



Deposition of Aerosols onto Upper Ocean and Their Impacts on Marine Biota

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Abstract: Atmospheric aerosol deposition (wet and dry) is an important source of macro and micronutrients (N, P, C, Si, and Fe) to the oceans. Most of the mass flux of air particles is made of fine mineral particles emitted from arid or semi-arid areas (e.g., deserts) and transported over long distances until deposition to the oceans. However, this atmospheric deposition is affected by anthropogenic activities, which heavily impacts the content and composition of aerosol constituents, contributing to the presence of potentially toxic elements (e.g., Cu). Under this scenario, the deposition of natural and anthropogenic aerosols will impact the biogeochemical cycles of nutrients and toxic elements in the ocean, also affecting (positively or negatively) primary productivity and, ultimately, the marine biota. Given the importance of atmospheric aerosol deposition to the oceans, this paper reviews the existing knowledge on the impacts of aerosol deposition on the biogeochemistry of the upper ocean, and the different responses of marine biota to natural and anthropogenic aerosol input.

Keywords: atmospheric aerosols; atmospheric deposition; upper ocean; atmospheric nutrients; mineral dust; organic aerosols; marine biota

1. Introduction

Atmospheric aerosols play an important role in climate because they can modify, both directly and indirectly, the Earth's radiative balance. Direct effects originate from the absorption and/or scattering of solar radiation by atmospheric aerosols, whereas indirect effects are mainly determined by the influence of aerosols on both cloud droplet and ice nuclei activation and the subsequent impact on cloud radiative properties and precipitation [1]. Aerosols can be emitted directly into the atmosphere (primary aerosols) from both natural (e.g., sea-salt, mineral aerosols (or dust), and volcanic dust) and anthropogenic sources (e.g., combustion of fossil fuels, waste and biomass burning, industrial activities, mining, and agricultural activities), or formed in the atmosphere (secondary aerosols) through gas-to-particle conversion processes (nucleation and condensation) of volatile organic compounds (VOCs) and/or photochemical processing of primary aerosols (atmospheric aging) [1,2]. Due to this multitude of sources and formation processes, aerosols vary in composition and size, which grants them an important role in eliciting deleterious effects on air quality and human health. As reviewed by Heal et al. [3], aerosols cover a size range of more than five orders of magnitude, from a few nanometers (nm) to several micrometers (μm) , which strongly influences particle lifetime in the atmosphere and, hence, the spatial extent of their influence. For regulatory purposes, the ambient PM is typically quantified using the PM_{10} (aerodynamic diameter less than 10 μ m) and $PM_{2.5}$ (aerodynamic diameter less than 2.5 µm) metrics. In terms of chemical composition, the atmospheric PM consists of a complex mixture of water-soluble inorganic salts (e.g., sulphate (SO_4^{2-}) , nitrate (NO_3^{-}) ,



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). and ammonium (NH4⁺)), insoluble mineral dust (e.g., calcium sulfate (CaSO4) and calcium carbonate (CaCO₃)), trace elements of both anthropogenic (e.g., Pb, Cd, and Ni) and natural (e.g., Si, Al, Fe, Ca, and Mn) origin, biogenic organic particles (e.g., pollen, spores, and plant fragments), and carbonaceous material (which includes water-soluble and water-insoluble organic compounds plus elemental carbon) [4–9].

Once in the atmosphere, aerosols can be transported over long distances and deposited onto terrestrial surfaces. Indeed, the lifetime of aerosols in the atmosphere is determined by wet and dry deposition, with wet deposition losses being typically estimated to account for the majority of submicron particle removal from the atmosphere [10,11]. Furthermore, wet deposition is eventually more important locally or regionally, close to aerosols emission sources, whereas dry deposition would be more important in areas far from emission sources. As far as the ocean is concerned, atmospheric deposition of aerosols can impact the biogeochemical cycles of several important nutrients in the ocean, especially Fe, N, and P [12], thus enhancing ocean productivity (Figure 1). For example, eolian dust fluxes from arid regions (e.g., Saharan dust) represent a source of nutrients, namely Fe, for the open ocean and induce important changes in the microbial plankton community [13]. On the other hand, the deposition of anthropogenic aerosol components (e.g., toxic elements, such as Cu, or dissociation products of strong acids (HNO₃ and H_2SO_4) and bases (NH₃)) can potentially exert toxic effects and change the patterns of marine primary production [14,15]. Atmospheric deposition can also contribute to the oceanic dissolved organic and black carbon pool, with the latter being one of the most refractory forms of organic matter in the ocean, thus potentially affecting regional ecosystems and global carbon reservoirs [16,17]. The impact of atmospheric aerosol deposition to the upper ocean is certainly very dynamic, altering both the surface seawater chemistry and marine biological production. It is, therefore, timely to address this topic and reflect on what is known about the deposition of aerosols onto the upper ocean and their impacts on marine biota.



Figure 1. Conceptual diagram illustrating the aerosols sources (natural and anthropogenic), transport, and deposition onto marine ecosystems, inducing biotic responses. Upon deposition onto the upper ocean, aerosols can impact nutrient and toxic elements availability, ocean biogeochemistry, ocean productivity, and carbon cycling. The description of these topics can be seen in the relevant sections. Fe—iron; N—nitrogen; P—phosphorous; OC—organic carbon; BC—black carbon.

2. Mineral Dust Deposition onto the Upper Ocean

The literature shows the considerable effort that has been devoted to the quantification and identification of major dust source regions and transport pathways until deposition onto the upper ocean [18,19]. The long-lived isotope thorium-232 (²³²Th) has been used to quantify the lithogenic inputs into the oceans (e.g., [20-22]) since it is supplied by dust, whereas other isotopes (such as ²³⁴Th and ²³⁰Th) are produced uniformly throughout the ocean by radioactive decay of dissolved uranium (²³⁸U and ²³⁴U, respectively) [23]. The rate at which the dissolved ²³²Th is released by mineral dissolution can be constrained by a Th removal rate derived from either ²³⁴Th:²³⁸U or ²³⁰Th:²³⁴U, assuming that the chemical speciation and residence time of each dissolved Th isotope is independent of its supply process [23]. The calculated fluxes of dissolved ²³²Th can in turn be used to estimate fluxes of other lithogenic-derived dissolved metals, such as Fe [21,24]. As highlighted by Hayes et al. [21], one must be careful when using measurements of 232 Th to quantify dust deposition to the ocean, since the influence of coastal input must be taken into account. The authors suggest that the combined measurements of Th isotopes and ²²⁸Ra, with the later originating in coastal waters in contact with sediments, will allow the assessment of the potential sedimentary sources of ²³²Th [21]. Besides Th isotopes, the cosmogenic radioisotope beryllium-7 (7 Be) in ocean surface waters has also been used as a tracer of atmospheric deposition processes (e.g., [25-27]). As described by Shelley et al. [27], this radioisotope is deposited in the ocean predominantly via precipitation and is subsequently homogenised in the mixed layer. As the mean life-time of 7 Be is 77 days, this radioisotope can be used as a tracer of atmospheric deposition on seasonal timescales. For example, ⁷Be data in the upper ocean has been used to quantify the atmospheric deposition flux of trace elements (e.g., Al, Mn, Fe, Ti, Y, Zr, Ni, Cu, Sr, Mo, Ag, and Pb) to surface waters of the North Atlantic at latitudes greater than 40° N and further compared to trace elements flux estimates obtained by the traditional "dry + wet deposition" approach [27]. The authors reported that the trace elements flux estimates obtained by the ⁷Be proxy and the traditional approach were in good agreement for approximately half of the analysed trace elements, but there were large offsets for other trace elements (up to $40\times$) [27]. The authors concluded that the ⁷Be approach is promising for regions where precipitation samples cannot be routinely collected, but it is important to consider factors that may contribute to the abovementioned variation, such as the timescale of integration, selection of representative deposition velocities, and precipitation rates [27].

In terms of the identification of major dust source regions and transport pathways, it has been possible to ascribe dust sources to natural and anthropogenic (primarily agricultural) origins, with the mineral dust sources accounting globally for up to 81% of the emissions (the remaining 19% is assigned to anthropogenic sources) [18,28], reaching almost 100% in hyper-arid regions according to MODIS data [28]. Chen and co-authors estimated that dust emissions from hyper-arid and arid regions contribute more than 97% to the global mineral dust emissions with emission fluxes reaching up to 50 μ g·m⁻²·s⁻¹ [28]. The simulations show that the main contributors are the Sahara, Taklamakan, and Arabia deserts, with annual average dust emissions of 12.19 \pm 0.24, 7.31 \pm 0.68, and 5.23 \pm 0.21 µg·m⁻²·s⁻¹, respectively [28]. It is, therefore, not surprising that the majority of studies on mineral dust deposition onto the upper ocean has been focused on the atmospheric transport and deposition of dust from the Sahara Desert (e.g., [29–32]). Two major source regions of Sahara dust can be distinguished: one from the Bodélé depression to the south of the Tibesti Mountains, and the second covering eastern Mauritania, western Mali, and southern Algeria. These source regions are almost nonaffected by anthropogenic activities and are driven basically by the typical variables of local climate and geomorphology [33]. The marine areas most affected by dust from the Sahara Desert are the Atlantic Ocean and the Mediterranean Sea. The dust supply from the greater North African region directly affects the tropical North Atlantic in late spring, summer, and early fall, when a dusty air mass forms over the Sahara Desert and typically moves westward over the tropical Atlantic Ocean every three to five days according to NOAA's Hurricane Research Division

(HRD) [34]. The Mediterranean Sea, on the other hand, has become a hotspot in terms of atmospheric deposition research. The Mediterranean Sea is an oligotrophic quasi-enclosed basin that receives a remarkable flux of dust, primarily coming from the Sahara Desert, in the form of strong pulses. Therefore, the Mediterranean Sea is considered a low-nutrient low-chlorophyll (LNLC) region, well suited for addressing the question of the planktonic response to aerosol input [29,35]. Because of the thousands of kilometers over which the Sahara dust is transported, its influence can also extend as far as the Amazon [36], southeast USA [37], coral reefs of the Caribbean [38], and northern and southern European countries [39,40], degrading the air quality of these regions. These events represent a public health issue being associated with more than 40 thousand deaths in Europe, based on recent estimates through GEOS-Chem modelling [40]. Since the mineral dust can be transported thousands of miles over the Atlantic and Pacific oceans and deliver important amounts of nutrients to the LNLC Atlantic and Pacific gyres, it could play a significant role in primary production in those marine regions [13,31,41].

The Sahara Desert dust is mainly composed of oxides, carbonates, quartz, silicates, sulfates, chlorides, soot mixtures, calcium-rich particles, hematite, phosphates, and mixtures of silicates and sulphates [42-45]. Due to the elemental composition of the earth crust, the atmospheric input of dust particles constitutes a major source of nutrients to the upper ocean, including Fe, N, and P. Of these, Fe has received much attention, driven by the realization that Fe can be a limiting nutrient in large areas of the global ocean [46]. Given its importance for primary productivity in the ocean [31], numerous studies have addressed the release of Fe from dust particles in seawater. For example, in 2013, Bressac and Guieu [47] investigated the seasonal abiotic processes occurring once simulated mineral dust deposited on surface seawater using a series of artificial seeding experiments in minicosms. The authors concluded that atmospheric dissolved Fe concentration is driven by the quantity and age of the dissolved organic matter (DOM) pool. Under high and fresh DOM conditions (i.e., in productive periods or areas), the fate of atmospheric Fe is primarily controlled by the intense and quasi-immediate aggregation process between aerosols and DOM [47]. Under low-DOM conditions (i.e., in mixing periods or in lowproductive areas), the absence of aggregation allows a strong and transient increase in dissolved Fe concentration to occur prior to being removed through adsorption on sinking particles [47]. Additional studies addressing the impact of atmospheric dust deposition on an oligotrophic ecosystem, i.e., the Mediterranean Sea, were conducted within the framework of the project "DUst experiment in a low-Nutrient, low-chlorophyll Ecosystem" (DUNE) [48]. Experiments involved the addition of dust onto large clean mesocosms (Figure 2), which allowed for the quantification of the dissolution and adsorption processes of dissolved Fe occurring from (or at) mineral particles surface [49]. These mesocosms have the advantage of enabling studies of processes both as a function of time and depth while the aerosols are sinking. The dust deposition events were reproduced through seeding experiments in seawater using a flux of 10 g m⁻². According to Guieu and co-authors [48], a dust flux of 10 g m⁻² was chosen based on the mean annual dust deposition flux of 12.5 g m⁻², which occurred in Corsica during the period 1984–1994. In 2001, strong events $(\sim 22 \text{ g m}^{-2})$ with a short duration have been also recorded in this region. Based on these records, the DUNE's researchers chose to mimic a high, but still realistic, Saharan dust deposition event of 10 g m⁻². Nevertheless, firstly, it was necessary to produce particulate material similar to the atmospheric aerosols being deposited onto the upper ocean. This was accomplished by simulating (i) the generation of desert aerosols and (ii) chemical aging, mimicking the transport and cloud processing of desert dust within the atmosphere [49]. The generated dust was then spread at the surface of the mesocosms using sprayers, after which the Fe stocks (dissolved and particulate concentrations in the water column) and fluxes (export of particulate Fe in sediment traps) were followed for 8 days. These seeding experiments showed a rapid decrease in dissolved Fe concentration in the water column after the dust spread onto the water surface. Wagener et al. [49] suggested that this decrease could be associated with dissolved Fe scavenging on settling dust particles and mineral

organic aggregates. As expected, the particulate Fe concentrations in the water column also decreased in the first 48 h after seeding, with the largest particles settling faster than smaller particles [49]. Interestingly, Wuttig et al. [32] performed consecutive artificial dust deposition events in the mesocosm, each mimicking a wet deposition of 10 g m⁻² of dust. The authors concluded that successive dust deposition events may induce the dissolution of Fe from the dust particles due to the excess of Fe binding ligand concentrations in the mesocosms [32].



Figure 2. Pictures and schematic representation of the mesocosms underwater developed within the framework of DUNE project. (Reprinted from the work of Guieu et al. [48], under the Creative Commons Attribution 4.0 License).

Wuttig et al. [32] also demonstrated that Mn and Al exhibit different behavior in comparison to Fe, i.e., the dissolved Mn and Al increased after consecutive seedings due to dissolution processes. The authors also reported that three days after the dust additions, Al concentrations decreased because of the increase in scavenging onto sinking biogenic particles induced by biomass increase after the first dust addition [32]. This mechanism was not observed for Mn, which the authors explained as being associated with complex redox and photochemical processes controlling Mn cycling in seawater ([32] and references therein). Based on the knowledge that Saharan dust could be an important supplier of phosphorus (P) to oligotrophic waters of the ocean, Gross et al. [50] concluded that dust P originates from widespread sedimentary sources and magmatic P "hot spots", in which the latter enriches the dust in bioavailable P. Ridame et al. [31], on the other hand, showed that a simulated wet deposition event of Saharan dust resulted in a strong increase in the nitrogen/phosphorous (N/P) ratio in surface waters due to a higher input of NO_3^- relative to PO_4^{3-} . The response of the phytoplanktonic community was positively correlated to these wet deposition events. In contrast, dry deposition was not a significant source of NO₃⁻, and the primary production was likely N-limited or co-limited by both N and P [31]. Ridame et al. [31] explained these contrasting results by the different N content of the dust used in each simulated deposition event. For the simulated wet deposition event, the dust was previously subjected to physicochemical

transformations through condensation/evaporation cycles that involved HNO₃ and H_2SO_4 , thus reproducing the photochemistry, the gradients in pH, and ionic strength during cloud processing of dust particles [31]. These processes led to a ten-fold higher N content compared to that of non-processed dust, which was used to mimic the dry deposition event. Due to these differences in the atmospheric supply of N, the dissolution of processed dust released a higher content of NO_3^- than that of non-processed dust, which was considered a negligible source of NO_3^- [31]. Louis et al. [51] also assessed the dynamics of NO_3^{-} and PO_4^{3-} in seawater after a dust event to better understand the impact of dust deposition in the low-nutrient waters of the Mediterranean Sea. Almost all PO_4^{3-} released from dust was scavenged back onto sinking ferric oxide-rich particles, leading to short-term bioavailability [51]. The release of NO_3^- from dust, on the other hand, remained high all through the seeding experiment timeframe, indicating that NO_3^- from dust is likely to be bioavailable for a longer period compared to PO_4^{3-} [51]. The authors also reported that the release of NO₃⁻ and PO₄³⁻ in seawater was intrinsically linked to dust particle dynamics, governed by the quantity and composition of marine DOM [51]. Although the aforementioned studies have been focused on the role of Saharan dust inputs on the functioning and phytoplankton dynamics of a well-known oligotrophic system—the Mediterranean Sea—one can extrapolate similar effects in other LNLC areas impacted by mineral dust deposition, such as the tropical Atlantic and Pacific oceans. The mineral dust can be transported thousands of kilometers across the Atlantic and Pacific oceans (e.g., [52,53]) and deliver large quantities of nutrients (Fe, P, N, and C) to the LNLC areas of the Atlantic and Pacific oceans, which in turn could play an important role in the biogeochemical cycles and primary production at those regions. Nonetheless, the current scenario of warm atmosphere and warm sea surface and ocean acidification, potentially leading to an increase in the stratification of the surface waters in the LNLC areas, may alter the biogeochemical impact of the deposited mineral dust in these regions. Additional field measurements of aerosol-derived nutrient deposition, aided by remotesensing observations and numerical models, are required for a better understanding of the response of the marine ecosystem to deposition events in regions where atmospheric inputs play an important role.

Other studies have evaluated the impact of the deposition of Sahara dust transported over long distances. For example, Baars and co-workers [54] performed multiwavelength polarization Raman lidar observations of optical and microphysical particle properties over the Amazon Basin in order to study the dust and smoke aerosol transported from Africa. Although very clean background conditions were frequently found, in about one-third (32%) of all lidar measurements, they observed advection of smoke and dust aerosol from Africa with only minor influence from marine aerosols. In about half of these cases, African smoke particles contributed the most to the total aerosol optical depth. Nevertheless, air masses loaded with Saharan dust were found to be enriched with fire smoke particles when these air masses cross the fire areas in Africa from northeast to southwest. During transportation, Saharan dust may also affect the cloud droplet size and, therefore, increase the coverage of shallow clouds over the North Atlantic Ocean, with implications in the radiative forcing [55] and precipitation regimes [56]. The impact of dust deposition on the carbon budget in the ocean is another matter of debate. For example, it has been reported that dust aerosols from the Taklamakan Desert can be transported to the subarctic North Pacific Ocean and promote an increase of 50% in Chl-a and particulate organic carbon (POC) after deposition in the ocean [57]. Bressac et al. [58] and Desboeufs et al. [59] also suggested that POC exported to the deep ocean is strongly influenced by lithogenic particles, such as those of Saharan dust, thus promoting a ballast effect of dust [58,60]. By coupling metabolic rates in the water column and export fluxes, it has been suggested that 50% of the flux is represented by POC flux directly linked to new production by autotrophs stimulated by the dust deposition. The remaining 50% of the flux was attributed to the "lithogenic carbon pump", a process due to the formation of organic-mineral aggregates, which occurred within the upper few meters of the water column, depending mainly on

the quantity and composition of marine DOM [58–60]. In artificial seeding experiments mimicking wet deposition events, Bressac et al. [58] reported sinking velocities of Saharan dust pool between 24 and 87 m d⁻¹, corresponding to the formation of organic-mineral aggregates within the upper few meters of the water column after seeding. Desboeufs et al. [59] further reported that wet deposition of aged dust enables triggering high POC fluxes, with the lithogenic fluxes (359 to 646 mg m⁻² d⁻¹) being typically 30-fold higher than the POC fluxes (10.9 to 21.9 mg m⁻² d⁻¹). In contrast, a much slower settling was observed in seeding experiments simulating dry deposition of dust (lithogenic flux of 63.7 mg m⁻² d⁻¹) [59].

Focusing also on long-term measurements of dust deposition transported over long distances, Prospero and co-workers [61] carried out a 3-year wet deposition study in a nine-station network in Florida (southeast USA), which is impacted by African dust every year, and compared the results with estimates from global dust models. The rainfall samples were collected at the top of a 15 m tall aluminum scaffold tower to minimize impacts from local sources. The authors concluded that concentrations of Fe and Al were highly correlated, suggesting that the temporal variability of African dust concentrations is relatively coherent over a large area of south Florida [61]. On the other hand, the concentration of the elements V, Cd, As, and Pb, which are commonly associated with pollution sources, shows a very different behavior of Al, Fe, and Mn. The authors concluded that those pollution-derived elements are primarily attributable to regional and local sources that are active all year long in contrast to African dust sources, which impact the region only during the warm season [61]. It was also concluded that the data acquired on dust Fe deposition were not comparable to those derived from global dust models, suggesting the need to acquire additional deposition data from geographical locations that are impacted by dust transported over long distances to improve those global dust models [61]. This is in agreement with the paper of Prospero and Mayol-Bracero [62] on the impact of African dust on the Caribbean basin, who suggested the need to implement a coordinated approach combining laboratory, field observations, and modeling experiments to assess the relative importance of African dust in this receptor region.

3. Deposition of Anthropogenic Aerosols to the Upper Ocean

The deposition (dry and wet) of atmospheric anthropogenic N, S, P, and C (organic and black C) onto the upper ocean has become a well-known environmental problem: it can alter not only the chemistry of surface seawater but also the marine biological production. This is even more critical when some studies suggest that anthropogenic fugitive, combustion, and industrial dust emissions are underrepresented in PM_{2.5} emission global inventories by at least 10% based on GEOS-Chem simulations [63].

Recent studies have shown an increase in anthropogenic air pollutants (e.g., Mn, Pb, Zn, SO_4^{2-}, NO_3^{-}) in the North Pacific Ocean transported by aerosols from pollution events in eastern Asian countries and the Taklamakan and Mongolian deserts [e.g., [64,65]. Fu et al. [64] signalized that remote areas of the western North Pacific Ocean are influenced by terrestrial biomass burning emission from East Asia, which was demonstrated by the relatively low depletion of Cl⁻ [64]. The continental long-range transport influences the chemical composition of secondary aerosol species (e.g., SO_4^{2-} , NO_3^{-} , NH_4^+); however, based on the collected data, Fu et al. suggested that the western North Pacific Ocean is the primary source of NH_4^+ [64]. In any case, the frequent occurrence of dust and haze events in East Asia seems to greatly contribute to the atmospheric deposition of macronutrients into the western North Pacific Ocean [64]. In fact, dust events contribute mostly to the dry deposition of P, Ca, and lithogenic elements, such as Al, Ti, or Fe, while haze events in polluted and populated cities of eastern Asian cities contribute to a sharp increase in the dry deposition of particulate inorganic nitrogen (sum of NO₃⁻ and NH₄⁺ 26,100 μ g m⁻² d⁻¹), SO₄²⁻ (20,700 μ g m⁻² d⁻¹), Zn (330 μ g m⁻² d⁻¹), and Cd $(1.3 \ \mu g \ m^{-2} \ d^{-1})$ in the western North Pacific Ocean [64]. By 2050, the net deposition of nitrogen to the oceans is estimated to be 36 Tg N yr^{-1} based on the RCP 6.0 scenario [66]. The same study highlighted that the atmospheric nitrogen fluxes are enhanced tenfold due to anthropogenic activities, mainly from combustion, agriculture, and shipping [66], which are disturbing the global nitrogen cycle and the marine ecosystems [67]. Doney et al. [15] demonstrated that atmospheric inputs of dissociation products of strong acids (HNO₃ and H₂SO₄) and bases (NH₃) alter surface seawater alkalinity, pH, and inorganic carbon storage. The effective net atmospheric input is acidic in the global ocean, leading to a decrease in surface alkalinity and a net air-sea outflow of CO_2 [15]. Additional contrasting impacts arising from anthropogenic nitrogen deposition onto the ocean include: (1) the potential increase in N_2O emissions, thus increasing radiative forcing, and (2) the increase in primary production and export production to the deep ocean, removing CO_2 from the atmosphere and, therefore, decreasing radiative forcing [68]. Furthermore, in a global change scenario where extreme episodes, such as acid rain or wildfires, will be more frequent, anthropogenic acids might affect the ocean biogeochemistry of some mineral elements [69,70]. Aerosols pH, chemical composition, particle size, and surface processes of the aerosols during transport play a key role in the dissolution of inorganic elements, such as phosphates and Fe (from insoluble to a soluble form), increasing their bioavailability in the ocean [69-71]. This is particularly critical since these elements are mostly present in the oceans in insoluble forms (e.g., only 3% of Fe is soluble [71]), and an increase in the anthropogenic input may increase the amount of bioavailable inorganic elements. A recent study also demonstrated that anthropogenic aerosol Fe fluxes, mostly associated with combustion sources, are underestimated over the world [72]. Using isotope measurements of Fe (δ^{56} Fe), Conway et al. [72] observed a dramatic difference in soluble aerosol δ^{56} Fe between samples collected from Saharan air masses and those collected from North American or European air masses. The authors reported that the aerosols dominated by the Saharan dust plume were characterized by near-crustal δ^{56} Fe in both the bulk and water-soluble phases (+0.12 \pm 0.03 and + 0.09 \pm 0.02‰, respectively). In contrast to the Saharan-dominated aerosols, the aerosol samples collected from North American or European air masses showed lighter δ^{56} Fe values in the water-soluble phase (-0.8 to -1.6%; mean -0.91%), an observation that was linked to anthropogenic activity [72]. The δ^{56} Fe measurements and geochemical modelling also showed that European and North American soluble aerosols comprise ~50–100% anthropogenic Fe [72]. In another recent study using δ^{56} Fe data, Pinedo-González et al. [73] reported evidence of anthropogenic Fe in seawater $(-0.23\% > \delta^{56}\text{Fe} > -0.65\%)$ collected across the central North Pacific Ocean (158° W from 25° to 42° N), downwind from industrial emissions in east Asia. The authors also measured the isotopic composition of dissolved Pb in the same surface samples collected for Fe isotopes, concluding that the ²⁰⁸Pb/²⁰⁷Pb and ²⁰⁶Pb/²⁰⁷Pb ratios are similar to those in anthropogenic Asian aerosols [73]. An isotope mass balance further suggested that anthropogenic Fe contributes to 21–59% of the dissolved Fe measured in the North Pacific Ocean [73]. These data demonstrate that anthropogenic Fe is an important source of Fe to some open-ocean regions, playing a key role in marine productivity and carbon cycling [73]. In terms of anthropogenic input of P, Mahowald [74] estimated that, globally, the average anthropogenic inputs are approximately 5 and 15% for total P and PO_4^{3-} , respectively, and may contribute as much as 50% to the deposition over the oligotrophic ocean where productivity may be phosphorus-limited [74]. In the same study, the authors estimated that anthropogenic PO₄³⁻ may contribute to a potential new primary production of 0.5 TgC a^{-1} , suggesting that atmospheric P might be an important source in the open ocean [74], although considerably less relevant (0.1 Tg P y^{-1}) than other sources, such as surface runoff (1.5 Tg P y⁻¹) [75]. However, the increasing frequency of extreme weather events will play a key role in the biogeochemistry of this macronutrient, intensifying the P limitation in coastal waters [76], increasing the ratios N:P in the ocean, and thus, posing risks on marine ecosystems [75]. Yu et al. [76] showed that dust events contribute to the increase in insoluble phosphorus as well as nitrogen, while haze events promote the increase in soluble forms of such macronutrients in the northern Chinese coast. In another recent study carried out between the Arctic Ocean and the South Pacific, it has been shown

that acidic substances with an anthropogenic origin, namely SO_4^{2-} , can react with sea-salt particles over the open ocean, thus shortening the lifetime of anthropogenic S in the marine boundary layer due to dry deposition [77]. These findings also suggest that the key role played by sea-salt particles on the climate are not being accurately accounted for in the simulation of clouds, aerosols, and air–sea exchanges and that chemically modified marine aerosols must be also considered in those climate models.

Atmospheric deposition of organic carbon (OC) and black carbon (BC) into the oceans is also important to the global carbon budget. Based on monthly satellite measurements of aerosol size distribution, temperature, and wind speed, Jurado et al. [17] suggested an important spatial and temporal variability of the atmosphere-ocean exchanges of OC and BC at different scales. These authors estimated that global dry and wet deposition of aerosol OC is approximately 11 and 47 Tg C y⁻¹, respectively, with wet deposition fluxes being more important locally and as a temporal source of OC to surface waters due to their pulsing variability [17]. Dry and wet deposition of aerosol BC to the global ocean was estimated to be 2 and 10 Tg C y⁻¹, respectively, with higher fluxes in the Northern Hemisphere and for inter-tropical regions [17]. In parallel measurements of dry (PM₁, PM₂₅) and wet (rain, snow, mixed) deposition of OC and elemental carbon (EC) into the Baltic Sea, Witkowska et al. [78] also showed that dry deposition load of carbon was on average two orders of magnitude lower than that introduced by wet deposition. Moreover, aerosol OC was more effectively removed from the air with rain than snow, while an inverse relationship was reported for EC [78]. The atmospheric cleaning of highly soluble OC was observed to be most effective on the first day of precipitation, while the hydrophobic EC was removed more efficiently when the precipitation lasted longer than a day [78]. Bao et al. [16] also reported that the concentration of water-soluble BC (WSBC) in marine aerosols was highly positively correlated with the concentration of water-soluble OC (WSOC), suggesting that these carbonaceous aerosol components might be released by similar processes (e.g., dissolution of soil particles and fossil fuel burning) [16]. According to Bao et al. [16], WSBC is defined as the BC that can be dissolved in ultrapure water, as most of the atmospheric BC deposited in the upper ocean is in the form of wet deposition [17]. It has been reported that oxidation of BC can increase its water solubility in aerosols [79] and rainwater [80]. As highlighted by Bao et al. [16], globally, a large amount of BC is emitted by combustion sources to the atmosphere $(4.3-22 \text{ Tg yr}^{-1})$, thus suggesting that the oxidation of BC might be a potentially major source of WSBC to the ocean. In fact, approximately 45% of the polycyclic aromatic compounds (which include BC molecules) identified in the aerosol samples were also found in seawater, suggesting a potential contribution of atmospheric deposition to the oceanic dissolved BC pool [16]. The authors estimated that the atmospheric contribution of dissolved BC to the global ocean was 1.8 ± 0.83 Tg yr⁻¹, with approximately 70–80% in the form of wet deposition [16]. These results suggest that the increase in anthropogenic BC emissions is likely to enhance the flux of the atmospheric deposition of BC to the ocean, with expected effects on biogeochemical cycles. In this regard, Mari et al. [81] studied the rain-mediated input of BC to the ocean and its consequences on nutrient concentrations and particle dynamics in Halong Bay, Vietnam, during a 24-hour cycle impacted by short and heavy rainfall events. The acquired data suggested that once introduced in the upper ocean via wet deposition, BC particles created a thin layer of sinking surface-active aggregates, acting as a net-like scavenger for DOM, nutrients (especially phosphate), and small particles ($<5 \mu m$, down to viruses) [81].

Research on the impact of atmospheric BC deposition into the Arctic Ocean has also been receiving increasing attention. It is well known that the Arctic region is especially sensitive to global changes and increases in BC emissions may contribute to the amplification of Arctic warming. It has been reported that atmospherically deposited BC becomes available into the Arctic Ocean with the increasing melting of sea ice during summer, thus influencing the abundance of dissolved BC in the surface waters [82,83]. Moreover, it has also been demonstrated that anthropogenic aerosol organic matter is the main source of fossil DOM in glacier waters and runoff [84]. As such, the deposition of labile anthropogenic organic aerosols is likely to exert an important impact on glacially dominated coastal ecosystems or the open ocean, where they could act as a primary driver of carbon cycling and alter the marine food webs [84]. Furthermore, the presence and the nature of organic ligands may also control the dissolution and subsequent size distribution of aerosol trace metals in seawater. Fishwick et al. [85] reported that the content of organic ligands (such as glucuronic acid and desferrioxamine-B) may impact the size distribution of aerosol-derived Mn in seawater, whereas changes in other key physico-chemical parameters in seawater (i.e., surface seawater temperature, pH, dissolved O₂ concentration) had little effect on the proportions of Mn, Co, Ni, and Pb released from aerosols. On the other hand, the same authors also concluded that aerosol source and composition had the most significant effect on the dissolution of aerosol Co and Pb, with the most anthropogenically influenced aerosol samples displaying the highest fractional solubilities in seawater, suggesting that anthropogenic aerosols may control the inputs of Co and Pb as well as of other (e.g., Fe) elements to the upper ocean making them more bioavailable [72,73,85].

4. Effects of Atmospheric Deposition on Marine Biota

Atmospheric deposition can favor the growth of some planktonic species (according to their nutritional needs), as it is a source of inorganic nutrients and organic matter [7,86,87] (Figure 3). The essential elements that limit marine phytoplankton productivity include C, N, P, and Fe, in an average proportion of 106 C:16 N:1 P:0.0075 Fe, known as the Redfield ratio [88]. Mineral dust deposition provides a source of these elements to the open sea surface waters and can have important impacts on marine biogeochemical cycles and possibly influence the climate [14,19,34,74,89,90].



Figure 3. Conceptual diagram illustrating the main processes of atmospheric aerosols (wet and dry) deposition and main changes occurring in the marine biota.

At the Mediterranean Sea, after the regular phytoplanktonic bloom during the spring season, the surface mixed layer is isolated from the deeper waters by thermal stratification [91,92]. Under these conditions, the N and P concentrations decreased, resulting in lower primary productivity and lower phytoplankton biomass [93,94]. During the entire stratification period, the main source of allochthonous nutrients to the oligotrophic surface waters is atmospheric deposition [31]. By bringing new macro-nutrients (N and P)

(e.g., [92,95]) and Fe (e.g., [32]) to the Mediterranean surface waters, Sahara dust deposition is considered to play a key role in controlling primary production within this region [31]. In this regard, Marín-Beltrán and co-workers [96] carried out aerosol seeding experiments in microcosms using seawater collected in different seasons (summer, winter, and spring) along the coast of Barcelona and Blanes and offshore of the Balearic Islands to assess the effect of Sahara dust and anthropogenic aerosols on the composition of the marine bacterioplankton community. Aerosol samples were collected on quartz fiber filters on days when Saharan dust particles arrived at the northeastern Iberian Peninsula (named "SD" aerosols) and during normal anthropogenic conditions ("AA" aerosols). The aerosol seeding experiments consisted of 0.8 mg L^{-1} of either SD or AA samples. In order to verify that no effect on the fertilization could be attributed to the filters' material, two additional microcosm containers were either not seeded or seeded with a blank filter processed identically as the filters with atmospheric aerosols. The authors concluded that bacterial diversity varies significantly between different seasons (with higher changes occurring in summer) and locations and that aerosols yielded major changes in the bacterial communities. The Alphaproteobacteria, Betaproteobacteria, and Cyanobacteria groups were stimulated by both types of SD and AA particles. On the other hand, the groups belonging to the phylum Bacteroidetes increased their abundance due to anthropogenic aerosols, while the Cyanobacteria phytoplankton groups were more stimulated by the Sahara dust. Marañón et al. [13] also assessed the effects of Saharan dust on the abundance, biomass, community structure, and metabolic activity of oceanic microbial plankton, but this time in the central Atlantic Ocean. The authors reported that different groups of phytoplankton and bacterioplankton responded differently to Saharan dust addition. Nonetheless, the sign and magnitude of the metabolic response to Saharan dust in terms of primary and bacterial productivity depended on the ecosystem's degree of oligotrophy, and it was modulated by competition for nutrients between phytoplankton and heterotrophic bacteria [13].

The deposition of anthropogenic aerosols to the upper ocean can also impact marine biota in different ways. To assess the short-term response of phytoplankton communities to aerosol deposition, Paytan et al. [14] performed bioassay experiments on northern Red Sea surface seawater using locally collected dry deposition aerosol samples (with different origin and chemical composition) that represent the bulk of the deposition in the studied area. The authors demonstrated that the response of phytoplankton growth to aerosol additions depends on specific aerosol constituents and differs across phytoplankton species. While aerosol additions enhanced growth by releasing N and P, aerosol samples with high Cu concentrations exerted toxic effects in picoeukaryotes and Synechococcus but not in Prochloro*coccus* [14]. Copper is a key trace element for marine organisms; however, above a given threshold, it becomes toxic causing growth inhibition on photosynthetic species (e.g., [97]), impairing the primary productivity, and changing phytoplankton community structure on a global scale [98]. According to Paytan et al. [14], the deposition of anthropogenic aerosols can potentially change patterns of marine primary production and community structure in high-aerosol, low-chlorophyll areas. Bioassay incubation experiments in the low-nutrient low-chlorophyll western tropical Atlantic Ocean of Barbados demonstrated that Fe- and N-rich Sahara dust deposition induces P limitation, favoring the growth of phytoplanktonic organism of the genus *Prochlorococcus* [99].

Desert dust and industrially derived $PM_{2.5}$ are also known to induce impacts in Asia, especially on the Japan Sea and in the Pacific Ocean. In a shipboard experiment, oligotrophic oceanic surface water was supplemented with aerosols collected in nearby coastal mountains, simulating the atmospheric input deposition from that region [53]. The input of N (as NO_3^-) was found to be the major driver for alleviating the low N concentrations to phytoplankton, increasing its biomass, and inducing changes in the microbial communities. In another study carried out at a coastal site on the east coast of Korea, Park and co-workers [100] reported that the total deposition (wet and dry) of anthropogenic NO_3^- and NH_4^+ could contribute to approximately 2.4 and 1.9% of the primary production in the coastal areas east of the Korean Peninsula and in the East Asian marginal seas, respectively. These percentage values are probably underestimated because the dry deposition of reactive nitrogen gas was not accounted for in this study [100]. Duarte and co-authors [101] demonstrated an increase in the phytoplankton biomass (7-fold) and production (10-fold) in phytoplankton communities from the subtropical NE Atlantic, favoring diatoms instead of picocyanobacteria after 4 days of exposure to aerosol inputs. Dinoflagellates, unicellular microorganisms associated with harmful algal blooms, also benefit from Asian Fe- and N-enriched aerosols, and therefore, an increase in anthropogenic aerosol emissions could lead to more algal bloom events in the future [101].

Liao et al. [102] found high trace metal concentrations, namely Fe, Mn, Zn, and Cu, in phytoplankton collected in the open ocean region of the Western Philippine Sea, which was assigned to anthropogenic aerosols from eastern Asia. For these elements, the size-fractioned plankton ratios were significantly higher than the reference intracellular ratios and lithogenic ratios (normalized to Al), suggesting a possible influence of extracellular adsorption or aggregation on plankton and the role of aeolian deposition of anthropogenic particles generated from fossil fuel burning in East Asia, respectively. Similarly, Liu et al. [103] reported the presence of fine particulates with a diameter of approximately 1 μ m (PM_{1.0}) in sea anemones, which are released from fossil fuel combustion into the air. These marine organisms are suspension/filter feeders and, therefore, can incorporate and accumulate suspended particles, including PM_{1.0} particles, in their bodies. The same research team reported that the bioaccumulation factor of $PM_{1.0}$ in sea anemones was approximately 5–7 orders of magnitude. Furthermore, a maternal transfer of PM_{1.0} was also suggested based on the existence of PM in sea anemone eggs and juveniles, suggesting that fine PM accumulation in marine biota is a long-lasting issue once it occurs [103]. The study of Liu et al. [103] also highlight that the hazardous fine PM may pose a serious risk to other marine organisms via the food web. In another study carried out in six coastal areas from the south of China (i.e., suburban industrial, suburban agricultural, and four urban areas), PM_{2.5} samples were collected and used as a simulation of wet deposition scenarios for exposure to the marine fish, the medaka Oryzias melastigma. Significant changes were observed in the fish gut microbiome diversity and lipid metabolism, consequently inducing a decrease in medaka growth [104]. It was also concluded that the PM_{2.5} samples from the industrial site with higher concentrations of Zn and Cr exert significant impacts than PM_{2.5} from the agricultural location that exhibit lower Zn and Cr concentrations [104].

Coral reefs are another example of a key marine ecosystem whose decline can be also exacerbated by the deposition of African dust [38]. In order to study the impact of Sahara dust deposition in the coral diseases at the Caribbean Sea, Hunter et al. [38] applied an association rule data mining (ARDM) algorithm alongside a combination of satellite remote-sensing and in situ data to establish a relationship between Saharan dust storms, Caribbean climate, and the prevalence of coral diseases. It was reported that the ARDM algorithm indicated a relationship between the African dust and coral disease observations in the Caribbean Sea, modulated by the sea surface temperature. The findings of Hunter et al. [38] support the assumption that African dust plays an important role in coral reef diseases alongside the sea surface temperature. This study also reported that the diffuse attenuation coefficient at 490 nm (K490), which is an important water property related to light penetration and availability in aquatic systems, and the Chl-a are not statistically significant parameters concerning the coral disease observations. It has been shown, however, that both Chl-a and K490 values are marginally influenced by the concentration of UV-absorbing aerosols [38]. Nutrient (N and P) enrichment in oceans can indirectly compromise coral health mostly by increasing the incidence and severity of coral diseases (e.g., white spot disease, white pox, aspergillosis) and predation susceptibility, affecting the population dynamics and ultimately causing corals mass extinction [75]. Therefore, all scenarios of increasing soluble forms of nutrients in worldwide oceans pose serious and real risks for the marine ecosystems [75], since corals are amongst the most ecologically and economically relevant marine ecosystems [105].

5. Summary and Outlook

In the preceding sections, this feature paper has outlined how atmospheric deposition of aerosols can have a significant impact on the biogeochemical cycles of several important nutrients in the ocean, namely Fe, N, and P. By bringing new nutrients to the upper ocean, aerosols deposition exerts contrasting and poorly constrained effects on ocean productivity, particularly in low-nutrient low-chlorophyll waters, such as in the case of the Mediterranean Sea. Understanding the long-range transport, deposition (wet and dry), settling, and chemical fate of mineral dust has been a priority, mostly because mineral dust is a dominant source of Fe and P to the open ocean. Nevertheless, atmospheric deposition of anthropogenic N and P is an increasingly important new, and yet poorly constrained, source of these nutrients to the ocean, with impacts on phytoplankton productivity. Considering how anthropogenic activities are modifying the concentration and chemical composition of aerosols, future research efforts should underpin the significance of the atmospheric deposition of anthropogenic N and P on the upper ocean biogeochemistry. Another open question is that the bioavailability of these nutrients is usually associated with their dissolved inorganic forms, often disregarding the fact that soluble organic forms of N or P can potentially also be considered bioavailable (e.g., Bikkina et al. [7] and Okin et al. [106]). Further efforts should explore the evidence for, and implications of, the deposition of organic forms of N and P in surface waters.

Atmospheric deposition is also an important pathway providing inputs of OC and BC to the ocean, with consequences for global carbon reservoirs. The increase in anthropogenic OC and BC emissions will be likely reflected in the amount and composition of these two carbonaceous components deposited in the upper ocean, with expected variable effects on biogeochemical cycles. For example, atmospheric OC binding ligands are likely to promote dust Fe solubility in seawater [70,107]. Furthermore, it has been shown that anthropogenic aerosol Fe is likely to be an important Fe source to the upper ocean and, therefore, could impact phytoplankton growth in the global oceans [73]. These types of evidence suggest that future research should consider the deposition of atmospheric OC/BC in conjunction with the deposition of aerosol Fe from anthropogenic sources and how these aerosol components could affect phytoplankton productivity in the ocean. Similarly, future research should also assess whether the atmospheric inputs of anthropogenic trace metals, such as Cu, Ni, Zn, or Cd, could be mediated by specific atmospheric organic aerosol-binding ligands, rendering them soluble, and thus bioavailable, in seawater. Furthermore, in the case of Fe, the quantity and nature of marine dissolved organic complexing ligands can also control the solubilization of atmospheric Fe in seawater [108]. In this milieu, it has become clear that the presence of specific organic matter ligands is an important factor that controls the solubility of atmospheric nutrients and trace metals, making marine DOM a key regulator of the post-deposition processing of aerosols. We can then assume that atmospheric and oceanic "organic factors", whilst independent from each other, together could control the overall atmospheric nutrients and trace metals dissolution in seawater. However, the molecular composition of those "organic factors" and the extent to which they exert a biogeochemical effect is yet to be fully explored. Other open questions remain to be answered and include: how does the input of "fresh" organic aerosols change the properties (e.g., optical properties) of the upper ocean? Does organic aerosol input make the existing DOM more labile or reactive, or does it just add a potentially more reactive fraction to the existing DOM pool? Addressing these questions is not straightforward, since the impact of organic aerosols inputs is controlled by different, and yet poorly understood, factors (several probably interconnected), namely atmospheric aerosol sources and its organic and inorganic composition, (photo)chemical processing, deposition processes onto the ocean surface, the ocean's degree of oligotrophy, and marine DOM composition. Our understanding of the effects of aerosol OC and BC on marine DOM dynamics is unclear and will require more detailed knowledge on the composition and fate of organic aerosols and marine DOM.

Finally, the existing literature also highlights examples of the negative effects of anthropogenic aerosols deposition in the composition of the oceanic phytoplankton community. These negative effects are likely related to the presence of toxic elements (e.g., Cu) in the aerosols, although ingestion, bioaccumulation, and transfer within trophic chains of atmospheric PM by marine animals cannot be disregarded. Yet, further research is necessary to better understand and predict the impact of anthropogenic aerosol deposition onto the upper ocean by integrating detailed and specific aerosol composition and their variable effects on the marine biota.

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References

- 1. Pöschl, U. Atmospheric Aerosols: Composition, Transformation, Climate and Health Effects. *Angew. Chem. Int. Ed.* 2005, 44, 7520–7540. [CrossRef]
- Tomasi, C.; Lupi, A. Primary and Secondary Sources of Atmospheric Aerosol. In *Atmospheric Aerosols*; Wiley-VCH Verlag GmbH & Co. KGaA: Weinheim, Germany, 2016; pp. 1–86.
- 3. Heal, M.R.; Kumar, P.; Harrison, R.M. Particles, air quality, policy and health. Chem. Soc. Rev. 2012, 41, 6606. [CrossRef] [PubMed]
- 4. Putaud, J.P.; Van Dingenen, R.; Alastuey, A.; Bauer, H.; Birmili, W.; Cyrys, J.; Flentje, H.; Fuzzi, S.; Gehrig, R.; Hansson, H.C.; et al. A European aerosol phenomenology—3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. *Atmos. Environ.* **2010**, *44*, 1308–1320. [CrossRef]
- Duarte, R.M.B.O.; Matos, J.T.V.; Paula, A.S.; Lopes, S.P.; Ribeiro, S.; Santos, J.F.; Patinha, C.; da Silva, E.F.; Soares, R.; Duarte, A.C. Tracing of aerosol sources in an urban environment using chemical, Sr isotope, and mineralogical characterization. *Environ. Sci. Pollut. Res.* 2017, 24, 11006–11016. [CrossRef] [PubMed]
- Duarte, A.C.; Duarte, R.M.B.O. Natural Organic Matter in Atmospheric Particles. In *Biophysico-Chemical Processes Involving Natural Nonliving Organic Matter in Environmental Systems*; Senesi, N., Xing, B., Huang, P.M., Eds.; John Wiley & Sons, Inc.: Hoboken, NJ, USA, 2009; pp. 451–485, ISBN 9780470413005.
- Bikkina, P.; Sarma, V.V.S.S.; Kawamura, K.; Bikkina, S. Dry-deposition of inorganic and organic nitrogen aerosols to the Arabian Sea: Sources, transport and biogeochemical significance in surface waters. *Mar. Chem.* 2021, 231, 103938. [CrossRef]
- Almeida, G.P.; Bittencourt, A.T.; Evangelista, M.S.; Vieira-Filho, M.S.; Fornaro, A. Characterization of aerosol chemical composition from urban pollution in Brazil and its possible impacts on the aerosol hygroscopicity and size distribution. *Atmos. Environ.* 2019, 202, 149–159. [CrossRef]
- Saarikoski, S.; Reyes, F.; Vázquez, Y.; Tagle, M.; Timonen, H.; Aurela, M.; Carbone, S.; Worsnop, D.R.; Hillamo, R.; Oyola, P. Characterization of submicron aerosol chemical composition and sources in the coastal area of Central Chile. *Atmos. Environ.* 2019, 199, 391–401. [CrossRef]
- 10. Emerson, E.W.; Katich, J.M.; Schwarz, J.P.; McMeeking, G.R.; Farmer, D.K. Direct Measurements of Dry and Wet Deposition of Black Carbon Over a Grassland. *J. Geophys. Res. Atmos.* **2018**, *123*, 12277–12290. [CrossRef]
- Emerson, E.W.; Hodshire, A.L.; DeBolt, H.M.; Bilsback, K.R.; Pierce, J.R.; McMeeking, G.R.; Farmer, D.K. Revisiting particle dry deposition and its role in radiative effect estimates. *Proc. Natl. Acad. Sci. USA* 2020, 117, 26076–26082. [CrossRef] [PubMed]

- De Leeuw, G.; Guieu, C.; Arneth, A.; Bellouin, N.; Bopp, L.; Boyd, P.W.; van der Gon, H.A.C.D.; Desboeufs, K.V.; Dulac, F.; Facchini, M.C.; et al. Ocean–Atmosphere Interactions of Particles. In *Ocean-Atmosphere Interactions of Gases and Particles*; Liss, P.C., Johnson, M.T., Eds.; Springer: Berlin/Heidelberg, Germany, 2014; pp. 171–246, ISBN 9783642256431.
- Marañén, E.; Fernández, A.; Mouriño-Carballido, B.; MartÍnez-GarcÍa, S.; Teira, E.; Cermeño, P.; Chouciño, P.; Huete-Ortega, M.; Fernández, E.; Calvo-DÍaz, A.; et al. Degree of oligotrophy controls the response of microbial plankton to Saharan dust. *Limnol. Oceanogr.* 2010, 55, 2339–2352. [CrossRef]
- 14. Paytan, A.; Mackey, K.R.M.; Chen, Y.; Lima, I.D.; Doney, S.C.; Mahowald, N.; Labiosa, R.; Post, A.F. Toxicity of atmospheric aerosols on marine phytoplankton. *Proc. Natl. Acad. Sci. USA* 2009, *106*, 4601–4605. [CrossRef]
- Doney, S.C.; Mahowald, N.; Lima, I.; Feely, R.A.; Mackenzie, F.T.; Lamarque, J.F.; Rasch, P.J. Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and the inorganic carbon system. *Proc. Natl. Acad. Sci. USA* 2007, 104, 14580–14585. [CrossRef] [PubMed]
- Bao, H.; Niggemann, J.; Luo, L.; Dittmar, T.; Kao, S.J. Aerosols as a source of dissolved black carbon to the ocean. *Nat. Commun.* 2017, 8. [CrossRef] [PubMed]
- 17. Jurado, E.; Dachs, J.; Duarte, C.M.; Simó, R. Atmospheric deposition of organic and black carbon to the global oceans. *Atmos. Environ.* **2008**, *42*, 7931–7939. [CrossRef]
- 18. Ginoux, P.; Prospero, J.M.; Gill, T.E.; Hsu, N.C.; Zhao, M. Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products. *Rev. Geophys.* **2012**, *50*, 3005. [CrossRef]
- Schulz, M.; Prospero, J.M.; Baker, A.R.; Dentener, F.; Ickes, L.; Liss, P.S.; Mahowald, N.M.; Nickovic, S.; García-Pando, C.P.; Rodríguez, S.; et al. Atmospheric Transport and Deposition of Mineral Dust to the Ocean: Implications for Research Needs. *Environ. Sci. Technol.* 2012, *46*, 10390–10404. [CrossRef]
- 20. Kienast, S.S.; Winckler, G.; Lippold, J.; Albani, S.; Mahowald, N.M. Tracing dust input to the global ocean using thorium isotopes in marine sediments: ThoroMap. *Glob. Biogeochem. Cycles* **2016**, *30*, 1526–1541. [CrossRef]
- 21. Hayes, C.T.; Anderson, R.F.; Fleisher, M.Q.; Serno, S.; Winckler, G.; Gersonde, R. Quantifying lithogenic inputs to the North Pacific Ocean using the long-lived thorium isotopes. *Earth Planet. Sci. Lett.* **2013**, *383*, 16–25. [CrossRef]
- 22. Hsieh, Y.T.; Henderson, G.M.; Thomas, A.L. Combining seawater 232Th and 230Th concentrations to determine dust fluxes to the surface ocean. *Earth Planet. Sci. Lett.* 2011, 312, 280–290. [CrossRef]
- 23. Anderson, R.F.; Cheng, H.; Edwards, R.L.; Fleisher, M.Q.; Hayes, C.T.; Huang, K.F.; Kadko, D.; Lam, P.J.; Landing, W.M.; Lao, Y.; et al. How well can we quantify dust deposition to the ocean? *Philos. Trans. R. Soc. A Math. Phys. Eng. Sci.* 2016, 374. [CrossRef]
- 24. Hayes, C.T.; Fitzsimmons, J.N.; Boyle, E.A.; McGee, D.; Anderson, R.F.; Weisend, R.; Morton, P.L. Thorium isotopes tracing the iron cycle at the Hawaii Ocean Time-series Station ALOHA. *Geochim. Cosmochim. Acta* 2015, *169*, 1–16. [CrossRef]
- 25. Kadko, D.; Landing, W.M.; Shelley, R.U. A novel tracer technique to quantify the atmospheric flux of trace elements to remote ocean regions. *J. Geophys. Res. Oceans* **2015**, *120*, 848–858. [CrossRef]
- Kadko, D.; Prospero, J. Deposition of ⁷ Be to Bermuda and the regional ocean: Environmental factors affecting estimates of atmospheric flux to the ocean. J. Geophys. Res. 2011, 116, C02013. [CrossRef]
- Shelley, R.U.; Castrillejo, M.; Sanial, V.; Masqué, P.; Landing, W.M.; Van Beek, P.; Planquette, H.; Sarthou, G. Quantification of trace element atmospheric deposition fluxes to the Atlantic Ocean (>40 °N; GEOVIDE, GEOTRACES GA01) during spring 2014. *Deep-Sea Res. Part I Oceanogr. Res. Pap.* 2017, 119, 34–49. [CrossRef]
- Chen, S.; Jiang, N.; Huang, J.; Xu, X.; Zhang, H.; Zang, Z.; Huang, K.; Xu, X.; Wei, Y.; Guan, X.; et al. Quantifying contributions of natural and anthropogenic dust emission from different climatic regions. *Atmos. Environ.* 2018, 191, 94–104. [CrossRef]
- Guieu, C.; Loÿe-Pilot, M.D.; Benyahya, L.; Dufour, A. Spatial variability of atmospheric fluxes of metals (Al, Fe, Cd, Zn and Pb) and phosphorus over the whole Mediterranean from a one-year monitoring experiment: Biogeochemical implications. *Mar. Chem.* 2010, 120, 164–178. [CrossRef]
- Pulido-Villena, E.; Baudoux, A.-C.; Obernosterer, I.; Landa, M.; Caparros, J.; Catala, P.; Georges, C.; Harmand, J.; Guieu, C. Microbial food web dynamics in response to a Saharan dust event: Results from a mesocosm study in the oligotrophic Mediterranean Sea. *Biogeosciences* 2014, 11, 5607–5619. [CrossRef]
- 31. Ridame, C.; Dekaezemacker, J.; Guieu, C.; Bonnet, S.; L'Helguen, S.; Malien, F. Contrasted Saharan dust events in LNLC environments: Impact on nutrient dynamics and primary production. *Biogeosciences* **2014**, *11*, 4783–4800. [CrossRef]
- 32. Wuttig, K.; Wagener, T.; Bressac, M.; Dammshäuser, A.; Streu, P.; Guieu, C.; Croot, P.L. Impacts of dust deposition on dissolved trace metal concentrations (Mn, Al and Fe) during a mesocosm experiment. *Biogeosciences* **2013**, *10*, 2583–2600. [CrossRef]
- 33. Middleton, N.J.; Goudie, A.S. Saharan dust: Sources and trajectories. Trans. Inst. Br. Geogr. 2001, 26, 165–181. [CrossRef]
- 34. Jickells, T.D.; An, Z.S.; Andersen, K.K.; Baker, A.R.; Bergametti, C.; Brooks, N.; Cao, J.J.; Boyd, P.W.; Duce, R.A.; Hunter, K.A.; et al. Global iron connections between desert dust, ocean biogeochemistry, and climate. *Science* **2005**, *308*, 67–71. [CrossRef]
- 35. Guerzoni, S.; Chester, R.; Dulac, F.; Herut, B.; Loÿe-Pilot, M.D.; Measures, C.; Migon, C.; Molinaroli, E.; Moulin, C.; Rossini, P.; et al. The role of atmospheric deposition in the biogeochemistry of the Mediterranean Sea. In *Proceedings of the Progress in Oceanography*; Pergamon: Oxford, UK, 1999; Volume 44, pp. 147–190.
- 36. Abouchami, W.; Näthe, K.; Kumar, A.; Galer, S.J.G.; Jochum, K.P.; Williams, E.; Horbe, A.M.C.; Rosa, J.W.C.; Balsam, W.; Adams, D.; et al. Geochemical and isotopic characterization of the bodélé depression dust source and implications for transatlantic dust transport to the Amazon basin. *Earth Planet. Sci. Lett.* 2013, *380*, 112–123. [CrossRef]

- Schuerger, A.C.; Smith, D.J.; Griffin, D.W.; Jaffe, D.A.; Wawrik, B.; Burrows, S.M.; Christner, B.C.; Gonzalez-Martin, C.; Lipp, E.K.; Schmale, D.G.; et al. Science questions and knowledge gaps to study microbial transport and survival in Asian and African dust plumes reaching North America. *Aerobiologia* 2018, 34, 425–435. [CrossRef]
- 38. Hunter, H.; Cervone, G. Analysing the influence of African dust storms on the prevalence of coral disease in the Caribbean Sea using remote sensing and association rule data mining. *Int. J. Remote Sens.* **2017**, *38*, 1494–1521. [CrossRef]
- Chuvochina, M.S.; Marie, D.; Chevaillier, S.; Petit, J.-R.; Normand, P.; Alekhina, I.A.; Bulat, S.A. Community Variability of Bacteria in Alpine Snow (Mont Blanc) Containing Saharan Dust Deposition and Their Snow Colonisation Potential. *Microbes Environ*. 2011, 26, 237–247. [CrossRef] [PubMed]
- 40. Wang, Q.; Gu, J.; Wang, X. The impact of Sahara dust on air quality and public health in European countries. *Atmos. Environ.* **2020**, 241, 117771. [CrossRef]
- Franchy, G.; Ojeda, A.; López-Cancio, J.; Hernández-León, S. Plankton community response to Saharan dust fertilization in subtropical waters off the Canary Islands. *Biogeosciences Discuss.* 2013, 10, 17275–17307. [CrossRef]
- 42. Kandler, K.; Benker, N.; Bundke, U.; Cuevas, E.; Ebert, M.; Knippertz, P.; Rodríguez, S.; Schütz, L.; Weinbruch, S. Chemical composition and complex refractive index of Saharan Mineral Dust at Izaña, Tenerife (Spain) derived by electron microscopy. *Atmos. Environ.* **2007**, *41*, 8058–8074. [CrossRef]
- 43. Kandler, K.; Schütz, L.; Jäckel, S.; Lieke, K.; Emmel, C.; Müller-Ebert, D.; Ebert, M.; Scheuvens, D.; Schladitz, A.; Šegvić, B.; et al. Ground-based off-line aerosol measurements at Praia, Cape Verde, during the Saharan Mineral Dust Experiment: Microphysical properties and mineralogy. *Tellus Ser. B Chem. Phys. Meteorol.* 2011, 63, 459–474. [CrossRef]
- Lieke, K.; Kandler, K.; Scheuvens, D.; Emmel, C.; Von Glahn, C.; Petzold, A.; Weinzierl, B.; Veira, A.; Ebert, M.; Weinbruch, S.; et al. Particle chemical properties in the vertical column based on aircraft observations in the vicinity of Cape Verde Islands. *Tellus Ser. B Chem. Phys. Meteorol.* 2011, 63, 497–511. [CrossRef]
- 45. Rodriguez-Navarro, C.; di Lorenzo, F.; Elert, K. Mineralogy and physicochemical features of Saharan dust wet deposited in the Iberian Peninsula during an extreme red rain event. *Atmos. Chem. Phys.* **2018**, *18*, 10089–10122. [CrossRef]
- 46. Martin, J.H.; Gordon, R.M.; Fitzwater, S.E. The case for iron. Limnol. Oceanogr. 1991, 36, 1793–1802. [CrossRef]
- 47. Bressac, M.; Guieu, C. Post-depositional processes: What really happens to new atmospheric iron in the ocean's surface? *Glob. Biogeochem. Cycles* **2013**, *27*, 859–870. [CrossRef]
- 48. Guieu, C.; Dulac, F.; Ridame, C.; Pondaven, P. Introduction to project DUNE, a DUst experiment in a low Nutrient, low chlorophyll Ecosystem. *Biogeosciences* **2014**, *11*, 425–442. [CrossRef]
- 49. Wagener, T.; Guieu, C.; Leblond, N. Effects of dust deposition on iron cycle in the surface Mediterranean Sea: Results from a mesocosm seeding experiment. *Biogeosciences* **2010**, *7*, 3769–3781. [CrossRef]
- 50. Gross, A.; Goren, T.; Pio, C.; Cardoso, J.; Tirosh, O.; Todd, M.C.; Rosenfeld, D.; Weiner, T.; Custódio, D.; Angert, A. Variability in sources and concentrations of saharan dust phosphorus over the Atlantic Ocean. *Environ. Sci. Technol. Lett.* **2015**, *2*, 31–37. [CrossRef]
- 51. Louis, J.; Bressac, M.; Pedrotti, M.L.; Guieu, C. Dissolved inorganic nitrogen and phosphorus dynamics in seawater following an artificial Saharan dust deposition event. *Front. Mar. Sci.* 2015, 2, 27. [CrossRef]
- van der Does, M.; Knippertz, P.; Zschenderlein, P.; Giles Harrison, R.; Stuut, J.B.W. The mysterious long-range transport of giant mineral dust particles. *Sci. Adv.* 2018, 4, 1–9. [CrossRef] [PubMed]
- 53. Maki, T.; Ishikawa, A.; Mastunaga, T.; Pointing, S.B.; Saito, Y.; Kasai, T.; Watanabe, K.; Aoki, K.; Horiuchi, A.; Lee, K.C.; et al. Atmospheric aerosol deposition influences marine microbial communities in oligotrophic surface waters of the western Pacific Ocean. *Deep-Sea Res. Part I Oceanogr. Res. Pap.* **2016**, *118*, 37–45. [CrossRef]
- 54. Baars, H.; Ansmann, A.; Althausen, D.; Engelmann, R.; Artaxo, P.; Pauliquevis, T.; Souza, R. Further evidence for significant smoke transport from Africa to Amazonia. *Geophys. Res. Lett.* **2011**, *38*. [CrossRef]
- 55. Amiri-Farahani, A.; Allen, J.R.; Neubauer, D.; Lohmann, U. Impact of Saharan dust on North Atlantic marine stratocumulus clouds: Importance of the semidirect effect. *Atmos. Chem. Phys.* **2017**, *17*, 6305–6322. [CrossRef]
- Karydis, V.A.; Tsimpidi, A.P.; Bacer, S.; Pozzer, A.; Nenes, A.; Lelieveld, J. Global impact of mineral dust on cloud droplet number concentration. *Atmos. Chem. Phys.* 2017, 17, 5601–5621. [CrossRef]
- 57. Luo, C.; Wang, W.; Sheng, L.; Zhou, Y.; Hu, Z.; Qu, W.; Li, X.; Hai, S. Influence of polluted dust on chlorophyll-a concentration and particulate organic carbon in the subarctic North Pacific Ocean based on satellite observation and the WRF-Chem simulation. *Atmos. Res.* **2020**, *236*, 104812. [CrossRef]
- Bressac, M.; Guieu, C.; Doxaran, D.; Bourrin, F.; Obolensky, G.; Grisoni, J.-M. A mesocosm experiment coupled with optical measurements to assess the fate and sinking of atmospheric particles in clear oligotrophic waters. *Geo. Mar. Lett.* 2012, 32, 153–164. [CrossRef]
- 59. Desboeufs, K.; Leblond, N.; Wagener, T.; Bon Nguyen, E.; Guieu, C. Chemical fate and settling of mineral dust in surface seawater after atmospheric deposition observed from dust seeding experiments in large mesocosms. *Biogeosciences* **2014**, *11*, 5581–5594. [CrossRef]
- 60. Bressac, M.; Guieu, C.; Doxaran, D.; Bourrin, F.; Desboeufs, K.; Leblond, N.; Ridame, C. Quantification of the lithogenic carbon pump following a simulated dust-deposition event in large mesocosms. *Biogeosciences* **2014**, *11*, 1007–1020. [CrossRef]
- 61. Prospero, J.M.; Landing, W.M.; Schulz, M. African dust deposition to Florida: Temporal and spatial variability and comparisons to models. *J. Geophys. Res. Atmos.* 2010, 115. [CrossRef]

- 62. Prospero, J.M.; Mayol-Bracero, O.L. Understanding the transport and impact of African dust on the Caribbean Basin. *Bull. Am. Meteorol. Soc.* **2013**, *94*, 1329–1337. [CrossRef]
- 63. Philip, S.; Martin, R.V.; Snider, G.; Weagle, C.L.; Van Donkelaar, A.; Brauer, M.; Henze, D.K.; Klimont, Z.; Venkataraman, C.; Guttikunda, S.K.; et al. Anthropogenic fugitive, combustion and industrial dust is a significant, underrepresented fine particulate matter source in global atmospheric models. *Environ. Res. Lett.* **2017**, *12*, 044018. [CrossRef]
- 64. Fu, J.; Wang, B.; Chen, Y.; Ma, Q. The influence of continental air masses on the aerosols and nutrients deposition over the western North Pacific. *Atmos. Environ.* **2018**, *172*, 1–11. [CrossRef]
- 65. Kim, I.N.; Lee, K.; Gruber, N.; Karl, D.M.; Bullister, J.L.; Yang, S.; Kim, T.W. Increasing anthropogenic nitrogen in the North Pacific Ocean. *Science* 2014, 346, 1102–1106. [CrossRef]
- 66. Jickells, T.D.; Buitenhuis, E.; Altieri, K.; Baker, A.R.; Capone, D.; Duce, R.A.; Dentener, F.; Fennel, K.; Kanakidou, M.; LaRoche, J.; et al. A reevaluation of the magnitude and impacts of anthropogenic atmospheric nitrogen inputs on the ocean. *Glob. Biogeochem. Cycles* **2017**, *31*, 289–305. [CrossRef]
- 67. Penuelas, J.; Janssens, I.A.; Ciais, P.; Obersteiner, M.; Sardans, J. Anthropogenic global shifts in biospheric N and P concentrations and ratios and their impacts on biodiversity, ecosystem productivity, food security, and human health. *Glob. Chang. Biol.* **2020**, *26*, 1962–1985. [CrossRef] [PubMed]
- 68. Duce, R.A.; LaRoche, J.; Altieri, K.; Arrigo, K.R.; Baker, A.R.; Capone, D.G.; Cornell, S.; Dentener, F.; Galloway, J.; Ganeshram, R.S.; et al. Impacts of atmospheric anthropogenic nitrogen on the open ocean. *Science* **2008**, *320*, 893–897. [CrossRef]
- 69. Zhu, Y.; Li, W.; Lin, Q.; Yuan, Q.; Liu, L.; Zhang, J.; Zhang, Y.; Shao, L.; Niu, H.; Yang, S.; et al. Iron solubility in fine particles associated with secondary acidic aerosols in east China. *Environ. Pollut.* **2020**, *264*, 114769. [CrossRef] [PubMed]
- López-García, P.; Gelado-Caballero, M.D.; Patey, M.D.; Hernández-Brito, J.J. Atmospheric fluxes of soluble nutrients and Fe: More than three years of wet and dry deposition measurements at Gran Canaria (Canary Islands). *Atmos. Environ.* 2021, 246, 118090. [CrossRef]
- 71. Marcotte, A.R.; Anbar, A.D.; Majestic, B.J.; Herckes, P. Mineral Dust and Iron Solubility: Effects of Composition, Particle Size, and Surface Area. *Atmosphere* **2020**, *11*, 533. [CrossRef]
- Conway, T.M.; Hamilton, D.S.; Shelley, R.U.; Aguilar-Islas, A.M.; Landing, W.M.; Mahowald, N.M.; John, S.G. Tracing and constraining anthropogenic aerosol iron fluxes to the North Atlantic Ocean using iron isotopes. *Nat. Commun.* 2019, 10, 1–10. [CrossRef] [PubMed]
- Pinedo-González, P.; Hawco, N.J.; Bundy, R.M.; Virginia Armbrust, E.; Follows, M.J.; Cael, B.B.; White, A.E.; Ferrón, S.; Karl, D.M.; John, S.G. Anthropogenic Asian aerosols provide Fe to the North Pacific Ocean. *Proc. Natl. Acad. Sci. USA* 2020, 117, 27862–27868. [CrossRef]
- Mahowald, N.; Jickells, T.D.; Baker, A.R.; Artaxo, P.; Benitez-Nelson, C.R.; Bergametti, G.; Bond, T.C.; Chen, Y.; Cohen, D.D.; Herut, B.; et al. Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts. *Glob. Biogeochem. Cycles* 2008, 22. [CrossRef]
- 75. Zhao, H.; Yuan, M.; Strokal, M.; Wu, H.C.; Liu, X.; Murk, A.; Kroeze, C.; Osinga, R. Impacts of nitrogen pollution on corals in the context of global climate change and potential strategies to conserve coral reefs. *Sci. Total Environ.* 2021, 774, 145017. [CrossRef]
- Yu, L.; Ma, X.; Gao, H.; Zong, H.; Yao, X.; Lin, Z.; Zhang, Z.; Zhang, C.; Yao, X.; Zhang, Z. Distribution and source identification of nitrogen and phosphorus in aerosols in the Qinhuangdao coast, north China. *Atmos. Environ.* 2020, 234, 117475. [CrossRef]
- 77. Yoshizue, M.; Iwamoto, Y.; Adachi, K.; Kato, S.; Sun, S.; Miura, K.; Uematsu, M. Individual particle analysis of marine aerosols collected during the North–South transect cruise in the Pacific Ocean and its marginal seas. *J. Oceanogr.* **2019**, *75*, 513–524. [CrossRef]
- 78. Witkowska, A.; Lewandowska, A.; Falkowska, L.M. Parallel measurements of organic and elemental carbon dry (PM1, PM2.5) and wet (rain, snow, mixed) deposition into the Baltic Sea. *Mar. Pollut. Bull.* **2016**, *104*, 303–312. [CrossRef] [PubMed]
- 79. Decesari, S.; Facchini, M.C.; Matta, E.; Mircea, M.; Fuzzi, S.; Chughtai, A.R.; Smith, D.M. Water soluble organic compounds formed by oxidation of soot. *Atmos. Environ.* **2002**, *36*, 1827–1832. [CrossRef]
- Mead, R.N.; Mullaugh, K.M.; Brooks Avery, G.; Kieber, R.J.; Willey, J.D.; Podgorski, D.C. Insights into dissolved organic matter complexity in rainwater from continental and coastal storms by ultrahigh resolution Fourier transform ion cyclotron resonance mass spectrometry. *Atmos. Chem. Phys.* 2013, *13*, 4829–4838. [CrossRef]
- Mari, X.; Guinot, B.; Van Thuoc, C.; Brune, J.; Lefebvre, J.; Ram, P.; Sriram, A.; Raimbault, P.; Dittmar, T.; Niggemann, J. Biogeochemical Impacts of a Black Carbon Wet Deposition Event in Halong Bay, Vietnam. *Front. Mar. Sci.* 2019, *6*, 1–18. [CrossRef]
- Doherty, S.J.; Warren, S.G.; Grenfell, T.C.; Clarke, A.D.; Brandt, R.E. Light-absorbing impurities in Arctic snow. *Atmos. Chem. Phys.* 2010, 10, 11647–11680. [CrossRef]
- 83. Fang, Z.; Yang, W.; Chen, M.; Zheng, M.; Hu, W. Abundance and sinking of particulate black carbon in the western Arctic and Subarctic Oceans. *Sci. Rep.* **2016**, *6*, 1–11. [CrossRef] [PubMed]
- Stubbins, A.; Hood, E.; Raymond, P.A.; Aiken, G.R.; Sleighter, R.L.; Hernes, P.J.; Butman, D.; Hatcher, P.G.; Striegl, R.G.; Schuster, P.; et al. Anthropogenic aerosols as a source of ancient dissolved organic matter in glaciers. *Nat. Geosci.* 2012, *5*, 198–201. [CrossRef]

- Fishwick, M.P.; Ussher, S.J.; Sedwick, P.N.; Lohan, M.C.; Worsfold, P.J.; Buck, K.N.; Church, T.M. Impact of surface ocean conditions and aerosol provenance on the dissolution of aerosol manganese, cobalt, nickel and lead in seawater. *Mar. Chem.* 2018, 198, 28–43. [CrossRef]
- Pinhassi, J.; Gómez-Consarnau, L.; Alonso-Sáez, L.; Sala, M.; Vidal, M.; Pedrós-Alió, C.; Gasol, J. Seasonal changes in bacterioplankton nutrient limitation and their effects on bacterial community composition in the NW Mediterranean Sea. *Aquat. Microb. Ecol.* 2006, 44, 241–252. [CrossRef]
- 87. Gallisai, R.; Peters, F.; Volpe, G.; Basart, S.; Baldasano, J.M. Saharan Dust Deposition May Affect Phytoplankton Growth in the Mediterranean Sea at Ecological Time Scales. *PLoS ONE* **2014**, *9*, e110762. [CrossRef]
- 88. Bristow, L.A.; Mohr, W.; Ahmerkamp, S.; Kuypers, M.M.M. Nutrients that limit growth in the ocean. *Curr. Biol.* 2017, 27, R474–R478. [CrossRef] [PubMed]
- Baker, A.R.; Kelly, S.D.; Biswas, K.F.; Witt, M.; Jickells, T.D. Atmospheric deposition of nutrients to the Atlantic Ocean. *Geophys. Res. Lett.* 2003, 30. [CrossRef]
- 90. Mahowald, N.M.; Baker, A.R.; Bergametti, G.; Brooks, N.; Duce, R.A.; Jickells, T.D.; Kubilay, N.; Prospero, J.M.; Tegen, I. Atmospheric global dust cycle and iron inputs to the ocean. *Glob. Biogeochem. Cycles* **2005**, *19*. [CrossRef]
- Marty, J.C.; Chiavérini, J.; Pizay, M.D.; Avril, B. Seasonal and interannual dynamics of nutrients and phytoplankton pigments in the western Mediterranean Sea at the DYFAMED time-series station (1991–1999). *Deep-Sea Res. Part II Top. Stud. Oceanogr.* 2002, 49, 1965–1985. [CrossRef]
- 92. Pulido-Villena, E.; Rérolle, V.; Guieu, C. Transient fertilizing effect of dust in P-deficient LNLC surface ocean. *Geophys. Res. Lett.* **2010**, *37*. [CrossRef]
- 93. Bosc, E.; Bricaud, A.; Antoine, D. Seasonal and interannual variability in algal biomass and primary production in the Mediterranean Sea, as derived from 4 years of SeaWiFS observations. *Glob. Biogeochem. Cycles* **2004**, *18*. [CrossRef]
- López-Sandoval, D.C.; Fernández, A.; Marañón, E. Dissolved and particulate primary production along a longitudinal gradient in the Mediterranean Sea. *Biogeosciences* 2011, *8*, 815–825. [CrossRef]
- Markaki, Z.; Loÿe-Pilot, M.D.; Violaki, K.; Benyahya, L.; Mihalopoulos, N. Variability of atmospheric deposition of dissolved nitrogen and phosphorus in the Mediterranean and possible link to the anomalous seawater N/P ratio. *Mar. Chem.* 2010, 120, 187–194. [CrossRef]
- 96. Marín-Beltrán, I.; Logue, J.B.; Andersson, A.F.; Peters, F. Atmospheric Deposition Impact on Bacterial Community Composition in the NW Mediterranean. *Front. Microbiol.* **2019**, *10*, 858. [CrossRef] [PubMed]
- 97. Echeveste, P.; Croot, P.; von Dassow, P. Differences in the sensitivity to Cu and ligand production of coastal vs offshore strains of Emiliania huxleyi. *Sci. Total Environ.* **2018**, *625*, 1673–1680. [CrossRef]
- Yang, T.; Chen, Y.; Zhou, S.; Li, H. Impacts of aerosol copper on marine phytoplankton: A review. *Atmosphere* 2019, *10*, 1. [CrossRef]
 Chien, C.-T.; Mackey, K.R.M.; Dutkiewicz, S.; Mahowald, N.M.; Prospero, J.M.; Paytan, A. Effects of African dust deposition on
- phytoplankton in the western tropical Atlantic Ocean off Barbados. Glob. Biogeochem. Cycles 2016, 30, 716–734. [CrossRef]
- 100. Park, G.H.; Lee, S.E.; Kim, Y.-I.; Kim, D.; Lee, K.; Kang, J.; Kim, Y.H.; Kim, H.; Park, S.; Kim, T.W. Atmospheric deposition of anthropogenic inorganic nitrogen in airborne particles and precipitation in the East Sea in the northwestern Pacific Ocean. *Sci. Total Environ.* 2019, 681, 400–412. [CrossRef] [PubMed]
- 101. Duarte, C.M.; Dachs, J.; Llabrés, M.; Alonso-Laita, P.; Gasol, J.M.; Tovar-Sánchez, A.; Sañudo-Wilhemy, S.; Agustí, S. Aerosol inputs enhance new production in the subtropical northeast Atlantic. *J. Geophys. Res. Biogeosciences* **2006**, *111*. [CrossRef]
- 102. Liao, W.-H.; Yang, S.-C.; Ho, T.-Y. Trace metal composition of size-fractionated plankton in the Western Philippine Sea: The impact of anthropogenic aerosol deposition. *Limnol. Oceanogr.* 2017, *62*, 2243–2259. [CrossRef]
- 103. Liu, L.L.; Hsieh, C.Y.; Kuo, M.Y.; Chen, C.; Shau, Y.H.; Lui, H.K.; Yuan, C.S.; Chen, C.T.A. Evidence for Fossil Fuel PM1 Accumulation in Marine Biota. *Environ. Sci. Technol.* **2020**, *54*, 4068–4078. [CrossRef] [PubMed]
- 104. Zhao, P.; Lu, W.; Hong, Y.; Chen, J.; Dong, S.; Huang, Q. Long-term wet precipitation of PM2.5 disturbed the gut microbiome and inhibited the growth of marine medaka Oryzias melastigma. *Sci. Total Environ.* **2021**, *755*, 142512. [CrossRef]
- 105. Lachs, L.; Oñate-Casado, J. Fisheries and Tourism: Social, Economic, and Ecological Trade-offs in Coral Reef Systems. In YOUMARES 9—The Oceans: Our Research, Our Future; Springer: Berling/Heidelberg, Germany, 2020; pp. 243–260.
- 106. Okin, G.S.; Baker, A.R.; Tegen, I.; Mahowald, N.M.; Dentener, F.J.; Duce, R.A.; Galloway, J.N.; Hunter, K.; Kanakidou, M.; Kubilay, N.; et al. Impacts of atmospheric nutrient deposition on marine productivity: Roles of nitrogen, phosphorus, and iron. *Glob. Biogeochem. Cycles* 2011, 25. [CrossRef]
- 107. Paris, R.; Desboeufs, K.V. Effect of atmospheric organic complexation on iron-bearing dust solubility. *Atmos. Chem. Phys.* 2013, 13, 4895–4905. [CrossRef]
- 108. Baker, A.R.; Croot, P.L. Atmospheric and marine controls on aerosol iron solubility in seawater. Mar. Chem. 2010, 120, 4–13. [CrossRef]