

# 海洋纳米塑料污染现状及其对双壳类的生态毒理效应

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**摘要:** 随着纳米塑料在海洋中的分布越来越广泛, 纳米塑料逐渐演变成目前海洋生态系统中面临的严重环境问题之一, 引起人们的广泛关注。纳米塑料比微塑料粒径更小, 具有更大的比表面积与吸附力, 成为海洋中污染物的重要载体之一, 影响深远。双壳类具有滤食特殊摄食方式, 可通过食物链影响其他营养级生物, 是食物链中重要一环。本文主要就纳米塑料的定义与来源、在海洋中的污染现状、对海洋双壳类的生态毒理效应进行阐述。纳米塑料可以通过海洋生物呼吸和进食过程中摄入体内, 在吞噬细胞中诱导氧化应激、线粒体损伤和细胞毒性并产生严重的炎症反应。研究表明, 在有其他污染物的存在下, 纳米塑料的存在, 会增加污染物在海洋生物体内的滞留时间, 从而加大其毒性。纳米塑料可以通过食物网对海洋生态系统构成威胁。

**关键词:** 纳米塑料; 污染现状; 双壳类; 生态毒理效应

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现在, 生态环境中存在的塑料成为最令人担忧的污染问题之一<sup>[1]</sup>。丢弃在环境中的塑料经过物理、化学和生物作用, 一步步形成小尺寸塑料颗粒, 并最终汇入海洋环境。当塑料颗粒粒径为 1~5 mm 时称为微塑料(microplastics, MPs)。海洋中 MPs 总量大约  $1.5 \times 10^{13}$  到  $5.1 \times 10^{13}$  个粒子不等<sup>[2]</sup>。从北极<sup>[3]</sup>到南极<sup>[4]</sup>的海水中, 以及河流<sup>[5]</sup>或湖泊<sup>[6]</sup>等淡水中都存在 MPs, 纳米塑料(nano-plastics, NPs)作为尺寸比 MPs 更小的塑料同时也广泛存在。纳米材料定义为具有 1~100 nm 纳米级的外部尺寸或具有纳米级的内部结构或表面结构的材料<sup>[7]</sup>。纳米颗粒被定义为具有所有 3 个纳米级外部尺寸的纳米物体, 其中纳米物体的最长轴和最短轴的长度没有明显差异。如果尺寸差异显著(通常超过 3 倍), 则纳米纤维等术语可能优于纳米颗粒, 但同属于纳米颗粒的范畴。对海洋生物的影响可能来自 MPs 和 NPs 的物理化学性质以及它们可能造成的潜在组织损伤, 也可能来自于其所吸附的有毒化学品的毒性<sup>[8-13]</sup>。

在现代生活中的各个方面塑料都必不可少, 例如包装、建筑、汽车、电子和电器、服装、农业等<sup>[14]</sup>。海洋环境中最常见的聚合物是聚乙烯(PE)、聚丙烯(PP)、聚苯乙烯(PS)、尼龙(PA)、聚对苯二甲酸乙二

酯(PETP)、聚氯乙烯(PVC)和醋酸纤维素(CA)<sup>[15]</sup>。有研究发现, 在水生系统中聚苯乙烯纳米塑料(polystyrene-nanoparticles, PS-NPs)比聚苯乙烯微塑料(polystyrene-microparticles, PS-MPs)显示出更大的急性毒性<sup>[16]</sup>。NPs 作为比 MPs 更小的颗粒, 其更容易被海洋生物所误食, 甚至有可能在呼吸过程中就会进入到生物体内。此外, NPs 具有更高的比表面积, 更加容易成为各种污染物的载体, 包括重金属<sup>[7, 15]</sup>、多环芳烃(PAHs)<sup>[2, 17]</sup>、多氯联苯(PCBs)<sup>[6, 9, 18]</sup>、二氯二苯基三氯乙烷(DDT)<sup>[6]</sup>、多溴二苯醚(PBDE)<sup>[19-20]</sup>, 以及其他污染物, 如多氟烷基物质(PFA), 药品和个人护理产品<sup>[21-23]</sup>。双壳类生物具有特殊的生存环境, 并且属于滤食性生物, 更加容易误食这些污染物, 在体内积累, 并通过食物链进入高营养级动物, 并在体内积累, 并产生不可逆的损伤, 导致不可忽视

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的毒性效应<sup>[24]</sup>。

## 1 纳米塑料定义与来源

根据形成过程, NPs 可分为初级纳米塑料和次级纳米塑料<sup>[5]</sup>: 在最初生产时, 制造的塑料尺寸就属于纳米塑料范围的, 称之为初级纳米塑料。初级纳米塑料使用较为广泛, 如作为化妆品或医学药物的载体<sup>[21]</sup>, 这些产品在使用之后, 会残留大量的 NPs, 而这些 NPs 可以进入水环境, 随水流进行迁移。Hernandez 等<sup>[25]</sup>研究发现, 个人护理产品, 如洗面奶、牙膏和去角质等, 为了达到更好的清洁效果, 其中都添加了 NPs 颗粒<sup>[26-27]</sup>, 使用之后会产生大量的 NPs 颗粒, 最终进入到水环境。在物理、化学和生物作用下, 较大的塑料结构发生损坏, 会形成尺寸更小的塑料颗粒, 当塑料颗粒粒径在纳米范围内称之为次级纳米塑料<sup>[28]</sup>。例如, 暴露在阳光下的塑料碎片会因为受到紫外线辐射, 导致聚合物基体氧化, 化学键断裂, 所以长时间暴露在环境中的塑料会逐步老化, 碎裂成更小的塑料颗粒形成 MPs/NPs<sup>[1, 29-30]</sup>。在陆地生态系统中, 如农田土壤<sup>[10]</sup>和滩涂地带<sup>[11]</sup>均检测出 NPs; 水生生态系统中, 如海洋<sup>[4]</sup>、河流<sup>[21]</sup>和湖泊<sup>[4]</sup>也检测出 NPs 的存在。

水生环境中 NPs 的来源很复杂, 一些学者认为, 80%的 NPs 来自于陆地<sup>[31]</sup>。被丢弃在环境中的塑料直接暴露于环境、个人护理用品随下水道或由污水处理厂进入河流和湖泊, 最终通过降雨或风力作用进入海洋<sup>[32-33]</sup>。在这些途径中, 污水处理厂所占比例最大。在芝加哥某高度城市化的河流中检测到 MPs/NPs 的数值, 远高于当前海洋水体中的 MPs/NPs<sup>[5]</sup>。常规的污水处理(过筛、沉淀、生物处理和深层处理)可以去除 90%以上的塑料颗粒, 但污水厂数量众多, 排水量巨大, 进入到水环境中的塑料仍然在持续增加<sup>[9]</sup>。事实上, 污水除去的那 90%的塑料进入到淤泥中, 通过降雨仍然会进入水体环境。码头上的塑料用品, 如常见的绳索和捕网, 在水环境下老化速度会加快。Welden 等<sup>[34]</sup>研究表明, 等足类浮游动物会加快海洋塑料垃圾破碎变成 MPs/NPs 颗粒, 一个等足类浮游动物大约会创造出上千个 MPs/NPs 颗粒<sup>[35-36]</sup>。在水体中塑料颗粒会受到风力运输作用、洋流作用, 然后被输送到水环境的各个角落, 目前的研究已经发现, 在淡水水体、

太平洋、大西洋、极地等水体中都检测出了 MPs/NPs 的存在<sup>[37-39]</sup>。

## 2 纳米塑料在海洋生物中污染现状

塑料污染已广受关注, Ter Halle 等<sup>[40]</sup>在北大西洋亚热带环流中对 NPs 的分布进行测定表明, 其浓度在 13~501 个/m<sup>3</sup>, 主要由 NP-PE(73%)、NP-PP(13%)、NP-PVC(8%)、NP-PS(2%)和 NP-PET(1%)等组成。因 NPs 的采集与鉴定方法的缺乏, 所以在海洋中分布情况与污染现状研究较少<sup>[41]</sup>。MPs/NPs 的存在可对生态系统产生影响<sup>[22]</sup>。MPs/NPs 在水环境中会出现凝聚行为, 凝聚的过程会使塑料大小发生改变, 影响其在环境中的行为<sup>[42-43]</sup>。MPs/NPs 如果稳定地在水体中悬浮或凝聚速率比较慢, 那么迁移的距离可能会更远, 对生态影响更深远。但是, 如果凝聚行为发生迅速, 那么就会沉降到沉积物中, 从而被底栖生物摄食<sup>[29]</sup>。

藻类吸附 NPs 主要依赖于 NPs 表面粒子电荷和表面功能团, NPs 在藻类代谢过程中会引起生理损伤。当塑料颗粒粒径为 53 nm 的聚苯乙烯纳米塑料(polystyrene nanoplastics, PS-NPs)时, 海洋微藻表面吸附大量的 NPs 颗粒可经过水蚤摄食进入体内, 再通过食物链进入下一营养级<sup>[23]</sup>。Ory 等<sup>[44]</sup>在 Rapa Nui 海岸采集的岛鲹(*Caramague Demtue*)样本中 80%的个体体内含有 MPs, 且其体内的塑料颜色、大小与其猎物高度相似。Baalkhuyur 等<sup>[30]</sup>采集了 26 种不同种类鱼样本, 体内也检测到 MPs, 其中以底栖无脊椎动物为食的红拟眶棘鲈(*Parascalopsis eriomma*)体内 MPs 浓度最高。MPs/NPs 可以由食物链传递, 并最终进入营养级更高的海洋生物体内产生积累, 甚至对生物造成不可逆的伤害。Chae 等<sup>[45]</sup>发现 NPs 可以由生产者传递给初级消费者再到次级消费者, 最终进入第三级消费者。此外, MPs/NPs 比表面积非常大, 可能会释放出塑料添加剂或吸收污染物<sup>[10, 46]</sup>。研究发现 MPs/NPs 可以与重金属等其他有毒元素相互作用, 可作为污染物的载体向生物体内转移<sup>[10, 47-48]</sup>。

MPs/NPs 不能从皮肤直接进入生物体<sup>[49]</sup>, 但是可以在体内蓄积<sup>[28]</sup>。NPs 对大型溞(*Daphnia magna* Straus)毒性显著比 MPs 的强, MPs/NPs 均能对其造成运动器官的损伤和堵塞其滤食系统; 浓度很大时富集在体表, 对其表皮造成损伤, 严重时导致其死

亡<sup>[33]</sup>。可以推断 MPs/NPs 粒径越小,可能更容易进入生物体内,产生的毒害作用更加显著<sup>[50]</sup>。当 MPs/NPs 被生物体摄入后,可以转移到生物体的各个部位。如河豚暴露在不同浓度直径 80 nm 的绿色荧光 PS-NPs 下,其肠、肝脏、胃、外套膜和鳃中均发现了 PS-NPs 的存在<sup>[51]</sup>。

### 3 纳米塑料对双壳类的影响

目前,各国海洋领域、环境领域学者开始关注各自国家内陆水体及沿海水环境中 MPs/NPs 污染及其对生物的毒性效应。双壳类在具有高密度 NP-PE 的环境中,会产生强烈的炎症反应,对其组织和亚细胞结构产生显著影响<sup>[23, 52]</sup>。MPs/NPs 具有潜在的生物学毒性及危害,主要包括两方面:一是 MPs/NPs 对生物体的直接伤害。环境中大量的塑料,经过物理、化学和生物的作用后,形成性质不同碎片及颗粒。具有锋利边缘的 MPs 甚至可以对组织和器官(如肠道)造成物理损害。二是 MPs/NPs 会吸附环境中的污染物<sup>[16, 47]</sup>。NPs 粒径更小,更容易通过血脑屏障,产生更严重的影响,且比表面积更大,携带更多污染物进行远距离运输,对生物产生更严重的危害。Hirai 等<sup>[53]</sup>研究发现,在海洋中收集的 MPs 样品表面吸附的持久性有机污染物比海水中的高 6 个数量级。MPs/NPs 性质稳定,很难在短时间内降解,海洋中初级消费者(浮游动物、双壳类和一些鱼类),在进食过程中都有可能将其当做食物进行摄食,并通过食物网传递对各个营养级生物造成潜在威胁<sup>[21]</sup>。

#### 3.1 纳米塑料对双壳类生态毒理学效应

Goldberg 等<sup>[54]</sup>最早提出使用双壳类作为评价河口和近岸海域环境质量状况的生物指示物。与其他海洋生物相比,双壳类不仅具有分布广、固着性强、生命周期长、采样检测容易等优点<sup>[55-56]</sup>,更重要的是,其体内的有机污染物含量可以更准确地反映环境的污染程度和变化。因此,双壳类被广泛用于监测海洋环境中的污染物,例如全球“Mussel Watch”计划。

双壳类直接从水体进行一系列的生命活动,对水体变化非常敏感。通过监测双壳类在受到外在胁迫时的生态生理变化,可用来反映水体环境污染状况。滤食是双壳类吸收外源物质的主要方式<sup>[57]</sup>。双壳类为减少压力影响,当过滤受到抑制,会根据应

力强度调节过滤速度,减少 NPs 进入体内<sup>[58]</sup>。此外,NPs 更容易穿透生物屏障进入体内逐渐积累。聚集性也是纳米材料的典型性特征之一,并且在生物体内比在自然环境中更容易聚集<sup>[59]</sup>,这种聚集会阻碍生物的呼吸和消化等,还可能会导致保护膜损坏并在吞噬细胞中诱导氧化应激、线粒体损伤、细胞毒性<sup>[60-62]</sup>和代谢变化,从而加大了 NPs 的毒性作用。纳米级的聚合物颗粒可以穿透并积聚在组织器官中,甚至改变基因表达<sup>[63]</sup>。

在双壳类血细胞中,50 nm PS-NH<sub>2</sub> 可诱导剂量依赖性吞噬细胞活性下降,溶菌酶增加,并导致溶酶体的损伤和凋零<sup>[64]</sup>。NPs 一般通过内吞作用释放到细胞质中,可干扰线粒体和细胞核等重要器官的正常代谢;可在质膜水平引起快速的细胞损伤,如对质膜泡的运输影响,丝状伪足消失并形成短的、水泡状伪足,常伴随可见的腔内空泡;酵素颗粒吞噬活性的大幅下降,吞噬/内吞途径被全面破坏<sup>[65]</sup>;干扰细胞内运输载体的运输;如果最终进入溶酶体,也很难被消化,干扰溶酶体的正常的降解功能;由 p38 MAPK 信号失调介导的损伤,最终会导致细胞损伤或死亡<sup>[66]</sup>。

NPs 粒径更小,在生物体内相较于 MPs 更加难以排出生物体。NPs 具有更长的肠道滞留时间(GRT),并可以携带更多的污染物进入生物体,加剧对生物体的毒性作用<sup>[42]</sup>。双壳类能够在肠道中选择不同粒径的纳米塑料颗粒<sup>[52, 67-68]</sup>,食物的 GRT 越长,通常表明该物质经历了更广泛的细胞外消化,并且可能被运输到消化腺进行更完整的细胞内消化。较长的 GRT 表明大多数 NPs 直接进入消化腺的小管,并可能通过内吞作用被消化细胞吸收。研究表明,紫贻贝(*Mytilus edulis*)的分离消化腺细胞和整个动物都可以吸收 NPs<sup>[69]</sup>。

#### 3.2 纳米塑料与污染物对双壳类的联合毒性

因 NPs 自身固有特性,在水环境中很难单独存在,所以通常以聚合物的形式存在。NPs 本身也会对生物体组织造成损伤,使生物体机体代谢紊乱。NPs 与有机污染物相互作用会增加其在生物体内富集,减慢代谢速率,降低了生物体对污染物代谢,增加了生物毒性,对器官造成损伤。在 NPs 与有机污染物共同存在时,可以通过破坏与免疫相关的分子途径激活,以此来触发双壳类生物的免疫反应<sup>[70]</sup>。细胞内 Ca<sup>2+</sup>在吞噬体的成熟过程中非常的重要<sup>[71]</sup>,其中

分泌因子 TLR4、IKK $\alpha$ 、TRAF6 和 NF- $\kappa$ B 的表达被明显抑制<sup>[70]</sup>。NPs 与有机污染物菲的联合暴露下显示出显著的联合效应和更大的潜在生物毒性风险。其中 NP-PS 对菲的吸附能力高于 NP-PE 和 NP-PP, 而 NP-PE 对菲的吸附能力又分别高于 NP-PP 和 NP-PVC<sup>[55]</sup>。NP-PS 具有大量的苯环,  $\pi$ - $\pi$  结构可以吸附更多的污染物, 加大了对四角蛤蜊(*Mactra veneriformis*)的毒性<sup>[72-73]</sup>, NPs 相较于 MPs 具有更大的比表面积<sup>[18, 74]</sup>, 更容易吸附在四角蛤蜊外套膜组织上<sup>[75]</sup>, 同时也能够携带更多的污染物和微生物<sup>[76]</sup>。具有疏水性的 NP-PS, 在水动力作用下更容易聚集成尺寸较大的颗粒, 减少对污染物的吸附量<sup>[74]</sup>。虽然一些颗粒可能会被鳃、外套膜和唇瓣的上皮细胞吸收, 但内部暴露和潜在影响的主要途径是通过颗粒的捕获和摄入<sup>[77-78]</sup>。

在紫贻贝(*Mytilus edulis*)中, PS 纳米颗粒(30 nm; 100~300  $\mu$ g/mL)诱导产生假粪便, 并随着 NPs 浓度的增加而增加, 表明 PS 颗粒可被视为非营养或低营养食物<sup>[59]</sup>。地中海贻贝血细胞暴露于 PS-NH<sub>2</sub>(50 nm; 1~50  $\mu$ g/L)中 30 min 后, PS-NH<sub>2</sub>会导致溶酶体膜稳定性降低和氧自由基生成增加, 导致细胞迅速受损, 如质膜泡和丝状伪足的损伤<sup>[64-65]</sup>。研究表明<sup>[79]</sup>, 在粒径为 30 nm、50 nm 的塑料颗粒与苯并芘和 17 $\beta$ -雌二醇作用下, 泥蚶(*Tegillarca granosa*)免疫活性的细胞类别减少, 从而减弱血细胞对异物的识别与降解。凋亡途径中的 Bcl-2 和 Caspase-3 在污染物暴露后分别显著上调和下调, 破坏细胞凋亡和 Ca<sup>2+</sup>信号相关的分子路径等一系列的免疫反应, 导致免疫能力下降。苯并芘和 17 $\beta$ -雌二醇的毒性在双壳类中受到塑料颗粒大小的影响, 塑料尺寸越大免疫反应就越弱, 尺寸越小免疫反应越剧烈<sup>[80-81]</sup>。NPs 与有机污染物相互作用会增加其在生物体内富集, 减慢代谢速率, 而 NPs 本身也会对生物体组织造成损伤, 使生物体机体代谢紊乱, 降低了生物体对污染物代谢, 增加了生物毒性, 对器官造成损伤。

## 4 结论

本文对海洋中纳米塑料的来源、污染现状以及对海洋双壳类的影响进行了阐述。与微塑料相比, 较小尺寸的纳米塑料有更高的比表面积, 吸附污染物的能力增强, 增加塑料在生物体内的滞留时间, 显著阻碍生物的解毒能力, 增强了其毒性, 对生物体造成严重损伤。

近年来, 更多的研究是微塑料对水生生物和生态系统的生态毒性和影响。有关纳米塑料对海洋生物的影响机制, 还知之甚少, 大多数停留在毒性影响层面。关于纳米塑料的来源与分布、与其他污染物的相互作用和影响机制, 缺乏相关的研究, 仍然需要进一步的探索。

就现在的研究而言, 大多数实验在室内完成, 进行短期急性生态毒理实验, 与实际海洋环境中的纳米塑料浓度有较大的差别, 且纳米塑料形态也较为单一。在以后的研究中应该考虑实际海洋环境中的塑料类型、形态等与实验条件的差别, 并进行长期的慢性毒理实验, 以期得到更具有现实意义的数据。

纳米级的塑料颗粒更容易穿透生物屏障进入生物体内, 会使生物的氧化应激、酶发生改变并导致细胞损伤等, 在以后的研究中应该考虑与组学相结合, 以期深入细致地探究其毒理机制。

对于海洋生物的生态毒性研究仍然处于探索阶段, 纳米塑料对双壳类的生物机制有待进一步探究。在以后的研究中应该考虑增加海洋中不同营养级的特征生物对纳米塑料在食物链传递、富集和生态毒性等对海洋生物的影响。并对纳米塑料的隐性影响进行探索。

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## Current status of marine nanoplastic pollution and its ecotoxicological effects on bivalves

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**Abstract:** The increased ocean-water contamination due to the increasing amount of nanoplastics has become one of the serious environmental problems in marine aquatic ecosystems, which has attracted widespread attention. Nanoplastics have a smaller particle size and larger specific surface area and adsorption power than microplastics, becoming one of the important carriers of pollutants in the ocean, with far-reaching effects. Further, they can enter the body during the respiration and feeding of marine organisms. Studies have shown that the presence of nanoplastics in the presence of pollutants increases the retention time of pollutants in marine organisms, thus increasing their toxicity. This can pose a threat to ecology and human health through the food web. Bivalves play an important link in the food chain because of their special feeding mode known as filter feeding, which responds significantly to changes in water bodies and is often used for water-body assessment. This review focuses on the definition and sources of nanoplastics, the current status of pollution in the ocean, and the ecotoxicological effects on marine organisms, mainly bivalves.

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