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Effects of coal microparticles on marine organisms: A review

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ABSTRACT

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Coal dust is a source of pollution not only for atmospheric air but also for the marine environment. In places of storage and handling of coal near water bodies, visible pollution of the water area can be observed. Coal, despite its natural origin, can be referred to as anthropogenic sources of pollution. If coal microparticles enter the marine environment, it may cause both physical and toxic effects on organisms. The purpose of this review is to assess the stage of knowledge of the impact of coal particles on marine organisms, to identify the main factors affecting them, and to define advanced research directions. The results presented in the review have shown that coal dust in seawater is generally not an inert substance for marine organisms, and there is a need for further study of the impact of coal dust particles on marine ecosystems.

1. Introduction

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Economic and social development is accompanied by an increase in energy consumption these days. Coal is an important fossil fuel that provides heat and electricity to people around the world. According to coal reserves data, the USA ranks 1st place in the world (233% of world reserves, 249537 million tons by the end of 2019), Russia ranks 2nd place (152%, 162166 million tons), Australia and China rank 3rd and 4th place (139%, 149079 million tons and 132%, 141595 million tons, respectively) (see Fig. 1) [1].

The international coal trade is heavily dependent on shipping. The leading importers of coal are China, India, the countries of Europe, and the Asia-Pacific region, while the leading exporters are Australia, Indonesia, and Russia as shown in Fig. 2 [1,2].

Handling and transportation of bulk and dusty cargo in ports are significant sources of air dust [3-5] that can also pollute the marine environment. For example, the content of suspended solids in coastal waters near some coal ports in Australia, Indonesia, China, and Columbia range from 10 to 511 mg/L [6–9]. These introduced coal particles are also a result of handling procedures of marine coal

terminals (when coal is loaded onto a ship or unloaded from a ship, when using conveyors and other uncovered transport equipment) and storage procedures, where coal is placed as open piles at coal terminals (dust and particles can enter the water area due to wind, cyclones, and heavy monsoon rains) [10,11]. The influence of wind during storage and transportation of coal, can lead to emissions of pulverized coal, which has a significant impact on the climate, human health, flora, and fauna [12–14]. Also, coal can enter the marine environment as a result of accidents on sea vessels, as well as various operations (discharge or unloading of residues after washing the cargo compartments). Although bulk cargo losses occur much more frequently than oil spills, they usually stay unrecorded [15,16].

Well-known dust suppression technologies, such as the installation of windbreak walls, irrigation of coal, use of closed conveyors and rotary car dumper systems, reduce the formation of coal dust but do not exclude it, as these measures are aimed primarily at reducing the concentration of coal dust in the residential area. Irrigation of coal can also contribute to the washout of coal particles into the sea if the storm collector is not properly designed.

To address the concentration of released coal dust, new technologies

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are also applied, for example, a smart installation in the port of Huanghua, China, allows compaction of a mixture of water and coal that is collected on the territory of the port and then sold as a product. However, the drawback of such systems is the time it requires to reach a sufficient profit revenue (the payback period for this installation is 30 years) [5].

Suspended coal particles in the absence of precipitation can remain in the atmosphere for about a week [17], which will allow them to mix with other aerosol components during their movement [18–20] and bring additional pollutants with them, into the marine environment. A significant deposition of coal particles takes place near coal terminals, where at the same time, the concentration of coal in seawater, decreases rapidly with increasing distance. Larger coal particles (> 2.36 mm) settle near the terminal, while smaller particles (<53 μ m) move much further [21].

Coal entering seawater not only changes the properties of the marine environment, but also can affect marine organisms, that are able to adapt to new habitat conditions, but possible reactions may reduce their capability to counteract any toxic effects. An important task is to assess these consequences and reactions and to determine the mechanisms that may cause them.

In recent years, a significant number of studies have been devoted to the effect of micro and nanosized particles of various origins on marine organisms [22–27]. Amongst them, some are devoted to the effects of coal particles and are presented in this review. Since there is no consensus regarding the safety of coal for marine organisms and the experimental results differ significantly depending on the species, the summary of the data will allow determining limitations. The continuing interest in this problem allows us to conclude that it is necessary to review the results of these studies, update the current review data on the biological effects of coal on aquatic organisms [28], and define directions for further possible research and solutions. In section 2 of this review, we discussed coal characteristics and their possible integrations with marine biota. Section 3 is devoted to the impact of coal particles on different groups of organisms, from plankton to fish, and sections 4 and 5 discussed future research and conclude the presented review.

2. Coal dust, its properties, and behavior in seawater

Coal is a sedimentary rock formed during two biochemical and

thermophysical processes, diagenesis and catagenesis [29]. As a sedimentary rock, coal is a complex heterogeneous mixture of organic matter and, to a lesser extent, inorganic matter of allogenic or authigenic origin. Organic matter consists mainly of non-crystalline constituents such as petrographic ingredients (lithotypes, microlitotype groups, and macerals), and in some cases, crystalline compounds (organic minerals). An inorganic substance consists of crystalline components (mineral substances from sulfides-thiosalts, oxides-hydroxides, silicates, sulfates, carbonates, phosphates, chlorides, vanadates, tungstates, etc.), to a lesser extent - semi- crystalline components (poorly crystallized mineraloids of some silicates, phosphates, and hydroxides) and, sometimes, amorphous compounds. Mineral matter, as a part of inorganic matter, includes minerals and mineraloids [30].

2.1. Chemical composition

Coal can contain various inorganic compounds of the following elements As, B, Ba, Cd, Cl, Co, Cr, Cu, F, Hg, Mn, Mo, Ni, Pb, Sb, Se, Th, U, V, Zn, and some organic compounds, with polycyclic aromatic hydrocarbons (PAHs) being especially toxic [30-36]. Coal can become a potential source of these substances in water due to their leaching [37]. For example, mercury (Hg) and lead (Pb) are elements lethal to organisms, arsenic (As) is potentially mutagenic, and As and Pb are both carcinogenic [38,39]. Hazardous compounds (Hg, Pb, As) are stable in the biosphere and bioaccumulate in food chains. Hg is toxic in both inorganic and elemental forms after being released into water, earth, and air [39]. Mn is important for algal growth [40], but its uptake by marine phytoplankton is insignificant due to its low affinity for metal [41]. However, it has been demonstrated that Mn accumulation in coastal marine phytoplankton can be prevented by high concentrations of metals such as Cu or Cd [42]. A study on three types of South African coal, showed that the leaching of metals such as Cu, Cr, Fe, and Pb in seawater at a pH level of 8 was insignificant due to the presence of humic substances in coal or seawater, which prevented them moving from coal to seawater [43]. At the same time, the high solubility of the compounds Ni and Mn was noted, and the that leaching efficiency is greatly reduced for coal with a high calcite content. Coal also contains (PAHS) in the number of hundreds, and some cases even thousands (mg/kg) [44]. The presence of PAHs was recorded in bottom sediments and suspended particles near the Hay Point coal terminal, Australia, in concentrations

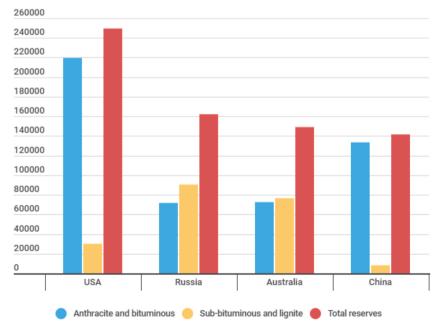


Fig. 1. Countries with the largest coal reserves in the world, million tons [1].

close to toxic levels for marine organisms [11]. It was reported that volatile organic compounds can inhibit the growth of algae while reducing zooplankton biomass [28,45]. Bituminous coal contains a significant quantity of PAHs [28,46], which is mentioned for coking coals [47]. PAHs bioavailability could cause toxic effects on marine organisms [48,49]. Physical and chemical properties of coal can be determined by sedimentation conditions in which peat is formed and its subsequent transformations [50]. For example, peat deposition conditions are related to elevated concentrations of some toxic components (such as Sulphur (S), and in some cases As and Hg), which have an adverse effect on the environment and human health [50–52]. Therefore, it is necessary to know the origin of coal and its type so as to evaluate the consequences if it enters the environment. At the same time, fossil coals are very diverse in their composition and properties [53–55].

The reactivity of organic compounds in seawater is certainly related to their chemical composition and structure, but the reactivity of organic carbon is explained by many factors, such as temperature, structure of the microbial community and the benthic ecosystem, type of mineral, surface area, redox potential, light, availability of nutrients, pH level, salinity, porosity, permeability, water content and time of exposure to oxygen [56–61]. These variables are biophysiochemical properties of the environment, therefore the reactivity of organic substances, including coal, is determined by the interaction of matter and the ecosystem [56].

2.2. Physical properties

The behavior of particles in seawater depends on many factors, especially on the vertical movement, which contributes to the deposition of particles to the marine bottom [56,62–64]. The behavior of large particles is more studied, however, small particles (<1 μ m) contribute to the chemical composition of organic carbon particles in the composition of marine bottom sediments [56,65], which indicates the importance of studying the behavior of particles of this size. Microparticles of coal dust (<53 μ m) form agglomerates in the form of spherical particles (up to 1 cm) are under static conditions in the water, or remain on the surface in the form of a film, which indicates the hydrophobicity of coal. Resistance to particle settling can also be related to surface tension. This effect however, disappeared when coal particles were placed in a container with seawater while shaken. A thin layer of fine coal particles

was observed during a sampling procedure near the coal terminal, even though no coal was being loaded onto the ship at that moment [21]. Coal was also found in bottom sediments [66]. Thus, it can be assumed that larger particles of coal will settle faster in seawater, while smaller particles will remain on the surface and prevent the penetration of light into the water column. The settling rate of micro-sized coal dust particles is much less than the settling rate of large particles, which contributes to their wider dispersion in the marine environment [21]. At the same time when the particle size decreases, they become more susceptible to aggregation caused by higher surface energy [67,68]. These aggregates settle more rapidly than coarse particles, which have fewer interactions. Important role plays the contact angle of coal particles, which associated with hydrophobicity and varies from different type of coal [69]. The interaction between coal particles and biota will depend on particles size distribution. It can be assumed that more species of flora and fauna inhabiting the marine environment will be affected by coal dust microparticles. In studies of the toxic effects of coal on marine organisms, samples of coal with different particle sizes were studied, for example, <38 µm [70] <40 µm [9], <63 µm [71,72], ≤ 425 µm [73]. Since coal has lower specific gravity than many other components of bottom sediments as the specific gravity of coal can vary, depending on the ash content $(1.2-2.9 \text{ g/cm}^3)$ [74] and by the movement of coal by water flow (larger particles of coal will move and settle with smaller and denser particles of sand and gravel) [28]. The movement of seawater promotes dispersion of suspended particles, but currents and waves can destroy large particles of coal, which will lead to continuous formation of smaller suspended particles and a longer absorption of light by particles of coal dust [71]. The spread of coal dust in seawater can affect benthic plants and organisms near coal terminals, which are most susceptible to coal dust and possible hypoxia [21]. Also, coal particles in water significantly reduces light penetration into the water by 44-99 %, depending on the coal concentration (from 38 to 278 mg/L) compared to unpolluted seawater [75].

3. Impact of coal dust particles on marine biota

Marine organisms can exhibit different responses to anthropogenic pollutants. A study evaluated whether if potentially toxic components of coal have a negative effect on aquatic biota, is actually determined by their bioavailability and concentration in the aquatic environment. Additionally, effects of coal on freshwater organisms were shown, as the

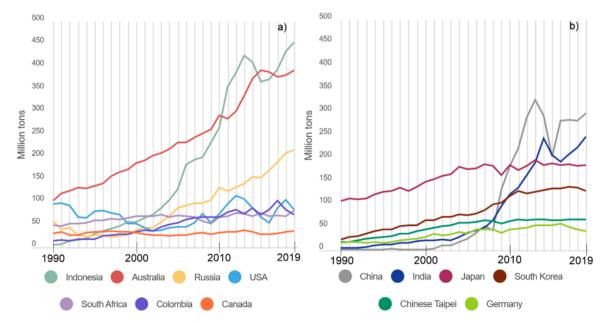


Fig. 2. Export (a) and import (b) of coal in countries for the period from 1990 to 2019, million tons per year, according to [2].

mechanisms of action are likely to be the same in seawater organisms [28]. The results of these researches are presented in Table 1.

3.1. Microorganisms, bacteria, and viruses

The effect of coal dust on microorganisms, bacteria, and viruses in seawater remains currently unexplored. It is impossible to make a conclusive assumption about the mechanism of action and its consequences due to the huge variety of species of these groups of organisms. However, some research results may allow to make an approximate estimate of the interactions between coal and some organisms. For example, in the sea bottom deposits with different oxidation-reduction states, signs of the presence of viruses were found [76–79], which can facilitate the processing of organic matter through lysis of microbial biomass [56,80,81]. It was shown that coal dust dispersed in the air after exposure to the bacterium *Bacillus subtilis* was exposed to overgrowth of the surface with calcium carbonate crystals [82].

3.2. Phytoplankton and zooplankton

Phytoplankton and zooplankton interact with dissolved and suspended substances in seawater through various processes, including active biological uptake, adsorption-desorption, particle aggregation, microbial decomposition, etc. [83]. In the ocean, phytoplankton forms the basis of the marine food chain [84], but under the anthropogenic impact, there is a significant decrease in its population (currently by 1% per year) [85] due to changes in water temperature, lighting intensity, increased acidity, deposition of substances from the atmosphere and stratification [86,87]. These changes affect the productivity of phytoplankton, giving an advantage to fast-growing species with increased adaptability [88], which creates conducive conditions for harmful algal blooms [87,89].

In a study of the impact of coal mines on phytoplankton, in particular on diatoms, it was noted that in the river, near the coal mining, the species diversity of phytoplankton was 24 % lower than in the river where coal is not mined [90]. However, this study does not indicate a direct relationship between the presence of coal in water and species diversity. A study investigated the impact of micro-sized coal particles (<20 $\mu m,$ 20–100 $\mu m,$ 100–250 $\mu m,$ and 250–500 μm fractions) on the viability of zooplanktonic crustacean Artemia salina was studied where the nauplii of A. salina were exposed to particles of four different types of coal at concentrations from 100 to 5000 mg/L at static and shaking conditions. During the 96 h period of exposure, there were no pronounced toxicological effects of coal in these used concentrations. However, the observed absorption of the coal particles in the guts of A. salina may cause possible various abnormalities in a longer-term experiment [91]. Zooplankton also plays an important ecosystem function in the marine food chain, since it has a key role in the transfer of energy from primary producers to the upper trophic levels [92,93]. With the help of zooplankton, pollutants, especially persistent organic pollutants (POPs), enter the food chain [94-96]. In seawater, zooplankton can accumulate PAHs both during sorption from water, and when feeding through phytoplankton, which is exposed to pollutants in the water [97]. Observation of zooplankton exposed to coal ash showed that the zooplankton community had been exposed to extensive restructuring over 30 years. Only 12 species of 35 species that lived in the lake in 1985, remained by the year 2015 [98].

3.3. Algae, macrophytes, and plants

The assumption that the decrease in the distribution and biomass of the green alga *Ulva lactuca L*. (Chlorophyceae) near the shores where coal waste is discharged is associated with the abrasive effect on the algae leaves was tested experimentally [99]. The impact of colliery waste particles of different sizes (3 categories: $<500 \mu$ m, $500-2000 \mu$ m, up to 2000 μ m) on *U. lactuca* samples was carried out under turbulent

and still conditions for 8 days. At the same time, ambiguous results were obtained: the coarse particles of colliery waste damaged macroalgae under turbulent conditions and could contribute decrease in their species diversity, however, under still conditions with the presence of coal particles, an increase in the growth of macroalgae was observed. The maximum weight loss was also observed in the presence of coal with a particle size of $500-2000 \mu m$ under turbulent conditions [99].

Metals leached from coal can also be toxic to seagrass [100–102]. When studying the effect of coal leachate on freshwater algae, it was found that growth inhibition occurred only in closed containers, and this effect stops when the container is aerated, which may indicate the effect of volatile organic compounds. Moreover, in an artificial ecosystem, when the concentration of coal from the leachate is from 1 to 20 vol. %, an increase of algae and bacteria and the death of zooplankton is observed [45]. Under the impact of coal waste on the sandy bottom or rocky shores, a significant decrease in the species diversity and abundance of algae is also observed [103].

The density of algae shoots and leaf growth are effective bioindicators in an environment with a lack of light [104], since reduced carbon fixation during photosynthesis reduces the amount of carbon distributed for plant growth [100,105,106]. The effect of coal particles on tropical algae (*Halodule uninervis*) was studied [75] where under laboratory conditions, an accidental discharge of coal was simulated, where organisms were exposed to five concentrations of coal (range, 0–275 mg/L and particle size <63 µm) for 28 days. This size of coal particles was chosen since such particles remain suspended in water for longer durations [21,75]. The attachment of coal particles to algae leaves was observed in less than 24 h and decreased growth was noted. The IC50 for algae was 275 mg/L at 28 days of exposure. Shoot density also decreased compared to the control group.

Coating the leaves directly with a layer of carbon particles can further reduce light penetration, leading to a decrease in chlorophyll a production [107], which prevents plant growth [75,105]. Thus, for algae, as noted earlier, it is possible that the decrease in the intensity of the light is due to the formation of suspended matter [108].

Also, the deposition of coal particles on the surface of plants (both marine and terrestrial) reduces the efficiency of photosynthesis, which was shown on mangroves [107] growing at one of the world's largest coal terminals in Richards Bay, South Africa. A 17–39 % reduction in photosynthesis was noted in leaves covered with coal dust.

Coal is dangerous for flora due to the duration of exposure. In experiments with the *Halodule uninervis* algae [75,100], the flow of water in the tanks did not promote the removal of coal particles from the leaves of the algae, just as the coal dust deposited on the leaves of mangrove trees near the port was not blown away by the wind or washed away by rain [107], thus the flora can be covered with coal for a long time [100].

3.4. Corals

An experiment similar to the H. uninervis algae was carried out for the corals Acropora tenuis [75]. In this case, coal particles settled on coral polyps and connecting tissue. Although branching corals such as Acropora tenuis are considered to be among the most resistant to sediment deposition [109], some tissues died and sloughed off the skeleton within 14 days at coal concentrations of \geq 38 mg/L. 100 % tissue mortality was observed at a carbon concentration exceeding 73 mg/L on exposure for 28 days, and at a concentration of 275 mg/L for 14 days. The LD50 for corals was 87 mg/L at 14 days of exposure and 36 mg/L at 28 days. Presumably, anoxia [110] was a lethal factor for corals, as well as increased energy consumption for a protective reaction from deposited coal particles due to light attenuation [75]. Reproduction of organisms is an important function in assessing toxicological effects. Most reef-building corals reproduce by spawning [111], like many other invertebrates. Successful reproduction plays an important role in the growth of the coral population [111], however, a decrease in the quality

Table 1

Effects of coal particles on marine organisms. The original table from [29] with adjustment and additions. The sources marked with * are presented in the review [29]. Sources marked with ** describe effects on freshwater organisms.

Species	Experiment type	Exposure condition	Coal type	Coal particles size	Experiment duration	Coal concentration	Assumed stressor	Observed effect	References
Green alga (Ulva lactuca)	Lab	Suspended colliery waste	NE England, U.K.	0-2000 μm	8 days in static and mixing conditions, 30 and 60 days in static conditions	29% by weight in waste, 1 g/L suspended waste	Abrasion by particulates	Reduced growth in the presence of waste and water movement, but increased growth with waste in still conditions	Hyslop, Davies [99]*
Seagrass (Halodule uninervis)	Lab	Suspended and settled coal particles	No data	<63 µm	28 days	0-275 mg/L	Reduced light penetration	Reduced growth, attaching of coal particles to leaves, decreased shoot density	Berry et al. [75]
Mangrove (Avicennia marina)	Field	Airborne coal	No data	5-12 μm	-	No data	Light reduction	Reduced CO_2 exchange by 17–39%, reduced photosynthetic performance	Naidoo, Chirkoot [107]*
Coral (Acropora tenuis)	Lab	Suspended and settled coal particles	No data	<63 μm	28 days	0-275 mg/L	Anoxia, reduced light penetration	Dying off and sloughed off tissues from the skeleton of corals within 14 days, 100% tissue mortality at concentrations of 73, 202, and 275 mg/L for 28 days	Berry et al. [72], Berry et al. [75]
Coral (Acropora tenuis	Lab	Suspended and settled coal particles, leachate	No data	<63 µm	72 hours	12.5-800 mg/L for suspended particles and 6.25-100 v/v of coal leachate with an initial concentration of 10000 mg/L	Physical impact	Reduced survival of embryos and larvae, subsidence of larvae reduced to 50%, no effect on fertilization, minimal anomalies in the development of embryos	Berry et al. [72]
Corals (Acropora tenuis, Montipora spp., Porites spp.)	Lab	Sediments, suspended solids	Coking coal, Queensland, Australia	63-125 μm	4 weeks for chronic exposure and 1.5 hours for acute	In the sediment - until the deposition layer reaches 30 mg*cm ² , in suspended form - 1250 mg /L	 Reduced light penetration; reduced gaseous exchange; increased expenditure of the coral's energy for cleaning the surface of tissues; potential chemical effects of leached metals 	Acute exposure led to a significant reduction in oxygen production (ranged from 112 to 135% depending on the species without prior chronic exposure and ranged from 68 to 104% with chronic exposure), reduction in light calcification rates (from 58 to 149% without chronic) and dark calcification rates (from 88 to 192% without chronic exposure and from 223 to 339% with chronic). In the presence of only chronic exposure, the reactions were less pronounced	Berry [100]
Crab (Cancer magister)	Lab	Coal mixed with sand in suspension	Westshore terminal, Tsawwassen, Canada	<300 μm	22 days	Up to 50% by weight mixed with sand	Smothering of gills by particulates	Accumulation of coal in gills at higher concentrations	Pearce, McBride [120]*
Crab (Cancer magister)	Lab	Coal mixed with sand at the bottom of the aquarium	Westshore terminal, Tsawwassen, Canada	3,9-500 µm	21 days	Up to 75% by weight mixed with sand	Smothering of gills by particulates	No measurable difference in ventilation and oxygen consumption relative to controls	Hillaby [121]*
Marine worm (Arenicola marina)	Field	Deposited colliery waste	NE England, U.K.	Average value from 209 to 283 µm		11% of sediment by weight	Physical destabilization of sediment by particulates	Worms avoided ingesting coal particles during deposit feeding (possibly based on particle size); avoidance of contaminated sediments in choice tests; reduced abundance	Hyslop, Davies [122] *
Marine predatory snail (<i>Hexaplex</i> <i>trunculus</i>)	Lab and field	Coal sediment	No data	No data	2 months in a lab experiment	No data	Cd from direct contact	The increased concentration of Cd in the hepatopancreas by 1.8 times, damaged outer epithelium and	Siboni et al. [123]*

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Species	Experiment type	Exposure condition	Coal type	Coal particles size	Experiment duration	Coal concentration	Assumed stressor	Observed effect	References
Scallop (Argopecten nucleus)	Lab	Suspended coal particles	Bituminous coal, Santa Marta, Colombia	<40 µm	12 hours	2, 9, and 40 mg/L	Availability of coal particles (as organic matter for food), physical effects of particles, chemical effects of metals	increased its permeability by 3.6 times, increased the level of metallothioneins by 3 times Increased water filtration rate by gills, reduced purification rate, selectivity in the absorption of particles, reduced oxygen consumption, physiological stress	Benitez- Polo, Velasco [9]
Bivalve mollusk (Villosa iris)	Lab	Coal mixed with sand at the bottom of the aquarium	Sydney Mine, Kentucky, USA	≼425 µm	20 weeks	Up to 50% by volume mixed with sand	Chemical effects of leached substances, the physical presence of coal particles	No significant decrease in survival, but sublethal effects were recorded: tissue necrosis in the gills, resorption of oocytes	Henley et al. [73]**
Bivalve mollusk (Crassostrea virginica)	Lab	Suspended coal particles, including leachate	No data	<40 μm	28 days	1 and 10 mg/L	PAHs	No significant adverse effect on oyster survival, shell growth, or pumping activity, no significant accumulation of PAHs in tissues of depurated oysters, assuming that coal particles have been removed from the intestine	Bender et al. [125]*
Bivalve mollusk (Modiolus modiolus)	Lab	Suspended coal particles	Lignite coal (rank 1B), Novoshakhtins k, Primorsky Krai	<10 µm	6 hours	1, 10, 100 and 1000 mg/L	Chemical effects of leached substances, the physical presence of coal particles	The number of living hemocytes' cells did not reduce, but enzymatic activity and partial depolarization of membranes reduces	Kirichenk o et al. [126]
Fish (Acanthochromis polyacanthus)	Lab	Suspended and settled coal particles	No data	<63 µm	28 days	0-275 mg/L	Availability of coal particles (as organic matter for food), physical effects of particles	Significantly reduced growth rate, no significant effect on survival	Berry et al. [75]
Fish (Danio rerio)	Lab	Methanolic coal dust extract	Mine in La Loma, Department of Cesar, Colombia	No data	48 hours	1-5000 mg/L	PAHs	Changes in the morphology of the head, tail, body, and heart. At a concentration of 500 mg/L, the survival was less than 20%, and at 5000 mg/L - 0% in 24 hours	Guerrero- Castilla et al. [138]**
Fish (Danio rerio)	Lab	Aqueous extract of coal dust	Bituminous coal, Department of Cesar, Colombia	<38 μm	72 hours	0,1-1000 mg/L	PAHs, chemical effects of leached substances	No effect on the mortality and morphological changes in embryos. Changes in genes associated with the development and function of cells of the connective tissue, hematological system, with immunological and inflammatory diseases, with cancer were revealed	Caballero -Gallardo et al. [70]**
Fish (Oncorhynchus tshawytscha)	Lab	Suspended coal particles	No data	No data	8 days	60-500 mg/L	PAHs	Increased CYP1A1 and ribosomal protein L5 expression in liver	Campbell, Devlin [48]*
Fish (Acanthochromis polyacanthus)	Lab	Suspended coal particles	No data	<63 µm	31 days	38, 73, and 275 mg/L	Chemical effects of leached substances, the physical presence of coal particles	Increased oxygen consumption, adhesion of coal to the gills, changes in their structure	Berry [100]

of water and substrate can affect these processes [72,112]. In an experiment with corals *Acropora tenuis* [72], indicators such as fertilization, the survival of embryos, larvae, and larval settlement decreased under the influence of various concentrations of coal particles (12.5, 25, 50, 100, 200, 400, 500, 600, 700, 800 and 10000 mg/L) with particle size <63 μ m and the duration of the experiment up to 96 h, depending on the stage of development. Early stages of development (gametes and embryos) were more sensitive to the effects of coal. However, these effects were likely caused by the physical impact of coal rather than toxic, as no effect on coral reproduction was noted in the experiment with coal leachate. Rather low concentrations of PAHs and metals were recorded in the leachate [72]. However, some studies have shown that Cu and Cd can be leached from coal and are toxic to corals [113–115] above threshold levels [100].

A study [100] was carried out on the effect of coal on 3 species of reef-forming corals Acropora tenuis, Montipora spp., Porites spp. to determine if there are differences in physiological changes during exposure to coal and suspended particles (carbonate deposits) with a size of 63-125 µm. In this case, 2 types of exposure were carried out, chronic (2 times a week for 4 weeks, coal and carbonate deposits were added to the container with corals in concentrations that provide a layer of sediment in the container of 30 mg^*cm^2) and acute (single exposure for 1.5 h of 1250 mg/L of coal and carbonate deposits). Acute impacts were assessed for both corals that were not previously exposed to any impact and for corals after chronic exposure. The experiment noted that the physiological responses of corals to coal particles differ from their responses to carbonate deposits, but these differences depend on the type of coral. Acute coal exposure resulted in a significant reduction in oxygen production and calcification that were not observed in sediment-exposed corals. Both chronic and acute exposure to coal and sediment significantly affected calcification. The author has identified the probable primary mechanisms of the effect of coal on corals: 1) attenuation of light, 2) reduced gas exchange; 3) increased expenditure of the energy for cleaning the surface of tissues, 4) potential chemical effects of leached metals [100]. Under acute exposure, a reduction in respiration rates was observed only in coal treatments for Porites spp., which is explained by the difference in physiological reactions in these species and the efficiency of tissue clearance from particles. However, Montipora spp. responded immediately by moving cilia and secreting mucus, thereby increasing the respiratory rate. Acropora tenuis accumulated fewer particles due to the vertical growth of branches. Also in the experiment, the concentrations of leached metals were determined, but they were not significant, so their effect was not evaluated [100]. However, metals can have long-term effects on biological processes in corals, such as respiration and reproduction [114,116], as well as PAHs can cause histological abnormalities [117] and affect coral growth [118].

Although the mechanisms of action of coal and carbonate deposits are similar, the characteristics of particles (color, adhesiveness) and changes in the abiotic environment caused by coal pollution (attenuation of light, leaching of metals) contribute to more serious changes in coral organisms [100].

3.5. Echinoderms

A study [119] examined the effects of coke-rich sediments (mainly PAHs) on the development of the sea urchin *Dendraster excentricus* (from fertilized egg to the echinopluteus stage), and no toxic effects were found. Although this study focused not on coal, but on its thermo-chemically modified product, the absence of mortality virtually eliminates any potential toxicity from coal that may have been present in these sediments [28].

3.6. Arthropods

As for arthropods, any suspended particles in the water can penetrate

the respiratory, vision, and nutrition organs, "clogging" them, and causing various damage. Accumulation of coal in the gills was observed in the experiment with crab *Cancer magister* in an aquarium with mixing coal and sand [120], which could affect oxygen consumption [28]. However, a later study found no effect of coal mixed with sand on oxygen consumption by *Cancer magister* or gill ventilation [121], although technical problems with the experiment (the coal was not in suspension, but was mixed with sand at the bottom of the aquarium) are regarded a drawback as the effect of coal on these organisms remains unclear.

3.7. Worms

In an experiment with *Arenicola marina* worms in the presence of coal in the sediments in an amount of 11 % weight/ratio, it was recorded that the worms avoided contaminated areas of bottom sediments, and their number decreased [122]. Comparing the sediment, gut contents, and fecal material of *Arenicola marina* from the heavily coal-contaminated area and the slightly polluted area, it was found that worms in the heavily polluted area selectively fed on sand grains and coal particles. Smaller particles of coal in the intestines and feces were found in worms from both sites. Despite the rejection of larger coal particles by worms in more polluted areas, there is no evidence that the lack of suitable sediment for feeding was responsible for the observed reduction in *Arenicola marina* abundance in heavily polluted areas. Perhaps the more important factor was the instability of sediments due to the presence of coal [28,122].

3.8. Mollusks

A study [123] suggested that the main factor affecting organisms might be leached Cd, which was confirmed by an experiment with the mollusk Hexaplex trunculus. The study was carried out on mollusks collected in the water area contaminated with coal and in the clean water area. An experiment was also carried out in aquariums. Both experiments showed that under the influence of coal, the concentration of Cd in the hepatopancreas of mollusks increased significantly. Damage to the outer epithelium and an increase in its permeability were recorded using microfluorimetry, which the authors explain by contact with Cd from coal deposits. Metallothioneins play an important role in the detoxification of metals in bivalve mollusks, especially of Cd [124]. Elevated levels of metallothioneins in mollusk tissues can also be caused by chemical components of coal [123]. Bivalve mollusks of the Caribbean Sea Argopecten nucleus were selected to assess the effect of coal on the growth factor, which were exposed to coal particles with a diameter of $<40 \ \mu m$ at concentrations of 2, 9, and 40 mg/L under laboratory conditions [9]. With the increasing concentration of coal, an increase in the rate of water filtration by the gills was observed, but the rate of purification was reduced. The selectivity of uptake has also been found, and it can be assumed that this species can select particles prior to feeding based on organic content or particle type, preferring coal particles with a higher organic content than microalgae cells that were added to the aquarium. The efficiency of absorption of particles at high concentrations of coal decreased, which may be related to the functional deterioration of the digestive gland caused by the greater availability of coal nanoparticles in seawater. The level of oxygen consumption also decreased due to inhibition of respiratory and metabolic functions as a result of either physical exposure to particles or chemical or biological effects of heavy metals present in coal nanoparticles [9]. Physiological stress was recorded in this experiment in the observed organism, which lies in the fact that the organism is not able to generate energy for its growth and reproduction. At the same time, for the mollusk A. nucleus, the concentration of coal and the exposure time required for the revealing of such effects were lower [9] than in corals, some bivalve mollusks, and fish [48,72,75,120,125]. The impact of pulverized coal in a sandy substrate with different percentages of coal (0, 10, 25, and 50 % vol.) on the Villosa iris mussels did not cause significant changes in the

survival of this species when exposed to the substrate for 7 weeks (3 of 40 mussels died during the experiment). The sublethal changes were observed when assessing the effect on the tissues of the mussel organs during a 20-week experiment at coal concentrations of 0 and 50 % vol. in a sandy substrate. For example, tissue necrosis was recorded in the gills, and the lipofuscin content in the kidney diverticulum was higher than in the control organisms. This experiment confirmed the hypothesis that coal in the substrate has a negative effect on reproductive and physiological functions in mussels [73]. Villosa iris is a freshwater mollusk, but histological findings are important for understanding how coal affects mollusks in general. On the contrary, the effect of PAHs was not recorded in an experiment with the mollusk Crassostrea virginica at coal concentrations of 1 and 10 mg/L in suspension (there were no changes in survival rate, shell growth, or water pumping activity) [125]. However, there are suggestions that Crassostrea virginica were exposed to high PAH contamination before coal exposure since high background PAH concentrations were recorded at the site of the collection of organisms [28]. Another experiment was devoted to determining the toxic effect of coal particles up to 10 µm size on hemocytes of Modiolus modiolus mollusks at concentrations from 1 to 1000 mg/L for 6 h, and the number of alive cells in comparison with the control remained approximately the same at all concentrations. At the same time, a decrease in enzyme activity and partial depolarization of membranes under the exposure of coal particles were recorded [126].

3.9. Fish

Fish are more subjected to sublethal effects than lethal ones when it comes to exposure to suspended particles since fish can move from an area with a higher concentration of particles to an area with a lower concentration, in contrast to sedentary or less mobile species [127]. The presence of suspended matter in the water, worsens the feeding conditions for fish, since the prey becomes less noticeable due to turbidity [128], which is a positive aspect for species that are food sources for fish. Besides, water turbidity contributes to the protection of fish larvae from large predators [127,129]. Histological analysis and observation of the gills of fish and oysters showed that solid particles of coal can adhere to the gills, having a suffocating effect, and clog lamellae [120,121], which are important for gas exchange [100,130]. The adhesion of suspended particles to the gill lamellae is expected to increase the resistance to gas transport through the gills [131]. Also, various pollutants can cause cell proliferation in the fish gills [132–134]. This structural modification can protect the gills from abrasive damage and/or reduce the permeability of the gills to toxins [133], but it also reduces the permeability of the gills to oxygen and may affect respiratory function [100,135]. An experiment on the effect of coal dust with a particle size of $<63 \,\mu m$ was carried out with the fish, Acanthochromis polyacanthus (the experimental setup was similar to *H. uninervis* and *Acropora tenuis*) [75]. At all coal concentrations, a decrease in the growth of fish is observed, as well as a change in their color. The IC50 was 73 mg/L with 28 days of exposure. With 14 days of exposure, growth inhibition was less than 50 %. Only two individuals died that were exposed to the maximum concentration of coal. An autopsy revealed the presence of coal in the fish's alimentary tracts, which could block digestion, contributing to starvation and exhaustion. There is also an assumption that coal, like suspended particles, influenced the respiration of fish [136,137]. The effects of coal on early fish life may be more significant. The effect of methanol extract of coal dust on embryos of freshwater fish Danio rerio at concentrations from 1 to 5000 mg/L showed changes in the development of embryos at the genetic level, and three malformed phenotypes were obtained depending on the concentration. All embryos exposed to a coal dust concentration of 5000 mg/L died within 24 h. The LC50 in this experiment was 223.68 \pm 29.48 and 161.55 \pm 17.16 mg/L at 24 and 48 h of exposure, respectively [138]. In a field experiment in water containing suspended solids from coal mine water, a 98-100 % mortality of Salmo gairdneri fish eggs during the incubation period was found due to a

decrease in dissolved oxygen concentration [139].

In another experiment [70] with embryos of freshwater fish Danio rerio at concentrations of coal dust extract up to 1000 mg/L and exposure times up to 72 h had no increase in mortality, but at concentrations of 0.1 and 1000 mg/L, a delay in hatching was found. Also, morphological changes in embryos were studied at concentrations up to 100 mg/L, where no differences were found. The genotoxic effect of coal was investigated, which was carried out at concentrations up to 100 mg/L by quantitative real-time PCR and transcriptomic analysis. As a result, transcripts with altered expression were identified. The altered genes are associated with the development and function of connective tissue cells, hematological system, immunological and inflammatory diseases, cancer [70]. Also, other studies have noted high mortality (more than 50 %within 30 days) and a negative effect on the reproductive function when exposed to Cd on Danio rerio individuals [140] and their sperm cells [141], which could be a concomitant factor for the impact of coal particles. Even though Danio rerio is a freshwater fish, these research findings are very useful and suggest a similar effect of coal on marine fish, which is generally consistent with such studies, the results of which are presented in this review. And the results of the investigation of changes in the transcripts emission are an important step in the further development of genotoxicology concerning the effect of coal dust on aquatic organisms. The effect of PAHs leached from coal on fish Oncorhynchus tshawytscha was also established [48]. Exposure to suspended particles of coal at concentrations from 60 to 500 mg/L for 8 days increased the expression of the CYP1A1 genes and the ribosomal protein L5. CYP1A1 plays an important role in the detoxification of such xenobiotic compounds as PAHs, while L5 plays an important role in ribosome biogenesis. The enzymes encoded by these genes have been used as a sensitive biomarker to assess the impact of organic pollutants [28,48]. However, in an experiment with fish Oncorhynchus mykiss gairdnerii, exposed to aqueous leachates of coal for 28 days, no changes in the liver were recorded as compared to the control groups [49]. It should be noted that in this experiment, the leachate was centrifuged, while in the experiment with Oncorhynchus tshawytscha [48], coal dust simply precipitated during the experiment, and the final leachate contained a significant amount of insoluble solids [28,48]. When Pimephales promelas fish were exposed to non-centrifuged coal leachate, a 100 % mortality rate was recorded within 96 h [142]. The level of oxygen consumption in tropical fish Acanthochromis polyacanthus after 5, 21, and 31 days of exposure at concentrations of suspended coal of 0, 38, and 73 mg/L was studied [100]. Oxygen consumption decreased by 17 % during 5 days of exposure, however, after 21 days of exposure at a concentration of 38 mg/L, this indicator increased by 47 % and at 73 mg/L by 18 % compared to the control groups. This indicator was also higher after 31 days for fish exposed to coal compared with control groups, by 30 % at a concentration of 38 mg/L and 38 % at 73 mg/L. Oxygen consumption is an estimate of the metabolic rate, and a significant increase in this indicator may indicate that coal was a stress factor that disrupted homeostasis [100,135,143]. Increased oxygen consumption is often stimulated by the release of stress hormones [143]. In response to stress, the immune function can be suppressed, as well as energy can be redistributed from the functions of reproduction and growth [100,144]. This study also evaluated the state of the gills of A. polyacanthus after 31 days of exposure at coal concentrations of 73 and 275 mg/L. Adhesion of a large number of coal particles to the gill surface and discoloration (darkening) of the gills were visually observed [100]. The morphology of the gills in some species can adapt to altered oxygen conditions, reversibly changing the morphology of the gills [100,135,145,146]. In this experiment, significant changes in the structure of the gills were observed, but they were different from the changes observed after exposure to other forms of suspended solids [100]. The author explains this by the "stickiness" of coal and its adhesion to the gill surfaces and clogging of the lamellae, which interferes with the transport of gas. In the gills of fish under control conditions, a thick epithelium was observed in filaments and lamellae in 48 % of cases, while under the influence of coal, only in 9%,

indicating that the morphology of the gills was reconstructed to increase the ability to absorb oxygen and compensate for the adhesion of coal particles and clogging of lamellas [100].

The accumulation of metals such as Fe > Zn > Mn > Cu > Pb > Ni >Cd and tissue abnormalities were shown when studying the consequences of transporting and dumping coal after catching fish Epinephelus sp., Lutjanus sp., Otolithes sp., Nemipterus sp., Thryssa sp. and Mugil sp. and carrying out histological analysis and spectroscopy of acid-destroyed tissues of the gills, liver, and muscles. Heavy metals can cause various deformities in fish, for example, Cd can cause deformities in bone tissue, Cd and Cu cause developmental delay, Pb has hematological and neurological effects, and Zn acts on the gills and can cause hypocalcemia [147]. Thus, in fish, the effect of coal particles can be a factor for the development of various pathologies. Reducing growth rates can also affect the reproductive function [75]. Direct physical interactions between fish and coal include the absorption of coal particles [75,120, 142], which can lead to extensive mucus secretion by the intestinal mucosa [142] and may also contribute to reducing growth rates [75, 100].

3.10. Mammals

There are no studies in the area of assessing the toxicity of coal to marine mammals, but it can be assumed that they will be indirectly affected through the food chain. It is unlikely that coal will have a direct impact, due to the more complex system of "barriers" in mammals in general. For example, in an experiment with monkeys [148], it was shown that PAHs leached from coal were unable to penetrate the skin.

4. Limitations and future research

Although various studies have been presented in the field of the effect of coal particles on marine organisms, some areas remain unexplored. There are very few studies devoted to the effect on organisms of coal particles of different size fractions, while many experiments have confirmed that the physical effect of particles is one of the most important factors. There is also insufficient information in the field of modelling the behavior of coal particles in seawater and, accordingly, of determination groups of organisms that are most susceptible to negative effects.

The limitations of the carried-out investigations allow us to point out several directions for further research. As noted earlier [28], it is necessary to investigate the bioavailability of various substances from different types of coal, which will determine the most dangerous types of coal. The study of the behavior of coal particles in seawater will reveal the most vulnerable areas, as well as elements of the marine ecosystem that may be affected. Long-term monitoring of sea areas is also promising to identify the frequency of their pollution near marine coal terminals, as well as the concentration of coal particles. Since micro-sized particles of coal (coal dust) mainly enter the marine environment, studies on the interaction of unicellular organisms and cells with such particles and their possible adaptation options are of great interest.

5. Conclusion

Coal particles in seawater may have a different effect on flora and fauna and can be divided into four reasons (the attenuation of light in the water column, the physical presence of solid matter, the release of inorganic substances and the release of organic substances) [103]. All these types of effects can be both sublethal and lethal for marine organisms [149–152], but it will significantly depend on the duration of the impact, the type of coal, and its concentration [100]. Physical effects can be conditionally divided into direct and indirect effects. The abrasion is related to direct impact, which means that the presence of a large number of coal particles in the water leads to injury, damage to animals and plants living at the seabed, on stones, or at berthing facilities [103,

153,154]. It is obvious that the presence of coal in seawater, as well as other suspended particles, will lead to the attenuation of light penetrating the water column [155], which will affect the growth of algae, seagrasses, and microalgae [71,156,157], and this will also affect the fauna that feeds on these algae [71]. Since coal from the suspended state settles on the seabed, the most direct impact is likely to be the "suffocating" effect on animals and plants [28]. The indirect physical effects consist of changing the habitat of organisms due to the deposition of coal particles at the bottom (in rock splits, etc.). But these effects can be both negative and positive [28]. For example, in naturally homogeneous muddy sediments, the presence of larger coal particles can increase the heterogeneity of the sediments, making it suitable for more animals to inhabit [155]. However, in general, the indirect effects are negative and can appear as a decrease in productivity, changes in food chains, as well as a decrease in the volume of fish and shellfish catch due to their death or biological pollution [100,158]. It was also concluded that the relevant indicator in assessing physical effects will be the dose of particles to which organisms are exposed (i.e., a function of the concentration of particles and the duration of exposure) and not the concentration of particles [159]. Different types of coal have different chemical compositions, and therefore it is difficult to provide generalized data on the chemical effects of coal on flora and fauna [100]. The content of PAHs and other toxins in different types of coal is different, and these substances can be leached out when interacting with water [37]. However, in a large volume of water, their concentrations are insignificant due to the dilution of these substances. Besides, the formation of insoluble salts upon contact with seawater, complex formation with dissolved organic substances in seawater, adsorption on particle surfaces, or redox reactions that lead to changes in mineralogical composition or solubility can cause these substances to become biologically inaccessible. Metals and metalloids that are readily-soluble under low pH conditions, such as Cd, Cu, Pb, Zn, and As, can become insoluble on contact and dilution with alkaline seawater. On the other hand, particle-bound metals and metalloids that are soluble under alkaline conditions, such as Cr and Se, can dissolve upon contact with seawater [28]. When determining the degree of leaching of substances from coal, it was shown that some chemical compounds, on the contrary, were adsorbed by coal from seawater, therefore, their concentrations in water decreased compared to control groups [160]. In general, studies have shown that the leaching of toxic trace elements [43,160] and PAHs [28,71,125] from coal is rather limited [100]. Complex mixtures of aliphatic and aromatic hydrocarbons can be leached also from open coal piles by rain [161]. The investigations have shown that coal dust is generally not an inert substance to marine biota. However, variances in the effects of exposure may be associated with a wide range of coal dust concentrations used in studies (from 1 to 1000 mg/L) and different levels of tolerance and/or sensitivity to stress factors in the studied species [9]. Several studies that have examined the effects of coal on marine organisms suggest that the physical presence of coal particles is more harmful than the effects of substances leached from coal [10]. It can be concluded that, in general, the marine fauna is more resistant to the effects of coal, than plants and protozoans. Acute toxic effects were observed more often in algae, as well as in corals. Nevertheless, an indirect effect on the fauna cannot be ruled out, due to the deterioration of conditions for feeding and reproduction.

Reactions in genes were recorded in fish at an early stage of life, which is an extremely valuable result in the field of genotoxic effects, and in the future, should be studied with other species as well. It is noted that in addition to assessing the acute impact of accidental coal discharge, studies of the impact of low coal concentrations in chronic exposure are needed [72].

The effects of coal particles on marine organisms at the cellular level remain poorly understood, although there are studies on the effects of coal on the DNA of other species cells. In this case, the main type of exposure is oxidative damage to DNA, which is caused by the production of active radicals by metals in the composition of coal particles, as well as chromosomal instability [162]. Metals can also cause genotoxicity and carcinogenicity by inhibiting DNA repair systems, which leads to genome instability and accumulation of mutations [163]. Oxidative DNA damage has also been observed in studies of effects on human cells [164].

Considerably more work will need to be done in the field of ecotoxicology and genotoxicology of the coal dust impact on marine organisms, which will allow us to assess the consequences of anthropogenic activities near the sea area and to predict possible consequences during its further development. It should also be noted that the studies do not carry out a comparative impact assessment of different types of coal in one experiment, which may cause different effects of coal particles in experiments due to different chemical compositions.

Author statement

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Declaration of Competing Interest

The authors declare no conflict of interest.

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