CENTER *for* BIOLOGICAL DIVERSITY *Because life is good.*

October 24, 2011

Sent via email and fax

State Water Resources Control Board
1001 I Street, 24th Floor
Sacramento, CA 95814
commentletters@waterboards.ca.gov
Fax: (916) 341-5620

Re: **Comment Letter – California Ocean Plan Amendments**

Dear Chair Hoppin and Board Members:

Thank you for the opportunity to submit comments on the Draft Amendment of the Water Quality Control Plan, Ocean Waters of California, dated August 11, 2011. Please consider the following comments, submitted on behalf of the Center for Biological Diversity:

1. Model monitoring

We support both alternatives 3 and 4 because robust monitoring of ocean water quality is essential for marine ecosystem health. We support minimum standards for monitoring. These monitoring data can and should also be used for water quality assessments.

> Alternative 3: Use a model ocean monitoring approach with minimum requirements.
> Alternative 4: Use a prescriptive approach to all ocean discharges from all sources.

Importantly, we urge the Ocean Plan to require monitoring for ocean acidification and its impacts. It would be inconsistent with the Clean Water Act and EPA's recent recommendations on ocean acidification to omit monitoring for this key threat to marine ecosystems. Because of the long time lag between carbon dioxide pollution and corresponding changes in ocean chemistry, it is necessary to begin to address ocean acidification immediately. For California to protect its designated uses for ocean waters, it should require monitoring for the biological and chemical changes from anthropogenic carbon dioxide pollution.

a. Amend to require monitoring for ocean acidification

With regard to ocean acidification monitoring, California should monitor physical and biological indicators, ecosystem changes and carbon dioxide sources over time. California should conduct water quality monitoring and coordinate those monitoring activities with the EPA and

cooperating federal agencies and research institutions. California's Ocean Plan can and should require monitoring of parameters relevant to ocean acidification by permit holders. It is relevant to require such monitoring of both coastal and ocean areas, as well as carbon dioxide emissions from permitted sources.

Lack of site-specific monitoring data in California waters on ocean acidification has been noted as a barrier to marine water quality assessments. In response to comments on ocean acidification for California's 2010 water quality assessment, California concluded that available information on ocean acidification did not meet the requirements for impaired waters listing policy, which requires that only data and information collected from waters of California to be used for 303(d) listing purposes. EPA affirmed this decision, "In the absence of specific data showing exceedance of the existing marine pH criteria, data showing impairment of California biota due to altered pH, or data demonstrating declining water quality due to acidification, EPA finds CA's omission of ocean acidification from its 303(d) list to be appropriate."

The lack of site-specific monitoring data, however, is no excuse to ignore the problem of ocean acidification. Lack of effort to detect ocean acidification may have serious repercussions. Ocean acidification is irreversible on human timeframes. To avoid the most severe consequences for California's fisheries, endangered species, and coastal economies we must act now to monitor and prevent ocean acidification.

Given the information and data gaps on ocean acidification, there is a need to monitor for both baseline conditions and long-term trends. This could easily attach to ongoing monitoring activities if the correct parameters are measured. Monitoring should include both chemical and biological factors.

At present, the proposed amendment to the Ocean Plan requires certain NPDES dischargers to measure receiving waters seasonally (at minimum fourtimes per year) turbidity, color/chlorophyll, dissolved oxygen (DO), pH, and salinity (at facilities discharging brine). This should be amended to provide simultaneous measurement of temperature, salinity and at least two of the four CO_2 system parameters, which are pH (with the pH scale identified), DIC, TA and pCO_2. Additionally, regional monitoring systems should also be required to gather these data. Moreover, the framework for gathering biological data should contemplate impacts from ocean acidification such as reduction of calcifying species during periods of low pH.

On November 15, 2010, EPA affirmed that states have the authority and duty to identify waters impaired by ocean acidification. In its memorandum EPA "recognizes the seriousness of aquatic life impacts associated with OA [Ocean Acidification] and describes how States can move forward" and instructs that "States should list waters not meeting water quality standards, including marine pH WQS, on their 2012 303(d) lists, and should also solicit existing and readily available information on OA using the current 303(d) listing framework" (Environmental Protection Agency 2010). Specifically, the EPA recommended that States:

(1) request and gather existing data related to ocean acidification, including temperature, salinity, dissolved oxygen, nitrate, total alkalinity, and pH;
(2) develop assessment methods for evaluating impacts of ocean acidification on marine waters based on existing pH and biological water quality criteria;
(3) track the progress of federal efforts to develop assessment and monitoring methods;
(4) development bioassessment methods and/or biocriteria for aquatic resources where data are unavailable; and
(5) prioritize TMDL development for ocean acidification.

In light of these recommendations, amending the model monitoring portion of the Ocean Plan to include ocean acidification parameters is warranted.

i. Ocean acidification

The global oceans have become about 30 percent more acidic since preindustrial times due to anthropogenic carbon dioxide pollution (Hall-Spencer et al. 2008). The best known consequence of ocean acidification is that impairs the ability of calcifying organisms to build their protective shells. Although the worst impacts of ocean acidification are yet to come, we are already experiencing some of the early warning signs along the coast of California. In a 2010 letter to the Environmental Protection Agency, the California Coastal Commission emphasized the importance of using the Clean Water Act to address acidification stating:

> The Coastal Commission fully supports the EPA's use of the Clean Water Act to address ocean acidification. The consequences of ocean acidification and related climate change are grave, and we must use every tool available to us, including the Clean Water Act, to slow down and reverse our contributions to these evolving environmental catastrophes.

(California Coastal Commission 2010: 1).

The California coast is among the most vulnerable to the early effects of ocean acidification because of the unique currents in this region of the Pacific. California Current System is particularly sensitive to ocean acidification with the pH of surface waters comparatively low and change in pH for a given uptake of anthropogenic CO_2 is particularly high (Hauri et al. 2009). Already the aragonite saturation horizon has shoaled by ~100 m and now reaches the euphotic zone in a few eddies and in near-shore environments during upwelling along the Pacific Coast (Hauri et al. 2009). A survey of the Pacific Coast revealed that the effects of ocean acidification are occurring more rapidly there than predicted (Feely et al. 2008). See Figure 1. Researchers found seawater undersaturated with respect to aragonite upwelling onto large portions of the continental shelf, reaching shallow depths of 40 to 120 meters (Feely et al. 2008). The areas where undersaturation occurred at the shallowest depths were off the coast of Northern California. Moreover, as a result marine organisms in surface waters, in the water column, and on the sea floor along the California coast are already being exposed to corrosive water during the upwelling season. According to the study, "the upwelled water off northern California (line

5) was last at the surface about 50 years ago, when atmospheric CO2 was about 65 ppm lower than it is today" (Feely et al. 2008: 1492).

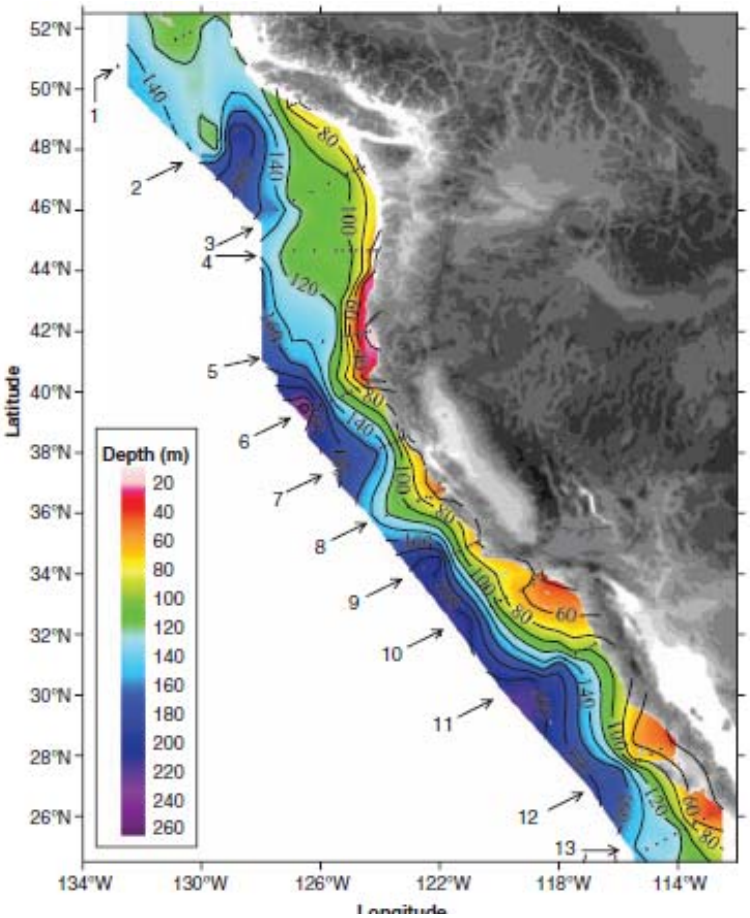

Figure 1. Distribution of the depths of water undersaturated with respect to aragonite on the continental shelf of western North America from Queen Charlotte Sound, Canada, to San Gregorio, Baja California Sur, Mexico. On transect line 5, corrosive water reaches all the way to the surface in inshore waters near the coast. The black dots represent station locations.
Source: Feely et al. (2008): Figure 1.

Coastal estuaries and temperate nearshore ecosystems are among the most biologically productive and maintain some of the most extensive and measurable ecosystem services (*e.g.*, commercial and recreational fisheries, fish and invertebrate nursery grounds, water purification, flood and storm surge protection, human recreation). Because they are shallower, less saline, and have lower alkalinity, these habitats are more susceptible to changes in pH than the open ocean and will likely experience more acute impacts from elevated CO_2 (Miller et al. 2009). A survey of the Puget Sound found corrosive waters that had shifted to being undersaturated with respect to aragonite (Feely et al. 2010). Feely et al. estimated that ocean acidification can account for 24-49 percent of the pH decrease already observed and will account for 49-82 percent of the pH decrease over time when carbon dioxide in the atmosphere reaches 560 ppm (*Id.*). Wootton et

al., studying waters off Tatoosh Island in Washington had experienced a decline in pH in excess of 0.3 units between 2000-2008, meanwhile species shifts occurred in tidepools (Wootton, Pfister, and Forester 2008). The waters in the North Pacific are increasingly lethal to shellfish larvae with recurring breeding failures occurring in oyster production (Cohen 2010; Welch 2009; Miller et al. 2009; Barton, Cudd, and Weigardt 2009). Some oysters in the Pacific Northwest have failed to reproduce for the past six years (Southern California Coastal Water Research Project 2010).

A meta analysis of studies on the biological responses to ocean acidification found that although the responses varied among animals, they were overwhelmingly negative (Kroeker et al. 2010). The review found that ocean acidification had a significant negative effect on survival, calcification, growth and reproduction in marine organisms (*Id*.). Kroeker et al. found that calcifying organisms were the most sensitive to ocean acidification (*Id*.). This shows that while our understanding of ocean acidification would benefit from more research, that we know enough to act now to prevent the worst consequences.

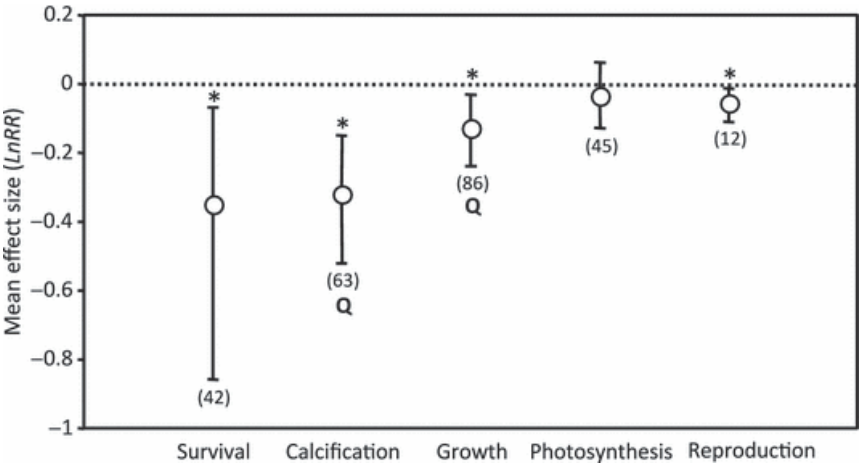

Fig. 2. The effect of near-future (2100) ocean acidification on different response variables of marine organisms from weighted, random effects meta-analyses. The mean and bias-corrected bootstrapped 95% confidence interval are shown for separate analyses of survival, calcification, growth, photosynthesis and reproduction. The number of experiments in each analysis is shown in parentheses. The zero line indicates no effect, and significance of mean effects is determined when the 95% confidence interval does not overlap zero. All responses are significantly negative (*) except for photosynthesis, which shows no effect. There is significant heterogeneity (underlying data structure, denoted with *Q*) within the mean effect for calcification and growth. (Source: Kroeker et al. 2010)

Plankton, which comprise the basis of the marine food web, are among the calcifying organisms likely to be adversely affected by ocean acidification. Studies of coccolithophorids showed that carbon dioxide related changes to seawater caused reduced calcification, resulting in malformed and incomplete shells (Riebesell et al. 2000). Pteropods similarly experience reduced calcification under elevated carbon dioxide levels (Comeau et al. 2009). Experiments also show that the shells of pteropods dissolve as seawater becomes undersaturated with aragonite (Orr et al. 2005). Elevated carbon dioxide concentrations also reduce the shell mass of foraminifera (Kleypas et al. 2005). Modern shell weights of foraminifera in the Southern Ocean are 30–35

percent lower than those from preindustrial sediments, which is consistent with reduced calcification induced by ocean acidification (Moy et al. 2009). While some species of plankton react differently under high concentrations of carbon dioxide, most calcareous plankton studied thus far exhibit reduced calcification (Guinotte and Fabry 2008). Ocean acidification's impact on calcifying plankton is especially troublesome because most of the ocean's primary production is from such plankton and effects will extend up the entire food chain.

The waters in the California Current system are increasingly lethal to shellfish larvae with recurring breeding failures occurring in oyster production in the Pacific Northwest. Most strikingly, oysters in Washington and Oregon have failed to reproduce for the past six years (Southern California Coastal Water Research Project 2010). Pacific Coast oyster hatcheries are already experiencing difficulties associated with increasing ocean acidification. Two of the largest hatcheries, including Taylor Shellfish Farms in Washington, report production rates down by as much as 80% (Miller et al. 2009). The oyster failures in recent years may foreshadow the widespread effects that increasingly acidic waters will have on the shellfishing and fishing industry. Assuming business as usual projections for carbon emissions and a corresponding decline in ocean pH and mollusk harvests, ocean acidification's broader economic losses for the United States would range from $1.5–6.4 billion through 2060 (Cooley et al. 2009).

California mussels are a dominant species in California's rocky intertidal area. The Wootton study in Washington found that calcifying species were replaced by non-calcifying species during years with low pH, this is a sample of what is likely yet to come for California's coasts. Wootton et al. noted significant implications of these findings for shoreline ecosystems:

> *Mytilus californianus* mussel beds are a dominant coastal habitat along the northeastern Pacific, and mussel beds, in general, are an important habitat on most temperate rocky shores (24). These habitats provide food and structure for a diverse array of species in an otherwise physically stressful environment, and the dominant calcareous species provide food resources for humans. Mussels also impact coastal water conditions through their filtering activities and influences on nutrient recycling pathways (25). Compared with other biogenic habitats, such as coral reefs and sea grass beds, evidence of substantial impacts of global change on rocky intertidal habitats has been sparse. This is perhaps because intertidal organisms naturally endure harsh physical fluctuations and are predisposed to tolerate varying environmental conditions. Hence, our results indicating that these robust systems are impacted by changes in pH may portend much broader-scale impacts in other marine habitats.

(Wootton 2008: 18851). A new study on California mussels supports Wootton's findings (Gaylord et al. 2011). Gaylord et al. experimented with larvae of California mussels finding that larvae exposed to conditions of ocean acidification precipitated weaker, thinner, and smaller shells (***Id.)***. Their findings suggested that the impacts of ocean acidification could have adverse impacts on mortality, fitness, with potential population impacts on distribution and abundance (*Id*.).

Ocean acidification also disrupts metabolism and other biological functions in marine life. Changes in the ocean's carbon dioxide concentration result in accumulation of carbon dioxide in the tissues and fluids of fish and other marine animals, called hypercapnia, and increased acidity in the body fluids, called acidosis. These impacts can cause a variety of problems for marine animals including difficulty with acid-base regulation, calcification, growth, respiration, energy turnover, and mode of metabolism (Pörtner, Langenbuch, and Reipschlager 2004). Squid, for example, show a very high sensitivity to pH because of their energy intensive manner of swimming (Pörtner et al. 2004; Royal Society 2005). Because of their energy demand, even under a moderate 0.15 pH change squid have reduced capacity to carry oxygen and higher carbon dioxide pressures are likely to be lethal (Pörtner et al. 2004). Studies have shown that squid under elevated carbon dioxide have a slowed metabolic activity and impaired behaviors, and researchers say warming waters will mean that the oxygen-poor zones the squid inhabit at night will be shallower reducing squid habitat and increasing their vulnerability to predators (Rosa and Seibel 2008). In fish, high concentrations of carbon dioxide in seawater can lead to cardiac failure (Ishimatsu et al. 2004).

Some studies show that juvenile marine organisms are particularly susceptible to ocean acidification (Ishimatsu et al. 2004; Kurihara and Shirayama 2004). Increased rates of CO_2 are reported to have had a pronounced negative effect on the survival of shellfish larvae, which in turn dramatically reduces the adult population (Talmage 2009). In conditions simulating future seawater with elevated carbon dioxide, larval clownfish lost their detection and homing abilities to find suitable habitat (Munday et al. 2009).

Ocean acidification can also decrease the sound absorption of seawater causing sounds to travel further with potential impacts on marine mammals and other marine life that may be sensitive to noise of vessel traffic, seismic surveys, and other noise pollution (Hester et al. 2008). Already sound travels 10-15 percent further with a change of 0.1 pH, and it is predicted to increase about 40 percent by mid century with corresponding ocean acidification (Hester et al. 2008). Moreover, ocean acidification may also enhance the mobility of mercury in the environment resulting in increased accumulation of mercury in fish, marine mammals, and humans (USGS 2000).

Although the specific ecosystem responses to ocean acidification are complex and not yet well understood, the rapid change in ocean chemistry means that wildlife has very little time to adapt. Monitoring the present status of marine ecosystems and how they change in response to carbon dioxide will be an important step in protecting and managing water quality and California's designated uses.

ii. Best protocols for ocean acidification chemistry monitoring

Ocean acidification concerns a broad range of changes to ocean chemistry including declining pH and saturation states of aragonite and calcium carbonate, all of which can directly affect marine biological processes. Accordingly, monitoring for ocean acidification should reflect these

various factors. The Ocean Carbon Biogeochemistry Program provided these primary recommendations to EPA concerning monitoring:

> a) Application of a pH criterion alone will be inadequate to monitor ocean acidification and its impacts on coastal marine ecosystems. It is therefore recommended that additional criteria such as carbonate ion concentration and saturation state be considered.
> b) Direct measurements of pH using spectrophotometric indicators are the most reliable and straightforward method for quantifying the changes in $[H^+]$ due to ocean acidification. Thus far, however, these measurements have limited spatial and temporal coverage.
> c) Full characterization of the seawater inorganic chemistry system and ocean acidification requires simultaneous measurement of temperature, salinity and at least two of the four CO_2 system parameters (pH, DIC, TA and pCO_2). The pH scale used must also be reported.

(Ocean Carbon and Biogeochemistry Program 2010: 9). Another excellent source for monitoring protocols is:

Dickson AG, Sabine CL, Christian JR (Eds) (2007) Guide to best practices for ocean CO_2 measurements, PICES Special Publication, 3, 191 pp., 2007 (available at http://cdiac.ornl.gov/oceans/Handbook_2007.html).

iii. Biological monitoring and biocriteria

California should require biological monitoring for responses to ocean acidification. Studies show a range of responses that can be grouped generally into those relating to survival, growth, and reproduction (OCB 2009; Ocean Carbon and Biogeochemistry Program 2010; Kroeker et al. 2010). Calcification and abundance and distribution of calcifying organisms should be measured along side the chemical conditions of seawater. The biological data paired with chemical data is essential to attribute the responses to ocean acidification (Ocean Carbon and Biogeochemistry Program 2010).

The use of biological criteria should also be fully utilized as a supplement to the numeric criteria that can provide a measure against which to evaluate ocean acidification and its impacts on aquatic life. As early as 1990, EPA provided guidance to states to develop biological information and criteria (EPA 1990). Biological criteria are numeric and narrative criteria that define the condition of an aquatic community such as its species richness, presence or absence of indicator taxa, distribution of classes of organisms (*Id.*). Biological assessments and biological criteria can then be used as a measure for determining the impacts of ocean acidification.

Although underutilized, biological criteria can significantly contribute to water quality monitoring and improve our understanding of pollutants on the marine ecosystem. The EPA recently released technical guidance on coral reef biological criteria. (EPA, Coral Reef Biological Criteria: Using the Clean Water Act to Protect a National Treasure, EPA/600/R-

10/054, July 2010.) In it, EPA acknowledges that the Clean Water Act can be used to address ocean acidification by assessing waterbody impairment caused by ocean acidification which may trigger action under the Clean Air Act. It also noted that biocriteria can be applied to specifically protect Endangered Species Act listed species.

Accordingly, biological monitoring and criteria are both components of monitoring ocean acidification.

b. Monitoring for plastic pollution

Plastic pollution has many ecosystem impacts including entanglement of and ingestion by marine wildlife, transfer of persistent organic pollutants and other contaminants, and transport of invasive species (OST 2011). The sum of available research suggests that plastics are a pollutant that California should regulate. Monitoring should begin now in order to collect data necessary to effectively protect California's ocean.

i. Coastal marine debris data

Coastal Cleanup Day statistics currently provide the most comprehensive long-term data set regarding the quantity and types of marine debris, including plastics, found on California coast. The California Coastal Commission organizes the volunteers and waste removal for the annual event and submits data to the Ocean Conservancy's international database. Although monitoring is occurring on this annual volunteer-led basis, California should institute standard marine debris monitoring at sources of discharge. All permitted dischargers should report observations of marine debris in receiving water bodies.

The question-driven monitoring framework is easily suited to addressing marine debris, yet the Draft Amendment to the Ocean Plan does not include questions targeted at marine debris. As soon as possible, California should establish a trash objective of "zero" in the Ocean Plan in order to comply with water quality standards in the Clean Water Act and guarantee protection of the beneficial uses of the ocean environment. Sufficient monitoring data is critical in order to determine whether water bodies are impaired. Therefore, California should include questions designed to monitor marine debris in the section "10. Receiving Water Characteristics." Tailoring the questions to be similar to the coastal cleanup data sheets will allow comparison to observations by dischargers.

ii. Microplastics

While some Regional Water Quality Control Boards have sought to meet water quality objectives regarding floating material and solid, suspended or settleable materials[1] by

[1] For example, the Los Angeles Region Water Quality Control Plan includes the following objectives: (1) "Waters shall not contain floating materials including solids, liquids, foams, and scum in concentrations that cause nuisance or adversely affect beneficial use" and (2) "Waters shall not contain suspended or settable material in concentrations that cause nuisance or adversely affect beneficial uses."

implementing trash total maximum daily loads (TMDLs), microplastics have not yet been addressed as a pollutant. To meet current trash TMDLs, captures systems like trash nets, mesh screens, vortex separation systems, and other devices filter out particles 5 mm and larger. Microplastics are smaller than this, by some definitions less than 1 mm, and so are not addressed by these filter systems.

Recent research has observed the ubiquitous nature of microplastics – on shorelines of six continents – and concluded that a large proportion derive from sewage (Browne et al. 2011). Ingestion of microplastic provides a potential pathway for the transfer of pollutants, monomers and plastic-additives to wildlife and humans (*Id.*). Browne et al. found plastic fibers in the same proportion of polyester and acrylic fibers used in clothing. Experiments sampling wastewater from domestic washing machines demonstrated a single garment can product > 1900 fibers per wash (*Id.*). While more research is needed on the effects of ingested microplastics, the scientists note the potential for release of monomers (*e.g.*, ethylene glycol, dimethyl terephthalate, propenenitrile, acrylonitrile, acrylonitrile, vinyl chloride, vinylidene chloride, vinyl bromide), dispersive dyes, mordants (*e.g.*, aluminum, chromium, copper, potassium, tin), plasticisers from manufacture and sorbed contaminants from sewage (*e.g.*, organotin, nonylphenol, and Triclosan (*Id.*).

Given the concern over the effects of microplastics in the environment, California should begin monitoring discharges for microplastics. Key to controlling pollutants is understanding the sources. Based on the research that a large proportion of microplastic pollution may come from point sources, California should start monitoring these sources for microplastics.

2. Conclusion

Thank you for consideration of these comments. Please feel free to contact me with questions.

Sincerely,

/s/ *Miyoko Sakashita*
Miyoko Sakashita
Center for Biological Diversity

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Accumulation of Microplastic on Shorelines Woldwide: Sources and Sinks

Mark Anthony Browne,[*,†,‡,§] Phillip Crump,[¶] Stewart J. Niven,[§,∥] Emma Teuten,[§] Andrew Tonkin,[¶] Tamara Galloway,[⊥] and Richard Thompson[§]

[†]School of Biology & Environmental Sciences, University College Dublin, Science Centre West, Belfield, Dublin 4, Ireland

[‡]Centre for Research on the Ecological Impacts of Coastal Cities, A11 School of Biological Sciences, University of Sydney, NSW 2006, Australia

[§]Marine Biology & Ecology Research Group, School of Marine Science & Engineering, University of Plymouth, Plymouth PL4 8AA, United Kingdom

[¶]School of Geography, Earth & Environmental Sciences, University of Plymouth, Plymouth PL4 8AA, United Kingdom

[∥]Waters Canada, Ontario, Canada

[⊥]School of Biosciences, College of Life & Environmental Sciences, University of Exeter, Exeter EX4 4PS, United Kingdom

ABSTRACT: Plastic debris <1 mm (defined here as microplastic) is accumulating in marine habitats. Ingestion of microplastic provides a potential pathway for the transfer of pollutants, monomers, and plastic-additives to organisms with uncertain consequences for their health. Here, we show that microplastic contaminates the shorelines at 18 sites worldwide representing six continents from the poles to the equator, with more material in densely populated areas, but no clear relationship between the abundance of miocroplastics and the mean size-distribution of natural particulates. An important source of microplastic appears to be through sewage contaminated by fibers from washing clothes. Forensic evaluation of microplastic from sediments showed that the proportions of polyester and acrylic fibers used in clothing resembled those found in habitats that receive sewage-discharges and sewage-effluent itself. Experiments sampling wastewater from domestic washing machines demonstrated that a single garment can produce >1900 fibers per wash. This suggests that a large proportion of microplastic fibers found in the marine environment may be derived from sewage as a consequence of washing of clothes. As the human population grows and people use more synthetic textiles, contamination of habitats and animals by microplastic is likely to increase.

INTRODUCTION

We use >240 million tonnes of plastic each year[1] and discarded 'end-of-life' plastic accumulates, particularly in marine habitats,[1] where contamination stretches from shorelines[2] to the open-ocean[3−5] and deep-sea.[6] Degradation into smaller pieces means particles <1 mm (defined here as microplastic[2,7,8]) are accumulating in habitats,[1] outnumbering larger debris.[7] Once ingested by animals, there is evidence that microplastic can be taken up and stored by tissues and cells, providing a possible pathway for accumulation of hydrophobic organic contaminants sorbed from seawater, and constituent monomers and plastic-additives, with probable negative consequences for health.[9−16] Over the last 50 years the global population-density of humans has increased 250% from 19 to 48 individuals per square km,[17] during this time the abundance of micrometer-sized fragments of acrylic, polyethylene, polypropylene, polyamide, and polyester have increased in surface waters of the northeast Atlantic Ocean.[1] This debris now contaminates sandy, estuarine, and subtidal habitats in the United Kingdom,[1,6] Singapore,[18] and India.[19] Despite these isolated reports, the global extent of contamination by microplastic is largely unknown. This has prompted the United Nations, Group of Experts on Scientific Aspects of Marine Environmental Protection, International Oceanographic Commision,[14] European Union,[15] Royal Society,[3] and National Oceanic and Atmospheric Administration (USA)[16] to all identify the need to improve our understanding about how widespread microplastic contamination is, where it accumulates, and the source of this material. If spatial patterns of microplastic result primarily from the transportation of natural particulates by currents of water, shores that accumulate smaller-sized particles of sediment should accumulate more microplastic. Alternatively, spatial patterns may be influenced by sources of microplastic; with more material along shorelines adjacent to densely populated areas which already have a greater abundance of larger items of debris[20] and receive millions of tonnes of sewage each year[21] which has also been shown to contain microplastic.[22−26] Although larger debris is removed in sewage treatment plants, filters are not specifically designed to retain microplastic and terrestrial soils that have received sewage sludge do contain

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microplastic fibers.[27] In the UK alone, over 11 km^3 of water is discharged into inland waters, estuaries, and the sea each year[21] from treatment plants. Certain subtidal marine sites may, however, contain large quanitites of microplastic in their sediments because for nearly 30 years, a quarter of UK sewage sludge was dumped at 13 designated marine disposal-sites around the coast, until this practice was stopped in 1998 through The Urban Waste Water Treatment Regulations 1994.[21,22] Since substantial quantities of sewage sludge and effluent are discarded to the sea, there is considerable potential for microplastic to accumulate in aquatic habitats, especially in densely populated countries.

To manage the environmental problems of microplastic it is important to understand and target the major pathways of microplastic into habitats with mitigation-measures. While sewage waste provides one potential route for entry of microplastics, others have been identified including fragmentation of larger items, introduction of small particles that are used as abrasives in cleaning products, and spillage of plastic powders and pellets. Forensic techniques that compare the size, shape, and type of polymers[28] may provide useful insights into the sources of the microplastic. For instance, if the material originated from fragmentation, the frequency-distribution of sizes of plastic debris would be skewed to smaller irrgeular fragments from the major types of macroplastic (e.g., polyethylene, polystyrene, polypropylene) found in habitats.[7] If, however, scrubbers in cleaning products were more important, we would expect most of the material to consist of fragments and spheres of polyethylene. These sources do not, however, account for the occurrence of microplastic fibers in sludge and effluent taken from sewage treatment works[26] and soil from terrestrial habitats where sewage sludge had been applied, the source of which is more likely explained by fibers shed from clothes/textiles during washing.[27] Work is therefore needed to gather forensic information about the number, type of polymer and shape, to assess the likelihood of microplastic entering marine habitats through this possible pathway.

Here, we investigate the spatial extent of microplastic across the shores of six continents to examine whether spatial patterns relate to its sources or sinks. We test the following hypotheses that there will be more microplastic in habitats that accumulate smaller particles of sediment (hypothesis 1) and in areas with larger population-densities of humans (hypothesis 2). Based on forensic analyses of the material we then tested the hypotheses that sediment collected from sewage-disposal sites contains more microplastic than reference sites (hypothesis 3), that microplastic found on the shoreline will resemble microplastic found in subtidal sewage disposal sites, sewage-effluent discharged from treatment works, and wastewater from washing clothes using washing machines (hypothesis 4).

MATERIALS AND METHODS

Global Sampling of Sediment from Shores. Samples of sediment were collected from sandy beaches in Australia (Port Douglas; 16°29S, 145°28E; Busselton Beach 33°39S, 115°19E), Japan (Kyushu 32°24N, 131°39E), Oman, United Arab Emirates (Dubai 25°17N, 55°18E), Chile (Vina Del Mar 32°56S, 71°32W; Punta Arenas 53°08S, 70°53W), Philippines (Malapascua Island 01°18N, 01°103E), Portugal (Faro 36°59N, 07°57W), Azores (Ponta Delgado 37°44N, 25°34W), USA (Virginia 36°56N, 76°14W; 36°57N, 76°14W; California 35°50N, 118°23W), South Africa (Western Cape 33°06S, 17°57E), Mozambique (Pemba 19°01S, 36°01E), and the United Kingdom (Sennon Cove 50°04N, 05°41W) from 2004 to 2007. During collection (and in subsequent sections), cotton clothing was worn rather than synthetic items (such as fleeces) to avoid contamination by plastic fibers. Samples were collected by working down-wind to the particular part of the highest strandline deposited by the previous tide. Sediment was sampled to a depth of 1 cm deep using established techniques.[7] As the sampling was opportunistic, the sampling design was unable to remove possible confounding due to intrinsic dfferences in the tidal range and position of the strandline that will vary spatially and temporally on the shores. The extraction and identification of microplastic, including the analysis of sediment particle-size, was done using established methods.[1,7] Microplastic debris was extracted from a 50 mL subsample of sedimentary material using a filtered, saturated solution of sodium chloride to separate particles of microplastic from sediments. This involved three sequential extractions using the saline solution and identifying the microplastic using Transmittance FT- IR and a spectral database of synthetic polymers (Bruker I26933 Synthetic fibres ATR-library).

Marine Sewage Disposal and Reference Sites. In 2008 and 2009, samples of sediment (n = 5) were haphazardly collected from each reference (Plymouth 50°14N, 04°10W and Tyne 55°06N, 01°18W) and sewage-sludge disposal site (Plymouth 50°14N, 04°18W; Tyne 55°03N, 01°17W) using van Veen grabs deployed from a boat. The surface 5−10 cm of sediment of each sample was placed into precleaned 500 mL aluminum foil containers and microplastic extracted as before. During collection, cotton clothing was worn rather than synthetic items to avoid contamination by plastic fibers.

Sewage Effluent. Microplastic was extracted from effluent discharged (n = 5) by two sewage treatment plants. Precleaned glass bottles (750 mL) with metal caps were used to collect effluent from discharges from Tertiary-level Sewage Treatment Plants at West Hornsby and Hornsby Heights (NSW, Australia) in 2010. Effluent was filtered and microplastic counted as before but without additional saline water and standardized to give the amount of microlastic per liter of effluent.

Washing Machine Effluent. Because the proportions of synthetic fibers found in marine sediments and sewage resembled those used for textiles, we counted the number of fibers discharged into wastewater from using domestic washing machines used to launder clothing. To estimate the number of fibers entering wastewater from washing clothes, 3 different front-loading washing machines (Bosch WAE24468GB, John Lewis JLWM1203 and Siemens Extra Lasse XL 1000) were used (40 °C, 600 R.P.M.) with and without cloth (polyester blankets, fleeces, shirts). Detergent and conditioner were not used because these blocked the filter-papers. Cross-contamination was minimized (<33 fibers) at the start of the experiment and in between washes, by running washing-machines at 90 °C, 600 R.P.M for 3 cycles without clothes. Effluent was filtered and microplastic counted.[1,7]

RESULTS AND DISCUSSON

Eighteen shores across six continents were contaminated with microplastic (Figure 1), and so we investigated whether spatial patterns relate to its sources or sinks. The abundance of microplastic per sample ranged from 2 (Australia) to 31 (Portugal, U.K.) fibers per 250 mL of sediment (Figure 2A), consisting of

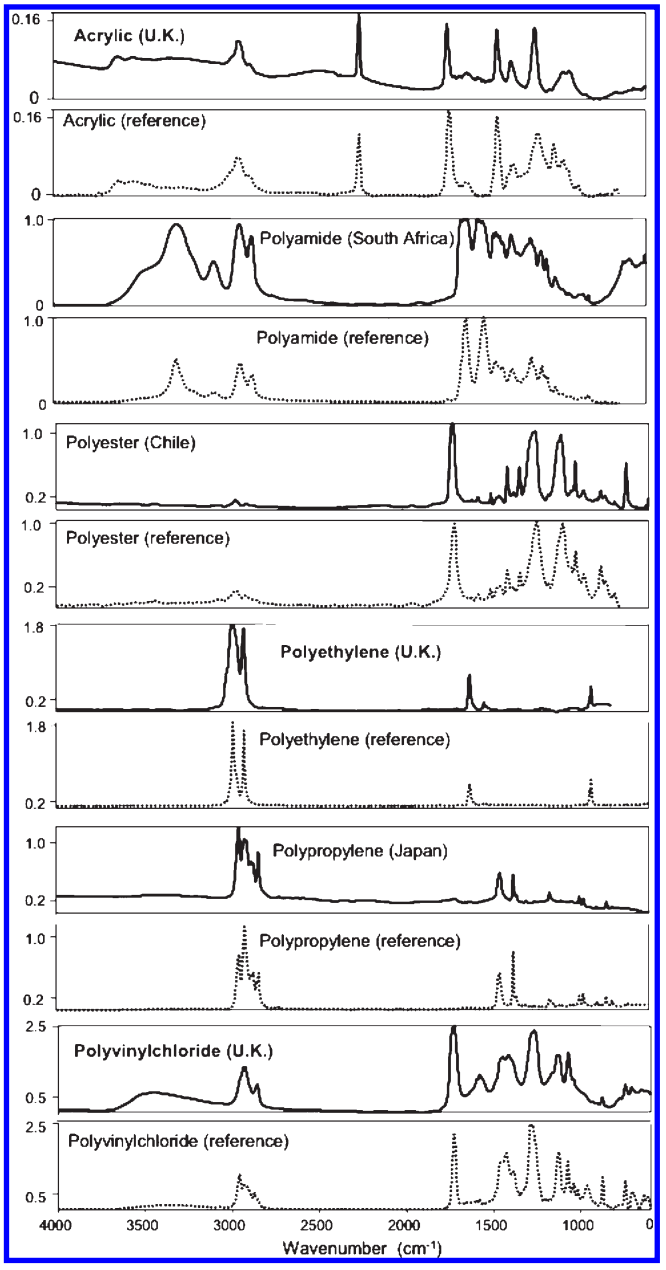

Figure 1. Examples of Fourier transform infrared spectra of microplastic and corresponding reference material from ATR spectral database, vertical axis represents transmission in standard optical density units. (Bruker Optics ATR-Polymer Library - a Collection of Synthetic Fibres, Copyright 2004 Bruker Optic GmbH).

polyester (56%), acrylic (23%), polypropylene (7%), polyethylene (6%), and polyamide fibers (3%). There was more microplastic in densely populated areas[24] with a significant relationship between its abundance and human population-density (Linear Regression, $F_{1,16} = 8.36$, $P < 0.05$, $n = 18$, $r^2 = 0.34$; Figure 2B), but no clear relationship with the mean-size of natural particulates (Spearman Rank rho = 0.39, $n = 18$, $P > 0.05$). As a consequence we explored the importance of sewage-disposal as a source of microplastic to marine habitats (Figure 2C). Despite sewage not being added for more than a decade, disposal-sites still contained >250% more microplastic than reference sites (2 Factor ANOVA, $F_{1,16} = 4.50$, $n = 5$, $P < 0.05$), mainly fibers of polyester (78%) and acrylic (22%). To further examine the role of sewage as a source, microplastic was extracted from effluent discharged by sewage treatment plants and compared with sediments from disposal-site. Effluents contained, on average, one particle of microplastic per liter. As expected, polyester (67%) and acrylic (17%) fibers dominated, including polyamide (16%), showing proportions of polyester and acrylic fibers in sewage-effluent resembled microplastic contaminating sediments from shores and disposal-sites. This suggests these microplastic fibers were mainly derived from sewage via washing-clothes,[26,27] rather than fragmentation[1,4,5,7,13−15,18,23] or cleaning-products.[2,7,11,14,16,23−25] Because proportions of polyester fibers found in marine sediments and sewage resembled those used for textiles (78% polyester, 9% polyamide, 7% polypropylene, 5% acrylic),[29] we counted the number of fibers discharged into wastewater from using washing blankets, fleeces, and shirts (all polyester). Here we show a garment can shed >1900 fibers per wash. All garments released >100 fibers per liter of effluent, with > 180% more from fleeces (Figure 2E), demonstrating that using washing machines may, indirectly, add considerable numbers of microplastic fibers to marine habitats. Because people wear more clothes during the winter than in the summer[30] and washing machine usage in households is 700% greater in the winter,[31] we would expect more fibers to enter sewage treatment during the winter. Research is therefore needed to assess seasonal changes in the abundance of plastic fibers in sewage effluent and sludge. In our study it was not possible to use detergent and conditioners because they blocked the filter-papers and prevented us from fitering the samples of effluent, so work is needed to investigate the effect of detergent and conditioner on the quantities of fibers in effluent.

Our work provides new insights into the sources, sinks, and pathway of microplastic into habitats. We show polyester, acrylic, polypropylene, polyethylene, and polyamide fibers contaminate shores on a global-scale, with more in densely populated areas and habitats that received sewage. Work is now needed to establish the generality of the relationship with population-density at smaller spatial scales, including freshwater and terrestrial habitats where sewage is also discharged. One source of these fibers of microplastic appears to be the disposal of sewage contaminated with fibers from washing clothes because these textiles contain >170% more synthetic than natural fibers[29] (e.g., cotton, wool, silk). The quantity of microplastic in sewage and natural habitats is, however, likely to be much greater. Brightly coloured fibers are easily distinguished from natural particulates, but microplastic from cleaning products and fragmentation will be discoloured by biofilms and resemble natural particulates, so better methods are required. In the future microplastic contamination is likely to increase as populations of humans are predicted to double in the next 40 years and further concentrate in large coastal cities[17] that will discharge larger volumes of sewage into marine habitats. To tackle this problem, designers of clothing and washing machines should consider the need to reduce the release of fibers into wastewater and research is needed to develop methods for removing microplastic from sewage. One means of mitigation may be ultrafiltration because fewer fibers have been found downstream from a sewage treatment plant that use this process as opposed to one that did not.[26] Work is urgently needed to determine if microplastic can transfer from the environment and accumulate in food-webs through ingestion. In humans, inhaled microplastic fibers are taken up by the lung tissues and can become associated with tumors,[32] while

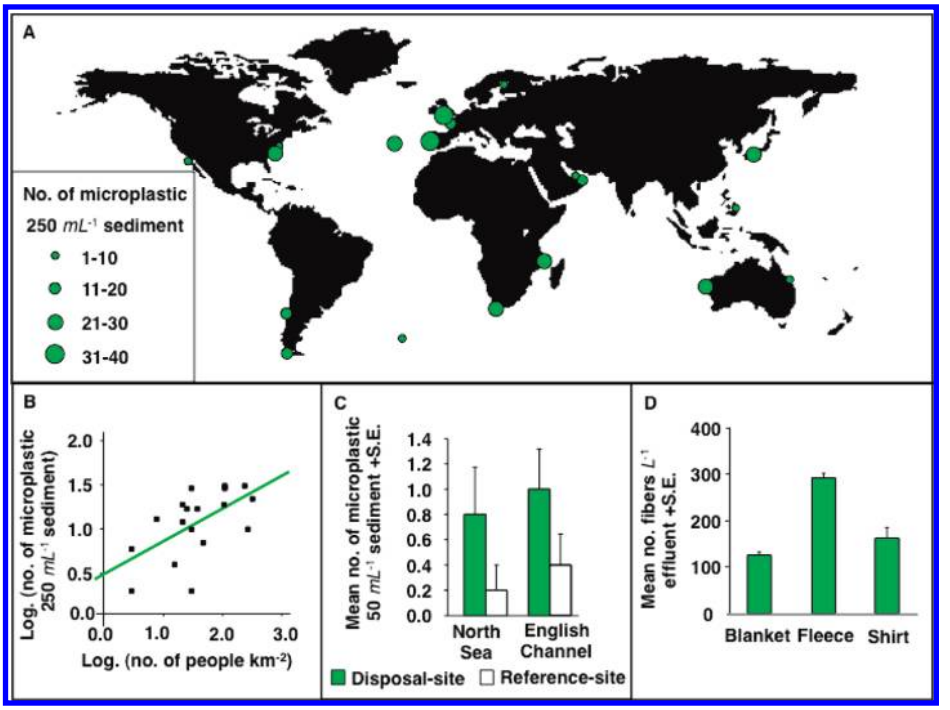

Figure 2. (A) Global extent of microplastic in sediments from 18 sandy shores and identified as plastic by Fourier transform infrared spectrometry. The size of filled-circles represents number of microplastic particles found. (B) Relationship between population-density and number of microplastic particles in sediment from sandy beaches. (C) Number of particles of microplastic in sediments from sewage disposal-sites and reference-sites at two locations in U.K. (D) Number of polyester fibers discharged into wastewater from using washing-machines with blankets, fleeces, and shirts (all polyester).

dispersive dyes from polyester and acrylic fibers have been shown to cause dermatitis.[33] Research is therefore needed to determine if ingested fibers are taken up by the tissues of the gut and release monomers (e.g., ethylene glycol, dimethyl terephthalate, propenenitrile, acrylonitrile, acrylonitrile, vinyl chloride, vinylidene chloride, vinyl bromide), dispersive dyes, mordants (e.g., aluminum, chromium, copper, potassium, tin),[34] plasticisers from manufacture and sorbed contaminants from sewage (e.g., organotin,[35] nonylphenol,[36] and Triclosan.[37] The bioavailability of these chemicals is likely to be greater from fibers of polyester and acrylic, compared to the more hydrophobic microplastics (e.g., polyethylene, polypropylene) that have more heterogenic atoms. In conclusion, our study shows the importance of testing hypothesis to improve our understanding about the sources and sinks of microplastic in habitats. Such experimental approaches are vital if we are to target the pathways of microplastic into habitats with effective mitigation-measures that reduce contamination by microplastic.

AUTHOR INFORMATION

Corresponding Author

*Phone: +353 (0) 870 916 484. Fax: +353 (0) 1 716 1152. E-mail: mark.browne@ucd.ie.

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