhalation rate values. These differences are illustrated in Figs. (3) and (4), which present the surface exhalation rate values calculated from the performed measurements using Eq. (2).

Fig. 3 presents a comparison of the Rn-222 surface exhalation rates for selected nodule samples analysed in the present study (2/2, 9/2, 11/

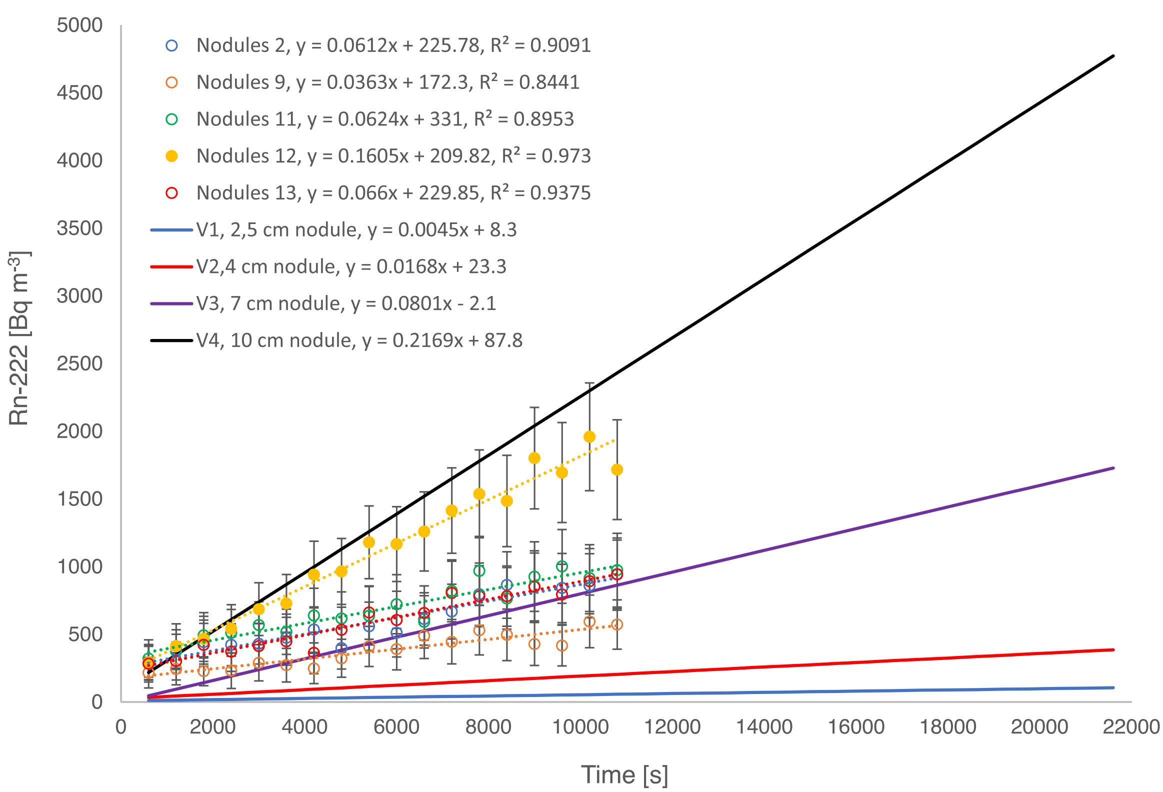

Fig. 2. Rn-222 activity concentration build-up during the first 3 h of accumulation for non-pulverised nodules analysed in the present study (symbols). Dashed lines represent linear regressions $C(t)$=a t + b used to determine the build-up rate (slope a). Solid lines show literature data from Volz et al. [15] for nodules of 2.5–10 cm diameter (samples V1-V4). Error bars represent the expanded uncertainty 2σ.

Table 2
Linear regression quality metrics for the standardized early-time interval (0–3 h; 10-min logging) used to determine the build-up slope *a* from C(*t*)=a*t* + b for pulverized samples.

Sample code	*a* (Bq m^{-3} s^{-1})	SE(*a*) (2σ) (Bq m^{-3} s^{-1})	R^2	Sample code	*a* (Bq m^{-3} s^{-1})	SE(*a*) (2σ) (Bq m^{-3} s^{-1})	R^2
1	0.0217	0.0043	0.866	9/1	0.0232	0.0076	0.699
2/1	0.0119	0.0063	0.472	10	0.025	0.0059	0.816
3	0.0141	0.0038	0.772	11/1	0.0497	0.0075	0.916
4	0.0152	0.0053	0.675	11/2	0.0442	0.0096	0.843
5	0.0075	0.0036	0.527	12/1	0.0925	0.0112	0.945
6	0.0332	0.0075	0.829	12/2	0.0645	0.0091	0.927
7	0.0424	0.0181	0.581	13/1	0.035	0.0099	0.757
8/1	0.1029	0.0122	0.947	14	0.0217	0.0051	0.824
8/2	0.0467	0.0068	0.922	15	0.0531	0.0082	0.913

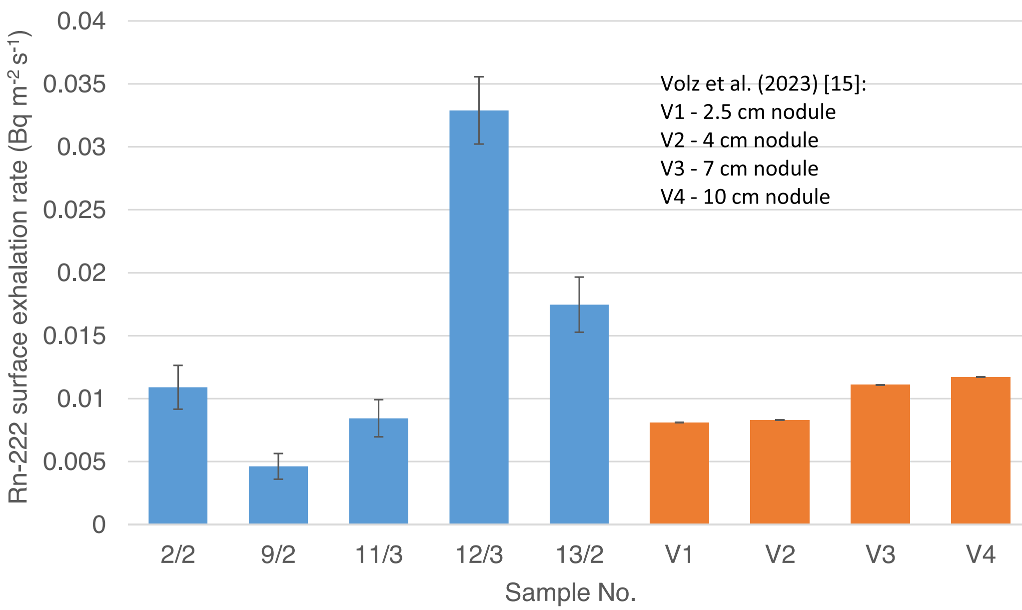

Fig. 3. Surface Rn-222 exhalation rate values for non-pulverised nodules examined in the present study (blue) and by Volz et al. [15] (orange). Error bars represent the expanded uncertainty 2σ.

3, 12/3, 13/2) and the samples studied by Volz et al. [15], which included nodules with diameters ranging from 2.5 cm to 10 cm (labelled V1–V4). The highest exhalation rate value of nearly $3.26 \pm 0.13 \times 10^{-2}$ Bq m^{-2} s^{-1} was recorded for sample 12/3. Such a high exhalation value may result from local enrichment in Ra-226, a highly porous nodule structure, or the presence of fractures and channels facilitating the transport of Rn-222. This sample stands out significantly from the others, both because of its high absolute exhalation rate and because its value clearly lies outside the range observed for all other samples. The other samples exhibited a wide range of exhalation rates, from approximately $0.46 \pm 0.05 \times 10^{-2}$ to $1.75 \pm 0.11 \times 10^{-2}$ Bq m^{-2} s^{-1}, indicating significant variability in the exhalation properties of the investigated nodules. The lowest exhalation rate value was recorded for sample 9/2, which may be associated with a denser internal structure or a lower radionuclide content. The values reported by Volz et al. [15] were more consistent, ranging from approximately 0.81×10^{-2} to 1.17×10^{-2} Bq m^{-2} s^{-1}, and showing a positive correlation with the increasing nodule diameter. Those samples originated from different locations but were analysed using a comparable methodological approach, making them suitable objects of reference for our study.

A comparison of the above results confirms that even under identical measurement conditions, samples derived from different geological settings may have significantly varying surface Rn-222 exhalation rates. This clearly indicates the significant influence of factors such as nodule porosity, Ra-226 content, the presence of microfractures, and local mineralogical characteristics on Rn-222 exhalation rate.

The differences in surface exhalation rates between non-pulverised nodules (Fig. 3) and ground nodules (Fig. 4) are primarily a consequence of the different surface areas available for Rn-222 exhalation. Pulverised nodules, due to material fragmentation, are characterised by a significantly larger specific surface area — ranging from approximately 1.15–7 m^2. In comparison, the specific surface area of the whole nodules used in the measurements was nearly 100 times smaller. Although an increased specific surface area in the pulverised samples would theoretically promote higher Rn-222 exhalation (due to shortened diffusion paths and a greater number of potential escape routes), the measurements conducted in this study revealed the opposite effect: higher exhalation rates were recorded for the whole nodules, while lower – for the pulverised samples. This phenomenon can be explained by the specific internal structure of polymetallic nodules. Whole nodules are characterised by high porosity of up to approximately 60%, meaning that a significant portion of their volume consists of voids. The presence of an extensive system of pores, channels and fractures facilitates effective migration of Rn-222 from within the nodule to its exterior, even in spite of its relatively small surface area. Under these conditions, Rn-222 can migrate more freely towards the nodule surface, resulting in high exhalation rates. In contrast, in pulverised nodules, despite their considerably larger contact area with the air, the exhalation of Rn-222 may be hindered by internal retention within the microstructure of fine particles, as well as by potential recombination and adsorption

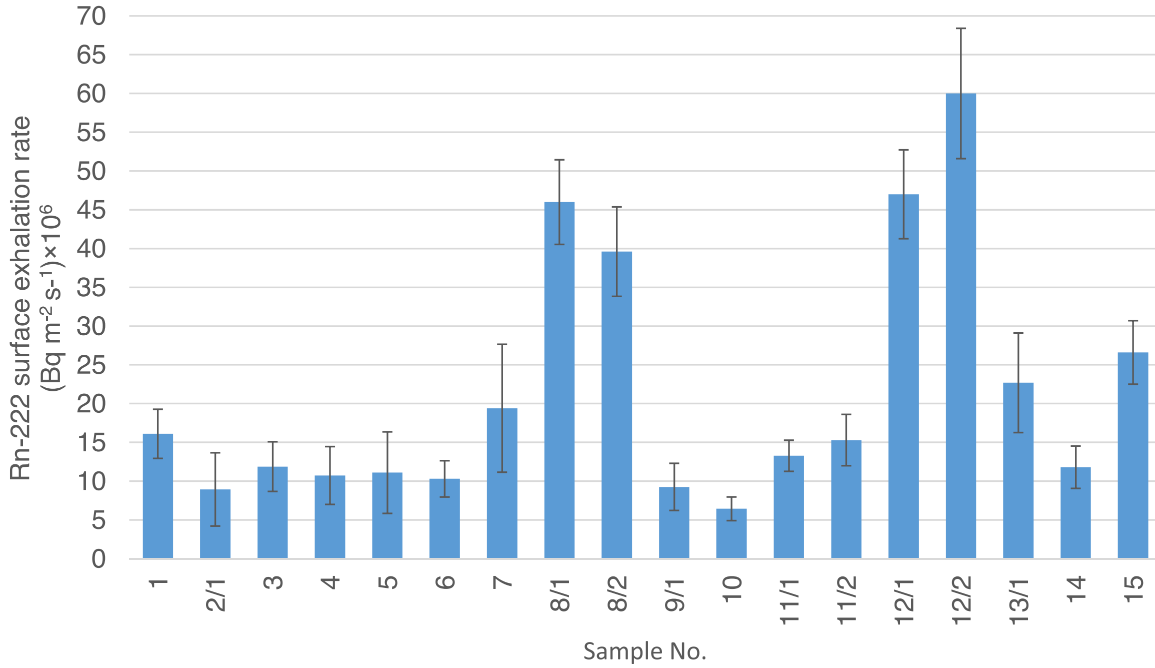

Fig. 4. Surface Rn-222 exhalation rate values for pulverised nodules examined in the present study. Error bars represent the expanded uncertainty 2σ.

processes occurring on the surfaces of these particles. Furthermore, the arrangement of grains in loose material may lead to partial pore closure and less efficient gas exchange with the surrounding air, ultimately reducing the effectiveness of Rn-222 escape. Nevertheless, exhalation rates recorded for pulverised samples can be useful when assessing indoor Rn-222 activity concentrations in confined dust-laden spaces like ship holds or storage facilities, particularly in situations where such particles remain suspended in the air or form surface layers with a large contact area with the air. Due to their high specific surface area, dust particles may represent a significant source of Rn-222 exhalation, and their presence in the air can substantially increase indoor Rn-222 activity concentrations compared to dust-free environments.

Further, to facilitate a direct comparison between intact nodules and pulverized material, the surface exhalation coefficient rate E was additionally normalized by sample mass (Fig. 5) to obtain the mass-based exhalation rate E_M (3). This normalization reduces the influence of strongly different effective surface areas between intact nodules and powders (Table 1) and highlights differences in radon release efficiency per unit mass.

Fig. 5 shows that expressing the results as a mass exhalation rate (E_M)

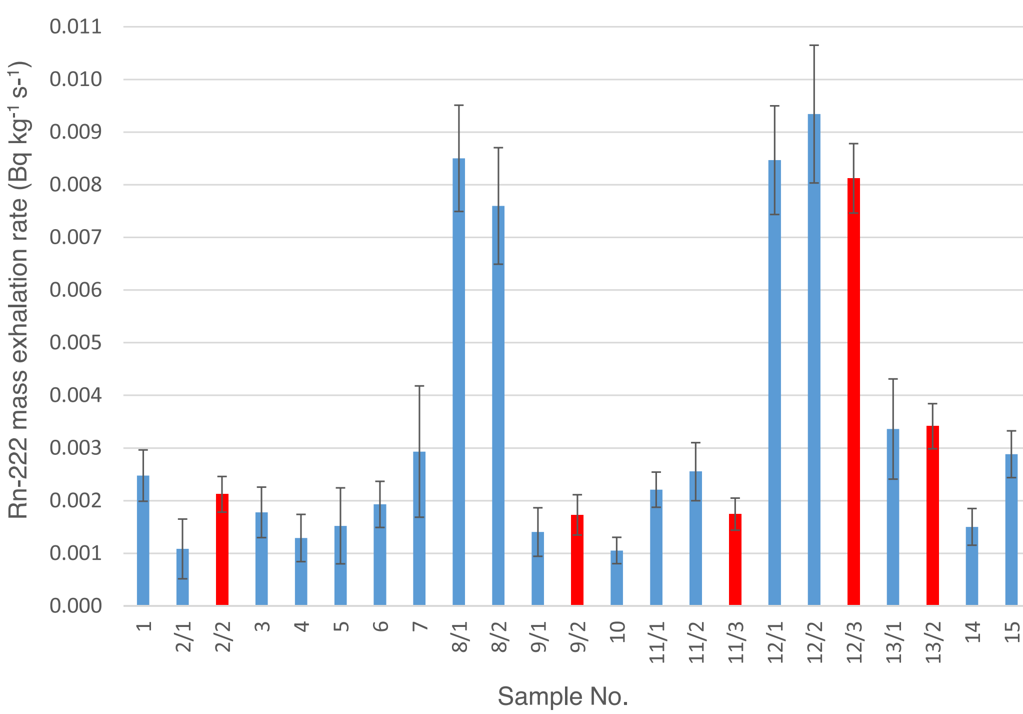

Fig. 5. Mass Rn-222 exhalation rate E_M (Bq kg^{-1} s^{-1}) for polymetallic nodule samples. Blue bars represent pulverized (powder) samples, while red bars represent intact (whole) nodules. Error bars represent the expanded uncertainty (2σ), consistent with the uncertainty reporting adopted in the manuscript.

enables a consistent comparison across sample forms. This is important because the effective emitting surface area differs by orders of magnitude between intact nodules and pulverized material: intact nodule sets have small geometric surface areas on the order of $S\approx 0.007–0.014$ m^2, whereas the pulverized samples have $S\approx 1.22–6.99$ m^2 (Table 1). Despite these large differences in S, several intact nodule sets exhibit E_M values comparable to, or higher than, many pulverized samples. This suggests that the observed variability cannot be explained by surface area alone, but also reflects sample-specific properties such as Ra-226 content, internal pore or fracture network, and radon emanation and transport pathways. Conversely, the wide spread among pulverized samples suggests that particle-size distribution and packing may also influence the measured release per unit mass, even when the nominal surface area is high.

A method similar to that used by Volz et al. [15] was also applied by Kunze et al. [17], who measured Rn-222 exhalation from polymetallic nodules using a closed chamber and a RadonScout detector. However, the authors reported only the initial slope of the radon build-up in the chamber and did not express exhalation rates per unit mass or per unit surface area. Moreover, the publication lacks details regarding the geometric parameters of the analysed samples, which makes it impossible to calculate Rn-222 exhalation intensity per unit of surface area. Therefore, the results presented by Kunze et al. [17] cannot be compared with the results obtained in our study.

3.2. Indoor Rn-222 activity concentration

The exhalation rate of Rn-222 from porous mineral materials is known to depend both on their intrinsic properties – such as radium content, grain size, porosity, permeability and effective surface area – and on environmental conditions, including moisture, temperature and air-exchange rate. Numerous studies for soils and building materials have shown clear correlations between radon exhalation and material characteristics [31–33] as well as a strong influence of moisture content and other ambient parameters on radon flux [19,34,35]. By analogy, the amount of Rn-222 exhaled from polymetallic nodules is also expected to depend on their size, porosity and surface development, and on the surrounding temperature, humidity and ventilation conditions. To date, commercial mining vessels designed for large-scale extraction of polymetallic nodules from the seafloor have not yet been deployed [36]. However, parametric analyses have been already carried out to explore design solutions for mining and transport vessels dedicated to seafloor massive sulphide extraction [37]. Future exploitation vessels will be required to meet strict safety standards regarding storage of material that may present any traces of radioactivity during transportation and transhipment. Given the current absence of operational vessels, further analyses must be based on assumed technical parameters for such ships. For example, Abramowski and Cepowski [36] assumed that the annual production rate would reach 3,000,000 tonnes of wet nodules, loading rates ranging between 5000 and 8000 h^{-1}, and a transshipment cycle lasting from 5 to 10 days. Based on these assumptions, the cargo capacity of a vessel should fall within the range of 60,000–120,000 tonnes, the unloading process being expected to take 12–18 h. Solheim et al. [37] proposed innovative concepts for deep-sea mining and transport vessels, drawing upon operational experience of the offshore oil and gas sector. Their designs included transport vessels with capacities of 7000, 24,500, and 49,000 tonnes.

Drawing on the above results of Rn-222 exhalation measurements, this section of our paper sets out to determine whether the concerns about exposure to radiation from polymetallic nodules during storage and transport are founded or not. As no nodule exploitation vessels are currently in service, we assumed a hypothetical ship of 60,000 tonnes deadweight capacity with six cargo holds, 8000 m^3 each. With an assumed cargo hold utilisation rate of 65%, a single hold would contain roughly 5200 m^3 of nodules. Given a bulk porosity of 40%, the corresponding mass of nodules per hold would be around 6500 tonnes. In order to estimate indoor Rn-222 activity concentration in the air inside a loaded cargo hold (as calculated with Eq. 12), it was necessary to determine the effective exhalation surface area – primarily the uppermost layer of nodules exposed directly to the hold's atmosphere. For this purpose, we had to assume specific dimensions of the hold, which in bulk carriers generally range from 20 to 25 m in width and 30–35 m in length. Under the assumption that nodule cargo is evenly distributed in the hold, the exposed upper surface of the cargo would be approximately 900 m^2. However, due to the inherently irregular, often spherical shape and rough texture of polymetallic nodules, combined with their high surface porosity, the actual effective surface area available for Rn-222 exhalation is likely to be substantially larger. For porous bulk materials, surface area development coefficients typically range between 1.2 and 3.0. Assuming conditions in which Rn-222 exhalation would be highest, a coefficient of 3.0 was applied in the study, leading to a threefold increase in the exhalation surface area to approximately 2700 m^2. The roughness factor scales the geometric upper surface area and should be regarded as an empirical sensitivity parameter rather than a mechanistic representation of convective or pressure-driven transport from deeper cargo layers. Accordingly, the adopted coefficient is used to explore the sensitivity of the scenario results to plausible increases in the effective source area.

To calculate indoor Rn-222 activity concentration in the air using Eq. (12), it was necessary to find out the air exchange rate for the assumed cargo hold. Ventilation of cargo spaces aboard vessels transporting polymetallic nodules is a critical aspect of both operational safety and the protection of crew health. Although specific technical standards for ventilation systems on nodule carriers have not yet been formally established, it seemed reasonable to refer to the general guidelines applicable to ships transporting bulk or potentially hazardous materials. Pursuant to the provisions of the International Convention for the Safety of Life at Sea (SOLAS) [38], enclosed cargo holds carrying bulk materials should be equipped with either mechanical or natural ventilation systems designed to effectively remove potentially harmful gases and ensure adequate air circulation. In the case of cargoes capable of releasing hazardous gases such as hydrogen sulphide, ammonia, or flammable vapours, high-capacity mechanical ventilation systems are recommended to ensure continuous and controlled airflow. For such materials, a minimum of six air exchanges per hour is typically required under SOLAS guidance. Although polymetallic nodules do emit Rn-222, the level of hazard from Rn-222 exhalation is incomparable to that posed by substances classified as dangerous under the International Maritime Dangerous Goods (IMDG) Code. At present, polymetallic nodules are not included in this category. However, due to their potential radionuclide content and capacity for Rn-222 release, it may be necessary to review relevant regulations in future or classify nodules as Naturally Occurring Radioactive Materials (NORM) [39]. In the light of this, we adopted an air-exchange rate of two air exchanges per hour as a conservative yet realistic assumption for cargo holds with nodules. This value lies within the typical range used for mechanically ventilated technical and storage spaces and is well below the minimum of six exchanges per hour prescribed for genuinely hazardous cargoes [38].

Fig. 6 presents the results of indoor Rn-222 activity concentration modelling for 9 nodule transportation scenarios based on the assumed cargo top surface area of 2700 m^2, a free cargo hold volume (i.e. not filled with nodules) of 2800 m^3, and indoor Rn-222 activity concentration for the cargo directly extrapolated from the results of measurements performed for non-pulverised nodule samples only. This is because in actual operational conditions polymetallic nodules will be transported and stored in non-pulverised form.

The calculated indoor Rn-222 activity concentrations for all the 9 scenarios are very low, not exceeding 65 Bq m^{-3} in the assumed conditions. Even if the effective exhalation surface area was hypothetically increased tenfold to 27,000 m^2, indoor Rn-222 activity concentrations in the majority of scenarios would still be well below the reference level of 300 Bq m^{-3} set out in the Council Directive 2013/59/EURATOM

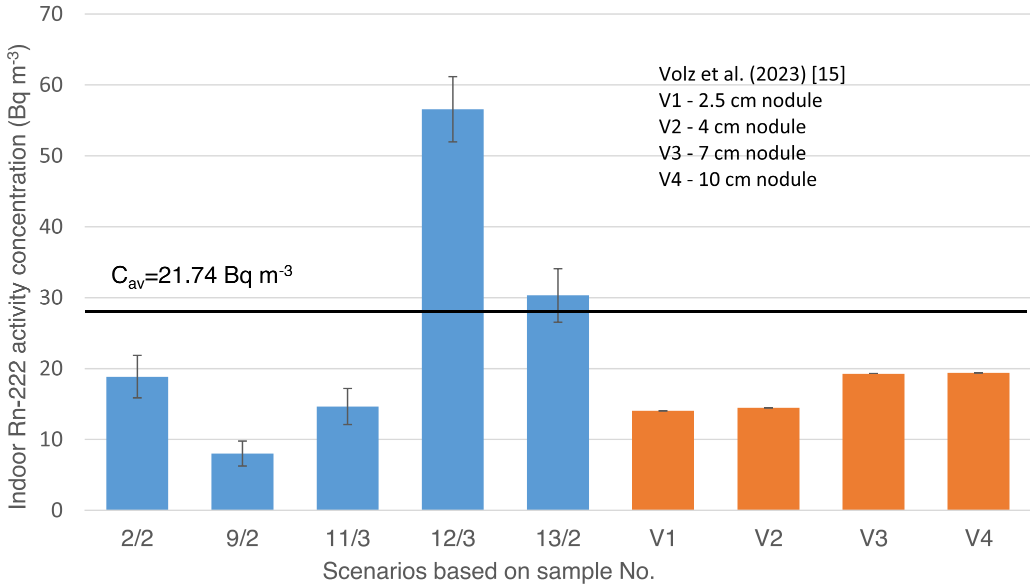

Fig. 6. Calculated indoor Rn-222 activity concentrations for non-pulverised nodules analysed in the present study and by Volz et al. [15], assuming an effective cargo surface area of 2700 m^2, a free cargo hold volume (not filled with nodules) of 2800 m^3, and total indoor Rn-222 activity concentration for the nodule cargo equal to extrapolated indoor Rn-222 activity concentration measured for the individual nodule sample. Error bars represent the expanded uncertainty 2σ.

[22]. The only exception would be the scenario based on sample 12/3 (the worst-case scenario), which is characterised by the highest calculated exhalation rate. Under this scenario, the predicted Rn-222 activity concentration would exceed the reference level, reaching 566 Bq m^{-3}. These findings indicate that, given the assumed air exchange rate and the effective exhalation surface area, exposure to polymetallic nodules did not indicate substantial radiological concern within the analysed screening scenarios from Rn-222. Even if substantially increased exposure is assumed, e.g. due to a larger effective exhalation surface area, the risk of exceeding the reference value remains limited to single cases with exceptionally high Rn-222 activity concentrations. The majority of nodules in a single cargo batch will have considerably lower surface exhalation rates as the average Rn-222 activity concentration was found to be C_{av}= 21.74 Bq m^{-3}.

The four scenarios modelled on the basis of data reported by Volz et al. [15] are comparable to those based on our own measurements. In their study, Volz et al. [15] estimated Rn-222 activity concentrations per cubic metre of polymetallic nodules using equation, which takes into account the actual cargo hold volume occupied total volume of the nodules and a time period equivalent to the half-life of Rn-222, i.e. 3.8 days. Importantly, their calculations were performed under the assumption of unventilated storage conditions. As a result, the theoretical Rn-222 activity concentration reached approximately 1 MBq m^{-3} after 3.8 days. Although mathematically correct, such a scenario is unrealistic under actual storage and transport conditions, where even minimal air circulation, system leaks, or diffusive processes effectively prevent the accumulation of Rn-222 to such high levels. In practical terms, for example in a ship's cargo hold equipped with a basic ventilation system, such concentrations are completely improbable.

In our study, we did not analyse Rn-222 diffusion from the pore spaces. Nonetheless, it was assumed that due to the limited diffusion distance, the presence of pore water, the share of the liquid phase, and the considerable tortuosity of the pore network, the effect of the diffusion mechanism on Rn-222 activity concentration in a well-ventilated storage environment would be negligible.

3.3. The radon emanation factor

However, to distinguish whether elevated exhalation values are primarily driven by higher parent nuclide content or by higher radon release efficiency, the Ra-226 activity concentrations for the tested intact nodule samples were determined and the emanation factor ε was estimated using the closed-loop equilibrium approach (4−5). This provides additional radiological context for interpreting sample-to-sample variability; for instance, a high surface exhalation coefficient may result from: elevated Ra-226 activity concentration, enhanced emanation, transport pathways associated with pore structure and micro-fractures, or a combination of both effects.

In Table 3, the closed-loop equilibrium activity concentration of Ra-226 [16], the equilibrium activity concentration of Rn-222, C_{eq}, and the derived emanation coefficient ε for intact polymetallic nodule samples are presented. The long-time growth curves were fitted to an exponential equilibrium approach model to obtain C_{eq} (5), and ε was calculated using the closed-cell formula (4). $C_{eq,max}$ is the theoretical maximum equilibrium concentration corresponding to $\varepsilon = 1$, included to separate the effect of the total Ra-226 activity in the sample ($mC_{Ra\text{-}226}$) from the emanation coefficient (ε).

Table 3 separates two key factors controlling radon release from polymetallic nodules: the radon production potential governed by the parent nuclide content, expressed here as $mC_{Ra\text{-}226}$, and the release efficiency quantified by the emanation factor ε. The equilibrium Rn-222 concentrations in the closed-loop gas phase (C_{eq}) increase with

Table 3
Summary of Ra-226 content, equilibrium radon concentration in the closed-loop system, and derived emanation factor for intact polymetallic nodule samples (V_{eff}=1.8 L).

Sample code	$C_{Ra\text{-}226}$ [Bq kg^{-1}]	m [kg]	$mC_{Ra\text{-}226}$ [Bq]	C_{eq} [Bq m^{-3}]	$C_{eq,max}$ for ε= 1 [Bq m^{-3}]	ε (-)
2/2	241 ± 17	0.0514	12.40	6061	6890	0.88 ± 0.06
9/2	367 ± 24	0.0374	13.67	3563	7595	0.47 ± 0.03
11/3	371 ± 34	0.0629	23.32	7364	12,957	0.57 ± 0.04
12/3	1315 ± 64	0.0364	47.91	19,078	26,619	0.72 ± 0.04
13/2	658 ± 34	0.0358	23.57	5586	13,096	0.43 ± 0.03

increasing production potential (e.g., sample 12/3 exhibits the highest $mC_{Ra\text{-}226}$ and the highest C_{eq}). However, the derived emanation factors vary substantially between samples ($\varepsilon \approx 0.43 - 0.88$), demonstrating that Ra-226 content alone cannot explain the observed differences in radon release behaviour. Notably, samples 9/2 and 13/2 have nearly identical $mC_{Ra\text{-}226}$ but differ markedly in C_{eq}, which directly translates into different ε values. This suggests that sample-specific physical properties, such as pore permeability, microfracture density, near-surface pathways, and moisture state, strongly influence the fraction of generated Rn-222 that becomes available in the pore/gas phase. The additional column $C_{eq,max}$ provides a transparent upper bound (corresponding to $\varepsilon=1$) and shows that the measured C_{eq} values represent roughly 43–88% of this theoretical maximum, further highlighting the role of emanation/transport processes beyond parent nuclide content.

The relatively high emanation factors obtained in this study (up to ~0.9 for intact nodules) are consistent with theoretical and experimental findings reported for other NORM-bearing materials, where emanation coefficients > 0.6 have been observed and explained by specific microstructural conditions. In particular, Sasaki et al. [40] showed that very high emanation can occur when Ra is associated with extremely fine Ra-bearing grains or is effectively distributed near grain surfaces (two-component grain configurations), yielding maximum emanation probabilities on the order of 0.6–0.75 [40]. Moreover, the emanation efficiency is known to be strongly controlled by microstructure—pore/particle size relationships and pore connectivity—rather than by parent-nuclide content alone; for example, Miklyaeva et al. [41] demonstrated systematic differences in emanation linked to microstructure types, with high-porosity cellular structures exhibiting emanation > 60% [41]. Therefore, the lack of a monotonic relationship between ε and Ra-226 content in our samples can be plausibly attributed to sample-specific microstructural differences (e.g., microfractures, pore connectivity, and possible near-surface enrichment of Ra-bearing phases), which modulate the fraction of produced Rn-222 that becomes available in the pore/gas phase.

3.4. Assessment of the risk of potential threats associated with Rn-222 exhalation during transport of polymetallic nodules

In their study, Volz et al. [15] identified three primary mechanisms that could lead to exposure to harmful α-radiation from polymetallic nodules: (1) inhalation or ingestion of radioactive dust from the surface of nodules; (2) inhalation of Rn-222 released from nodules stored in enclosed spaces; and (3) inhalation of fine nodule particles and a potential increase in the concentration of selected radionuclides during nodule handling and processing. It is important to note, however, that such exposure mechanisms do not typically occur during the transport of such cargo.

Fragmentation of polymetallic nodules during vertical transport has been investigated by de Hoog et al. [42] and van Wijk et al. [43], who demonstrated that nodule disintegration is strongly dependent on the hydraulic transport system and is primarily accelerated by mechanical impacts, chipping, and abrasion. Further insights from Borkowski et al. [44] indicate that controlled fragmentation of nodules at the mining site could significantly reduce many of the adverse phenomena that complicate both extraction and the effective hydraulic transport of polymetallic nodules through pipelines. Fragmentation of nodules is facilitated by their inherently low mechanical strength, which results from their internal structure: they are composed of multiple layers, including those containing seafloor sediments, which substantially weaken their overall integrity. Larger nodules tend to be more fragile than smaller ones due to the greater number of such layers, whereas smaller nodules are usually characterised by greater mechanical resilience [43,45].

Due to the fragmentation mechanism described above, polymetallic nodules mined and lifted in bulk to a vessel's deck would differ significantly in terms of their integrity from those collected in an intact form during scientific research expeditions. In such bulk material, the outer nodule layers, typically containing elevated radionuclide concentrations [15,17,46], are intermixed with fragments from the inner layers, which have considerably lower radiation levels. In cases where whole nodules are lifted from the sea floor for scientific purposes in the form of solid, poorly dusting lumps several centimetres in diameter, direct external exposure to α-radiation is virtually negligible. Therefore, the primary radiological concern in such scenarios arises from the possible inhalation of Rn-222 - the gaseous decay product of Ra-226. Consequently, any assessment of radiological risk associated with industrial exploitation of polymetallic nodules should not be based on the characteristics of single nodules but nodules mined and transported in bulk. Moreover, it has been shown that seawater constitutes approximately one-third of the total nodule mass [47], which results in a slow drying process, substantially minimising the risk of dust generation. As the material mined and lifted from the sea floor would consist of wet, fragmented nodules, the release of dust from nodule surfaces would be significantly limited.

According to Kunze et al. [17], the Rn-222 emanation factor in water-saturated nodule samples can be high and, depending on conditions, may exceed values observed for dry samples. This indicates that the influence of moisture on radon release is not unidirectional and may reflect competing mechanisms, including changes in recoil retention/emanation, partitioning between pore water and pore air, and gas-phase transport pathways. From a practical standpoint, transport and storage involve evolving moisture conditions (wet at recovery, followed by partial drying). Therefore, the present study uses air-dried samples primarily as a repeatable screening condition for quantifying exhalation under controlled laboratory settings. We do not claim that dry conditions universally represent the worst case for radon release; rather, the net effect of moisture remains uncertain and should be quantified in dedicated wet-dry experiments.

As outlined in Section 3.2, the reference indoor Rn-222 activity concentration value of 300 Bq m^{-3} set out in the Council Directive 2013/59/EURATOM [22] would only be exceeded for the sample with the highest recorded exhalation rate (labelled 12/3) provided that the effective exhalation surface area was substantially larger. A detailed simulation illustrating the relationship between indoor Rn-222 activity concentration and exhalation surface area, based on Eq. (12), is presented in Fig. 7. Also shown is an alternative scenario with improved ventilation using six air exchanges per hour ($n = 6$) and one air exchange per hour ($n = 1$), as opposed to the minimum scenario of two exchanges per hour ($n = 2$). Accordingly, Fig. 7 is presented as a sensitivity analysis over plausible effective-area ranges and ventilation rates, highlighting the conditions under which exceedance may occur.

As can be seen in the figure, an increase in the effective exhalation surface area leads to a linear rise in indoor Rn-222 activity concentration in the cargo hold atmosphere. In the case of two air exchanges per hour ($n = 2$), the reference value of 300 Bq m^{-3} would be exceeded if the exhalation surface area was approximately 14,000 m^2. The scenario with improved ventilation ($n = 6$) significantly reduces the increase in concentration - under this ventilation rate, the reference level would only be exceeded if the effective exhalation surface area was over 45,000 m^2. For the lower-ventilation scenario ($n = 1$), included here to represent a more conservative condition (e.g., temporarily reduced ventilation during weather-related hold closure or port stay), the reference level of 300 Bq m^{-3} would be exceeded at an effective exhalation surface area of approximately 10,500 m^2. Ventilation intensity thus plays a key role in controlling indoor Rn-222 activity concentrations in enclosed spaces. Even a substantial increase in the active exhalation surface area does not necessarily lead to the exceeding of the regulatory limit, provided that sufficient air exchange is maintained. As a result, with appropriate ventilation and reduced dust emissions, polymetallic nodules containing natural α-emitting radionuclides do not fall into the radioactive material category under the Regulations for the Safe Transport of Radioactive Material [48], as long as their activity concentration remains within the exemption limits established for

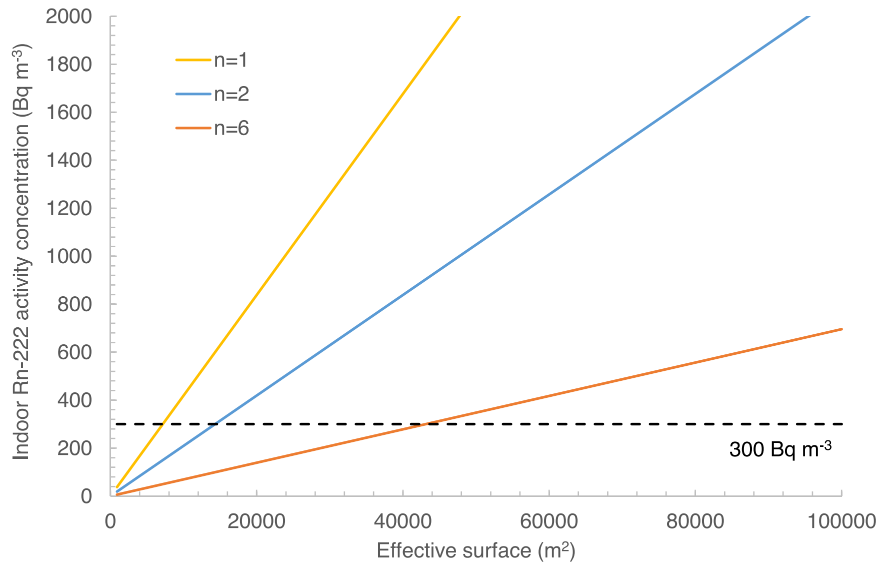

Fig. 7. Indoor Rn-222 activity concentration in a model cargo hold with a capacity of 8000 m³ containing 5200 m³ of polymetallic nodules as a function of the effective exhalation surface area for three ventilation scenarios: one air exchange per hour ($n = 1$, yellow line), two air exchanges per hour ($n = 2$, blue line) and six air exchanges per hour ($n = 6$, orange line).

NORM.

As previously mentioned, in our study we did not analyse Rn-222 transport via diffusion from the voids within a bulk nodule cargo. This was due to the fact that the limited number of samples available for our study made it impossible to build a representative laboratory-scale model of an actual nodule cargo. Nevertheless, we made an attempt to estimate at what thickness of a layer of the pulverised nodule material the outward diffusion of Rn-222 becomes ineffective, and its exhalation to the surrounding air is substantially lower. For this purpose, we performed additional measurements using the 30 g sample 11/2 arranged in three different setups. In the first setup, a 0.45 cm thick powder layer was placed in an exhalation chamber with a diameter of 10 cm (surface area $S = 78.5$ cm^2). In the second setup, a beaker with a diameter of 5 cm ($S = 19.6$ cm^2) containing a 1.8 cm thick powder layer was placed in the chamber,. In the third case, we used a beaker with a diameter of 3 cm ($S = 7$ cm^2), and a 5 cm thick layer of pulverised nodules. The measurements for all the three setups were performed with high frequency over a time frame of 300,000 s.

In the first of the setups, the pulverised nodule sample was spread evenly in the exhalation chamber as a very thin layer, which allows us to assume that Rn-222 was exhaled from virtually the entire sample volume. The layer thickness of 0.45 cm is substantially smaller than the typical Rn-222 diffusion length in loose porous materials, and the isotope's half-life ($T_{1/2} = 3.8$ days) allows the majority of atoms to reach the layer surface before their decay. The result obtained for this setup can therefore be regarded as a reference value reflecting the maximum potential Rn-222 exhalation from the entire sample mass. The measurements performed for the other two setups with thicker layers enabled us to assess the degree to which exhalation is limited by diffusion processes and radioactive decay occurring in the deeper parts of the layer. The results for all the three setups (Fig. 8) were used to assess how the efficiency of Rn-222 diffusion changes with the varying material

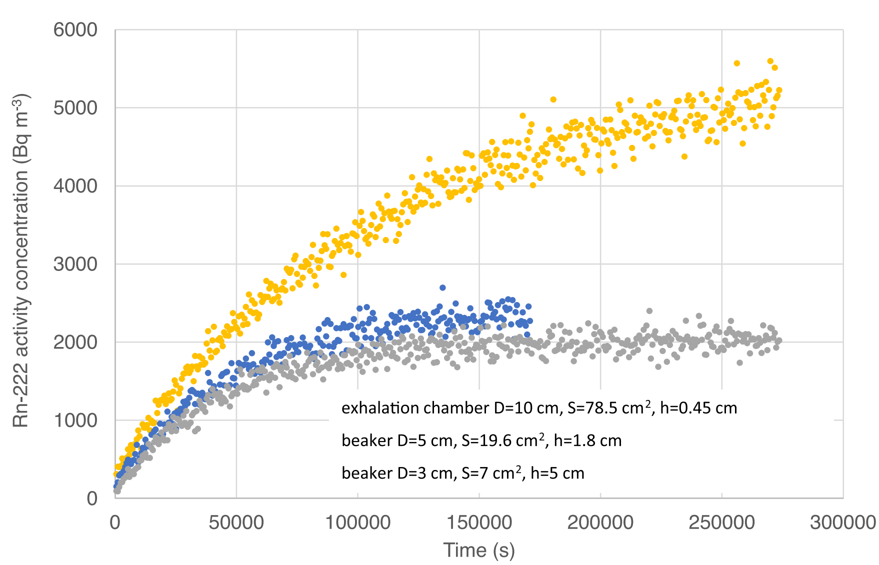

Fig. 8. Variation of Rn-222 activity concentration over time for sample 11/2 (m = 30 g) arranged in three different geometric configurations: a very thin layer (height $h = 0.45$ cm) placed in an exhalation chamber (diameter $D = 10$ cm), a medium-thickness layer ($h = 1.8$ cm) in a beaker ($D = 5$ cm), and a thick layer ($h = 5$ cm) in a beaker ($D = 3$ cm).

layer thickness.

In an ideal closed chamber without leakage and back-diffusion, the time constant of the build-up curve is determined only by the radioactive decay constant of Rn-222; under such conditions, reaching 90–95% of the steady-state concentration would require approximately 12–17 days. The much earlier levelling observed in Fig. 8 indicates that additional loss and retention processes are present and that transport limitations within thicker powder layers reduce the effective contribution of deeper material to the measured release. In closed-system radon accumulation experiments, this behaviour is commonly described using an effective build-up constant, λ_{eff}, which includes not only radioactive decay but also leakage and back-diffusion (11) [26,49,50].

The results presented in Fig. 8 clearly indicate that there is a negative relation between the thickness of the layer and Rn-222 activity concentration in the exhalation chamber. In the measurement setup with the thinnest layer (0.45 cm), Rn-222 diffused freely into the chamber, reaching a concentration of approximately 5000 Bq m^{-3}. This effect is particularly evident in the measurement setup with the thickest layer (5 cm), where the increase in Rn-222 activity concentration was slower and reached a plateau at an earlier stage compared with the setup with a thinner layer.

The long-term build-up curves also show an earlier apparent leveling-off in the thicker-layer configurations, which is consistent with the higher fitted λ_{eff} values and indicates that processes other than radioactive decay alone contribute to the observed behaviour. In the present study, leakage and back-diffusion were not determined independently; therefore, their effects are interpreted jointly as a combined non-decay contribution to λ_{eff}, following the general approach used in radon build-up analyses [26,49]. This interpretation is used here to explain the long-time behaviour shown in Fig. 8, whereas the surface exhalation rates reported in Figs. 2 and 3 were determined from the initial accumulation stage using the ISO-based linear approximation [23].

These results confirm that the spatial arrangement of pulverised material (including layer thickness) is a critical factor controlling the effective release of Rn-222 to air. Therefore, in the context of nodule transport and storage, not only the intrinsic radioactivity of the material but also its physical arrangement should be considered. Under real technical conditions, the cargo would consist mainly of irregular, porous nodules of centimetre size rather than a fine powder bed. In such systems, radon transport would occur through a complex network of interparticle voids, and the effective contribution of deeper cargo layers to airborne Rn-222 is expected to be lower than that inferred from laboratory tests on pulverised material, especially when adequate ventilation and cargo arrangement are ensured.

A key uncertainty in the cargo-hold source-term estimation is the extent to which Rn-222 generated within the bulk cargo can migrate to the free air volume. While a powder layer can strongly suppress diffusion, a pile of blocky nodules may exhibit larger inter-particle voids and potential preferential flow paths, especially under ventilation-induced pressure gradients and mechanical vibrations. Conversely, ship motion and vibration may also promote compaction of the cargo, reducing porosity and permeability over time. Therefore, our baseline assumption (effective exhalation from the upper part of the pile) should be interpreted as a simplified screening approach. The potential contribution from deeper layers is treated as an uncertainty and should be further investigated (e.g., via permeability/pressure-drop measurements, tracer tests, or CFD/porous-media modelling).

Moisture content can affect Rn-222 release in a non-linear manner by altering both emanation and gas-phase transport in the pore network. The present cargo-hold scenario calculations were parameterized primarily using exhalation rates measured for air-dried samples. This choice was made as a screening-level assumption to ensure repeatability and comparability of the laboratory measurements. However, the direction and magnitude of moisture effects on Rn-222 release remain uncertain. Moisture may enhance emanation under some conditions while limiting gas-phase transport under others. Therefore, the present scenario results should be interpreted within this uncertainty. Systematic measurements under controlled moisture contents representative of real cargo conditions are needed to better constrain this effect.

4. Recommendations

The findings of our study help to improve our understanding of the Rn-222 exhalation process in polymetallic nodules found in the Clarion–Clipperton Zone in the Pacific Ocean. The study revealed considerable variability in the Rn-222 exhalation rate between individual samples, resulting primarily from differences in their internal structure. The results enabled an approximate estimation of Rn-222 concentrations in enclosed spaces, such as cargo holds of transport vessels.

Although there are no grounds for classifying polymetallic nodules as radioactive materials under current IAEA regulations [48], it remains essential to implement basic radiological protection measures to mitigate the potential hazards to human health associated with Rn-222 exhalation. The most effective method for reducing Rn-222 concentration in enclosed spaces is to ensure sufficient ventilation. For rooms and cargo holds where Rn-222-emitting materials are to be stored, an air exchange rate of between 2 and 6 air changes per hour is recommended, depending on the exhalation intensity and operational conditions. These conclusions apply to the assumed effective exhalation area and ventilation scenarios, because the effective source area in a real cargo hold may vary with cargo structure, permeability and ventilation-induced pressure gradients, the results should be interpreted as a screening-level sensitivity assessment rather than a physically constrained prediction. To maintain effective control of radiological conditions, indoor Rn-222 activity concentration should be monitored at regular intervals using continuous radiation sensors such as the AlphaGUARD [24,25] system, particularly in areas where personnel are required to work in the proximity of nodule cargo. Where the presence of humans in areas potentially exposed to radiation is necessary, the ALARA principle (As Low As Reasonably Achievable) should be applied by reducing exposure time, increasing distance, and employing appropriate protective measures. During routine transport and storage operations, the use of personal protective equipment is generally unnecessary, provided that adequate ventilation is ensured and dust generation is effectively controlled. However, in situations where fine nodule particles are released (for example, as a result of mechanical erosion or fragmentation), the use of protective respiratory masks fitted with P3-class filters is recommended, as these effectively capture aerosols containing Rn-222 decay products. Another simple and effective measure to reduce the risk is to transport and store nodules in a moist condition as the presence of moisture binds particles and limits their suspension in the air. It should be noted that moisture control primarily mitigates dust exposure and does not automatically imply a conservative assumption for Rn-222 release, because moisture can influence emanation and gas-phase transport through competing mechanisms (e.g., changes in emanation efficiency, partitioning between pore water and pore air, and reduced gas-phase diffusion and permeability). Consequently, moist storage is recommended primarily as a practical dust-suppression measure during handling. The findings of this study may serve as a valuable reference for future deep-sea mining operators as well as regulatory authorities, such as the International Seabed Authority and national agencies responsible for radiological safety, in developing guidelines for the safe handling of materials containing naturally occurring radioactive isotopes.

The scenario analysis presented here should be treated as a first-step screening assessment. Future work should include: in situ radon concentration measurements in realistic transport and storage environments and probabilistic modelling (e.g., Monte Carlo simulation) to account for variability and uncertainty in exhalation parameters, ventilation rate, loading ratio, and environmental conditions (temperature and moisture).

Environmental Implication

Deep-sea mining may introduce large volumes of polymetallic nodules into transport and storage chains. Because nodules contain natural radionuclides, Rn-222 release may pose exposure concerns in enclosed spaces. We measured surface Rn-222 exhalation from whole and milled nodules and estimated radon concentrations in a cargo-hold scenario. Under realistic ventilation, predicted Rn-222 levels remain below the 300 Bq m^{-3} reference level and can be managed with standard controls such as ventilation and dust suppression. These findings support decision making for safe handling of nodules and provide input for future environmental and occupational risk assessments as deep-sea mining activities scale up.

CRediT authorship contribution statement

Agnieszka Dołhańczuk-Śródka: Writing – review & editing, Writing – original draft, Supervision, Investigation, Formal analysis. **Daniel Janecki:** Writing – review & editing, Writing – original draft, Visualization, Resources, Methodology, Investigation, Formal analysis, Conceptualization. **Zbigniew Ziembik:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis. **Andrzej Kłos:** Writing – review & editing, Writing – original draft, Supervision, Methodology. **Agnieszka Strzelecka:** Writing – review & editing, Supervision. **Artur Skowronek:** Writing – review & editing, Supervision. **Tomasz Abramowski:** Writing – review & editing, Supervision, Methodology. **Kamila Mianowicz:** Writing – review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

The raw data supporting the findings of this study are available in the University of Opole Research Data Repository: https://repo.uni.opole.pl/info/researchdata/UO9afef3fe573d461e953a6512c5bd2c96/

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