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Global Pattern of Microplastics (MPs) in Commercial Food-Grade Salts: Sea Salt as an Indicator of Seawater MP Pollution

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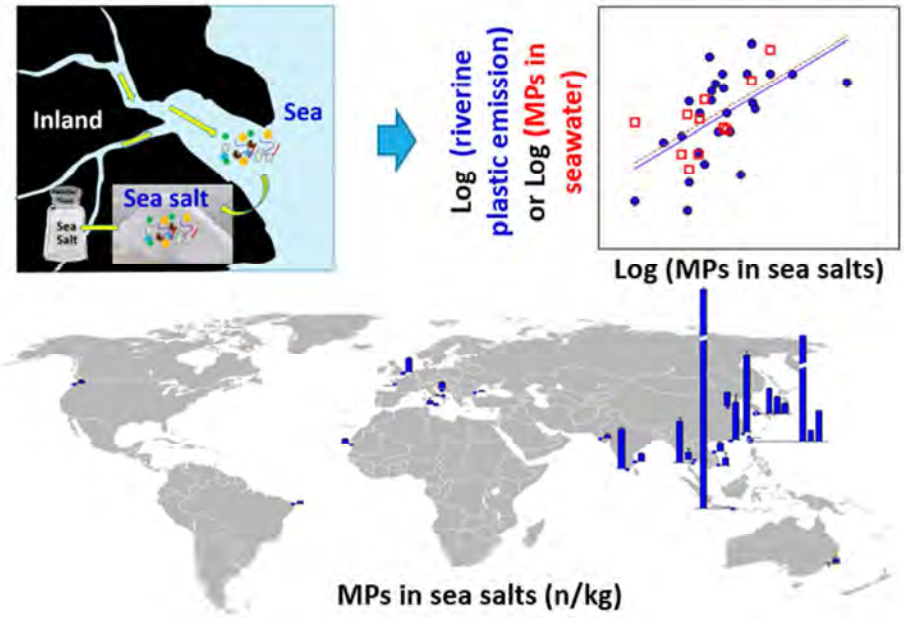

Table of Contents/Graphical Abstract

ABSTRACT

Previous studies have identified microplastics (MPs) in commercial table salts but could not exactly address the origin of the MPs because of several limitations. The present study is based on the hypothesis that commercial sea salts can act as an indicator of MP pollution in the surrounding environment unless the MPs are filtered out during the manufacturing process. A total of 39 different salt brands produced at geospatially different sites, including 28 sea salt brands from 16 countries on six continents, were investigated. A wide range of MP contents (in number of MPs per kg of salt; n/kg) was found: 0 – 1,674 n/kg (excluding one outlier of 13,629 n/kg) in sea salts, 0 – 148 n/kg in rock salt, and 28 – 462 n/kg in lake salt. Relatively high MP contents were identified in sea salts produced in Asian countries. The abundance of MPs in unrefined sea salts (n=25) exhibited significant linear correlations with plastic emissions from worldwide rivers (r=0.57; p=0.003) and with the MP pollution levels in surrounding seawaters (r=0.68; p=0.021) in the published literature. The results indicate that not only is Asia a hot spot of global plastic pollution, as previous studies have suggested, but also that sea salt can be a good indicator of the magnitude of MP pollution in the surrounding marine environment.

1. Introduction

Plastic debris entering the marine environment is a global concern. It was estimated that eight million metric tons of plastics with a range of 4.8 – 12.7 million metric tons (MMT) were released into the world's oceans in 2010.[1] Another study estimated that plastic input to the world's ocean via rivers could reach 1.15 – 2.41 MMT/year.[2] Despite the wide range of the estimates, it is commonly accepted that Asia is a global hot spot or major emitter of the world's ocean plastic pollution. Most of the top 20 emitting countries[1] and the top 20

26 polluting rivers[2] are located in Asia, accounting for approximately 70% of the global total of
27 plastic emissions. This plastic debris fragments into fine particles with diameters of less than
28 5 mm, so-called 'microplastics (MPs)', which are of critical concern due to their easier
29 transport via currents, biological/human exposure, and harmful effects.[3-5] Thus, the
30 ecosystem and human health in Asian marginal seas could potentially be at greater risk
31 because of severe maritime MP pollution. Some monitoring data for seawater,[6,7] sediment,[8-9]
32 and organisms[10,11], which showed relatively high MP abundance in the Asian region, seems to
33 support such concern.

34 So far, numerous studies have focused on monitoring seawater as a medium of MP
35 pollution. However, due to inconsistencies in measurement and sampling methods[6,12]
36 between studies and the limitations of snap-shot grab sampling, it is difficult to identify the
37 geographical distribution of MP pollution by solely direct comparison of different seawater
38 monitoring data. Furthermore, seawater monitoring is financially expensive and labor
39 intensive and has limitations with regard to the kinds of MPs collected because of the mesh
40 size (mostly >300 μm)[13] of the nets used. Compared with the restrictions of monitoring
41 seawater, sea salt is not only much easier to obtain but it might better reflect the
42 characteristics of the MP pollution present in the seawater because it is directly produced
43 from bulk seawater. We hypothesized that commercial sea salt would be a good indicator of
44 the MP pollution level in the source seawater, as well as of human exposure to MPs
45 originating from marine environment, unless disrupted by additional refining or
46 contamination during manufacturing.

47 Previous studies have found the presence of MPs in commercial table salts but could
48 not exactly address the origin of the salt-hosted MPs because of several limitations, such as
49 geographical constraints on the investigated samples, limited information on production
50 processes, and differences in MP analysis methods.[14-19] In works on Chinese and Turkish

table salts, researchers attributed the significantly higher content of MPs in sea salt relative to rock/well salt or lake salt to the much higher MP pollution in the coastal environment of China[14] or high MP pollution pressure of Turkish coast.[15] On the other hand, Iñiguez *et al*.[16] suspected the background presence of MPs in the environment as a possible reason of MP pollution in sea salt, because the MP content in Spanish salt (in number of MPs per kg of salt; n/kg), with values of 50 – 280 n/kg, was not significantly different from that in other types of salt (sea salts versus well salts). Compared to these three studies, much lower amounts of MPs (only 30 MP particles from all salt samples; a range of zero – 10 n/kg) were present in 16 different salt brands from eight origin countries purchased from a Malaysian market, and the source of the salt-hosted MPs was not addressed.[17] Kosuth *et al*.[18] found "anthropogenic debris" at concentrations of 47 – 806 n/kg in table salts purchased in Minneapolis, Minnesota, without any further investigation of the cause of the salt pollution. Lastly, Renzi and Blašković[19] mentioned dense human activities around salt production sites and polluted Mediterranean Sea as causes of several thousands to ten thousands of MP particles detected in 1 kg of Italian and Croatian fine sea iodate salts.

Notably, in the aforementioned six studies, different techniques and protocols were used to quantify the amount of MPs and to identify the polymers of MPs. For instance, the identities of only randomly selected MP-like particles[14,16] or clear fibers in only selected some samples[19] were verified using Fourier transform infrared spectroscopy (FTIR) analysis. Meanwhile, in other studies, MPs in salts were measured with Raman spectroscopy analysis[15,17] or visual observation using biological staining without any spectroscopic verification.[18] These differences in identification methods are related to the uncertainty in the determination of MPs in environmental samples, as seen in previous studies[20-24] and discussed later (section 4.1), and can cause inconsistency in occurrences of MPs among salts investigated, resulting in ambiguity in the geospatial distribution of MP contents and sources

of MPs in sea salts.

In the present study, we compared 28 sea salt brands produced from geospatially different sites, i.e., 16 countries on six continents, to reveal the relationships between riverine plastic inputs and the magnitude of MP pollution in other marine compartments. Additionally, rock and lake salts were also investigated for comparison. For identification of MPs, most of the individual MP-like particles were analyzed using FTIR to reduce uncertainty in the occurrence and determination of MP particles, as mentioned above.

2. Materials and Methods

2.1. Sample collection

A total of 39 brands of commercial table salt were collected from the supermarkets of 17 different countries on four continents between January and September 2017 (Table 1). The samples represent 21 countries from six continents because some samples, although purchased from the same country (i.e., consumption location), have a different country of origin (i.e., production location), as depicted in Figure S1 and S2. Among the off-the-shelf salt brands from each country, the target samples were selected based on available information, such as geospatial location of the source, raw material of the source (seawater, lake water, or halite), and the method of manufacturing (solar-dried, refined, or unrefined). Information on each sample was collected through analysis of package labels, web research, and phone interviews. The exact production sites of two salt brands, PH-R1 and SN-S1, could not be confirmed due to lack of available information; however, they are assumed to be produced in the Philippines and Senegal, respectively, because they were known to be local salts. The salts are classified as sea salt, rock salt, and lake salt depending on whether they are sourced from seawater, halite, or lake water, respectively. The present research focused on collecting diverse salt types but mainly focused on collecting sea salt.

Based on raw material and production location, the sampled salt products consist of 28 sea salts from 16 countries, 9 rock salts from 8 countries, and 2 lake salts from two countries (Table 1). Packages of individual samples contained salt amounts of 100 to 1000 g. In the present study, one to five packages of each salt brand were purchased to obtain 500 g as a minimum weight for analysis because the analysis of each 250 g sample was duplicated for each brand of salt. For two sea salt brands from Chinese Taipei (CT-S3 and S4), only one sample (250 g) each was analyzed one time because these samples were additionally collected to compare with CT-S1.

2.2. Sample treatment and microplastic measurement

For duplicate analysis, 250 g was taken from each of two different packages for brands with a package unit weight of 250 – 500 g. Two 250 g samples were taken from one package for brands with a package unit weight of 1000 g. For brands with a package unit weight of less than 250 g, two 250 g samples were taken from a well-homogenized composite sample of three to five packages.

The analysis of MPs in salts mostly followed the procedure presented in previous studies[14,16] with some modification (see the details in section S1 of supporting information). In brief, a 250 g unit sample was digested and dissolved using 17.25% H_2O_2 solution and filtered through a filter (GF/D; ø 2.7 μm; Whatman), and all of the MP-like particles on the filter were analyzed using microscopic and spectroscopic analysis.

For all the MP-like particles, excluding mineral particles and biological soft tissues, their physical characteristics, such as shape (four classifications: fragment, fiber, sheet, and spherule), size (100 – 5000 μm), and color (13 classifications: white, ivory, yellow, orange, red, pink, purple, green, brown, gray, blue, black, and transparent) were visually observed under a dissecting stereo-microscope (Leica S8APO) equipped with a Leica MC170 HD

126 Camera & SW kit with a magnification range of 10× – 80×. Thereafter, each MP-like particle
127 was carefully hand-picked using microforceps and analyzed to identify the associated
128 polymer using a FTIR instrument (Agilent Cary 630 FTIR spectrometer) equipped with
129 Diamond ATR mode. All the sample spectra were recorded as 32 scans in the spectral range
130 of 650 – 4000 cm^{-1} at a resolution 4 cm^{-1} and were compared to the polymer library
131 (KnowItAll, Bio-Rad) to identify the polymer type. A total of 14 different polymers were
132 identified at least once: polypropylene (PP), polyethylene (PE), polystyrene (PS),
133 polyethylene terephthalate (PET), polyvinylchloride (PVC), nylon, polyacrylate (PA),
134 ethylene-vinyl acetate (EVA), acrylic, polycarbonate (PC), phenoxy resin (PR), polyurethane
135 (PU), Teflon, and paraffin wax (PW). Details of the spectral analysis were described in
136 previous studies.[6,25] In the present study, matches between the polymer and reference spectra
137 better than 70% were accepted, as suggested in previous studies.[26,27] MP particles with a
138 matching rate between 60% and 70% were identified individually based on both the
139 characteristic features of synthetic polymers observed in visual examination and the closeness
140 of their absorption frequencies to those of chemical bonds in the known polymers.

141 Indonesian sea salt (ID-S1), which contained a tremendous number of MP particles
142 (over 3,000 MP particles per 250 g sample), required too much time for polymer
143 identification (see Figure S3). Therefore, MP particles with sizes of <500 μm in only 12
144 rectangular zones with an area of 4.7×3.6 mm (which represents 15% of the entire MP
145 retained area) were fully analyzed, and the results were extrapolated to the number of MPs in
146 the whole area. All particles with sizes >500 μm were analyzed for the entire filter zone (ca.
147 13.8 cm^2).

148

149 **2.3. Quality control and quality assurance**

150 To rule out the possibility of contamination,[28] strict control protocols were followed

during the entire procedure for sample treatment and analysis (see section S2 in the supporting information). To monitor potential contamination during the sample handling process, two procedural blanks and one air blank were processed together with ten samples for every analysis batch. Using the air blank, potential airborne contamination was monitored by exposing the GF/D filter paper to the laboratory air whenever the samples were exposed to the air. Procedural blanks containing only filtered water were treated simultaneously with the samples. No MPs were observed in any of the eight air blanks, and only one PET fiber was found in one of the 16 procedural blanks. The method detection limit (MDL) values, equal to three times the average MP number found in the procedural blanks, were 0.72 n/salt-kg for PET fiber and 0 MP/salt-kg for other MP types. Therefore, the potential contamination was negligible, and thus, MPs found at >1 n/kg originated from the samples.

To check the variation in the analysis, we measured MP contents in duplicate for each salt brand. Duplicates are not sufficient to statistically result in a validated coefficient of variation (CV); however, the numbers provide some reference. The CV for the duplicates was on average 33%, with CVs of <15% for the samples with MP number of more than 10.

2.4. Statistical analysis

Statistical analysis of data was performed using IBM SPSS® Statistics 23 software. As normality in the distribution of MP content was rejected by the Kolmogorov-Smirnov or Shapiro-Wilk test, a nonparametric test (Mann-Whitney U-test) was conducted to determine the mean differences in MP contents among salt groups.

3. Results and Discussion

3.1. Identity of MPs in salt samples

A variety of MP particles, with varying shape, color, size, and polymer type, were

found in the salt samples (Figure S4). The polymeric identity of all MP-like particles ≥100 μm in a total of 76 salt samples including replicates was verified through FTIR. Overall, 91% (n=9,752 particles) of all identified particles (n_{tot}=10,723 particles) were synthetic MPs, and 2.5% (n=272 particles) were natural polymers, such as cotton, with mineral particles accounting for a small percentage (0.7%; n=74 particles). Cotton accounted for 89% of the natural polymers. The remaining 5.8% (n=625 particles) of the analyzed particles were assigned to "unidentified" due to their unclear spectroscopic spectra.

An extremely high MP content was observed in ID-S1, the duplicates of which accounted for 63.8% (n=6,844 particles) of the total particles and 70% (6,815 MPs) of the total MPs found in the 76 salt samples. When excluding ID-S1, 75.7% of the total 3,879 particles were MPs, followed by unidentified particles (15.4%), natural polymers (7%), and mineral particles (1.9%). Only three brands of investigated salts (CT-S2, CN-R2, and FR-S1) did not contain any MP particles in either replicate sample. CT-S2 and CN-R2 were refined sea salt and refined rock salt, respectively, but FR-S1 was an unrefined sea salt produced by solar evaporation. All other salt samples contained MP particle amounts of ≥2 n/kg above the MDL values.

3.2. Comparison of MP characteristics among salt types

The abundance of MPs found in each salt brand was 0 – 13,629 n/kg (average=675±2,560; median=82; n=28) in sea salts, 0 – 148 n/kg (average=38±55; median=14; n=9) in rock salts, and 28 – 462 n/kg (average=245±307; median=245; n=2) in lake salts (Figure 1(a)). There were significant differences between sea salts (n=28) and rock salts (n=9) (Mann-Whitney U-test; p=0.020) and between sea salts and the others (n=11) (Man-Whitney U-test; p=0.047). High MP contents were found in the two lake salt brands, but statistical analysis was not performed due to the limited sample number (n=2).

The samples of sea salt produced in Asia contained relatively high MP contents than those from other continents: 1,028±3,169 n/kg (median=137; n=18) in Asian salt brands versus 39±9 n/kg (median=31; n=10) in brands from other continents. Additionally, nine of the top 10 sea salts with the highest MP contents were produced in Asian countries (Figure 1(a) and Figure S5). As a result, the MP contents in sea salt were significantly different between the brands produced in Asia and those producing on other continents (Mann-Whitney U-test; p=0.001). On the other hand, no significant difference in MP content was observed between rock salts from Asia and those from other continents (Mann-Whitney U-test; p=0.389).

The major polymers were PE, PP, and PET, but their contributions varied among salt types. The average composition of MPs was PE (35%) > PP (30%) > PET (30%) in sea salts, PET (41%) > PE (26%) > PP (23%) in rock salts, and PP (47%) > PE (28%) > Teflon (11%) > PET (10%) in lake salts. The other 10 polymers were rarely found and had average contributions of <5% (Figure S6(a)). In terms of shape type, fragments were predominant for all salt types, with an average frequency of 63% in sea salt, 54% in rock salt, and 88% in lake salt, followed by fibers (31%, 45%, and 11%, respectively) and sheets (6%, 1%, and 1%, respectively). The spherule type was not found in any samples (Figure S6(b)). MP particle colors were dominantly white, transparent, black, blue, gray and green, together accounting for 89.5% of MPs in sea salts, 81.4% in rock salts, and 97.9% in lake salts (Figure S6(c)). The average size class distribution revealed a marked prevalence of smaller particles (Figure S6(d)). MP particles <500 μm accounted for 47% in sea salts, 61% in rock salts, and 55% in lake salts.

Despite the average characteristics of the MPs discussed above, it is notable that the composition profiles of MPs in terms of polymer type, shape, color and size class varied among the different salt types, different brands of the same salt type, and even the same salt

types with the same origin country (Figure 1(b-e)). For example, PP comprised >50% of MP particles in some of salts with MP contents of >10 n/kg (KR-S1, CN-S1, CNS3, ID-S1, US-S1, BR-S1 for sea salts; CN-R1, PH-R1, and IT-R1 for rock salts; and SN-L1 for lake salt), while PET accounted for >50% in CN-S2, CT-S3, TH-S1, and TH-S2. Although fragments were prevalent in most of the moderately contaminated salts (e.g., >10 n/kg), >50% of the MPs found in CN-S2, CT-S3, TH-S1, TH-S2, IN-S1, VN-S2, and IT-R1 were fibers, the majority of which were composed of PET. In the most polluted sea salt (ID-S1), the predominant MPs had the following characteristics: PP (85%) and PE (15%) polymers, a sheet shape (98%), and a white color (83%); overall, white-colored PP sheets accounted for 83% of the total MP particles in ID-S1. ID-S1 will be discussed further in section 4.3.

FTIR analysis results of the packaging material were consistent with the polymer contents recorded on the packaging of some brands and revealed that the packaging materials of most of the salt products without packaging information were PE (Table 1). All the PE packaging materials were made of transparent sheets, but MPs with the same morphological features, i.e., transparent PE sheet particles, were observed in only four salts, KR-S1, TH-S2, IN-S3, and IT-S3, accounting for 2.3%, 2.1%, 11.8%, and 10% of the total PE particles found in each salt, respectively. For ID-S1 for which transparent PP as packaging material was used, 83% of the total 6,815 MP particles from the replicate were white-colored PP sheets (97.4% of the PP particles), and only one transparent PP sheet was found (Figure 1). Thus, it can be concluded that the packaging materials of the products on sale do not substantially affect the occurrence of MPs present in each salt. Iñiguez *et al.*[16] also observed that grinding or the packing process did not influence the number of MPs in salts.

3.3. Effect of the refining process

Table salt collected in the present study was categorized as ‘solar-dried’, ‘refined’,

and 'unrefined' according to the manufacturing method because these processes could have influenced the MP content of the samples. Solar-dried salt is salt produced through evaporation of salt brine by solar heat. Refined salt is salt manufactured through brine evaporation using heat sources other than solar heat. The brine of most refined salt is purified through electric dialysis. Unrefined salts are rock salts that are mined and mechanically crushed and are mostly not purified.

Most sea salts (n=26) were solar-dried except for only two brands (CT-S2 and UK-S1). CT-S2, the most popular salt brand in Chinese Taipei, in which no MP particles were found, was produced through the electric dialysis and vacuum evaporation tank method. This refining process can remove MP particles. Meanwhile, three solar-dried sea salts from Chinese Taipei contained 115–1,674 n/kg. Another refined sea salt (UK-S1) contained one of the top 10 highest MP contents among global sea salts, 136±5.7 n/kg, although UK-S1's manufacturing procedure is known to filter seawater and boil the brine with the pan-heating method.

Among the 9 rock salts, four brands (CN-R1, CN-R2, DE-R1, and BY-R1) were confirmed to be refined. Although the difference between refined and unrefined rock salts was not statistically significant (Mann-Whitney U-test; p=0.065), unrefined salts contained relatively more MP particles (4 – 148 n/kg; n=5) than refined salts (0 – 14 n/kg; n=4). Only PET fibers were detected in DE-R1 (2 n/kg) and BY-R1 (6 n/kg), while PE fragments were abundant in CN-R1; no MPs were found in CN-R2. PH-R1, known as iodized salt, was not categorized as 'refined salt' because the brand was not included in the list of 'refined iodized rock salts' in the available literature.[29] If PH-R1 is refined salt, the inefficiency of the refining method or the presence of another contamination source during manufacturing is suspected, considering its high degree of contamination (148±5.7 n/kg). These results, based on the limited number of refined salt samples, cannot clearly confirm whether refining processes

remove MPs. Further study for before and after refining process would help more clearly elucidate the effect of the manufacturing process.

3.4. Global distribution of sea salt MPs as an indicator of seawater plastic pollution

The MP pollution level based on particle number in salt (in n/kg) exhibited a distinct geographical distribution. Sea salt containing relatively high MP numbers, including the top 9, were mostly produced on the coasts of eastern and southern Asia (Figure 2(a) and S5). This geographical distribution is similar to that of plastic emissions from rivers into the world's oceans (in the unit of ton/year) estimated by Lebreton *et al*.[2] To understand the correlation between riverine plastic emissions and MP contents in sea salts, we extracted the "midpoint values" of river-to-ocean plastic emissions from geospatial vector data for river mouths located within ±25 km (doi:10.6084/m9.figshare.4725541)[2] of the production sites of the sea salts investigated in our study. Then, the averaged value of the "midpoint values" extracted from the same district was compared with paired sea salt MP content (Figure 2(b) and Table S1). In the comparison, the two refined sea salts (CT-S2 and UK-S1) and one solar sea salt with no MPs (FR-S1) were excluded. Due to the large variation in the original values, the logarithmic values of the two metrics were compared. Despite the limitations caused by the different units, n/kg versus ton/year, the values of the two pollution indices produced a significant correlation ($r=0.57$; $p=0.003$; $n=25$) (Figure 2(b)). This finding shows that sea salt MP pollution is correlated with riverine plastic emissions.

In addition to riverine plastic emissions, we collected seawater and bivalve monitoring data originating from the same or similar regions as the sea salt to directly compare MP pollution levels with the same unit, i.e., number-based MP contents, for both seawater (or bivalves) and sea salt. Only 12 seawater-sea salt pairs were available – the coastal or neighboring off-shore seawaters of China (n=3; the number on basis of sea salt origin),[30-32]

Korea (n=1),[33] Indonesia (n=1),[34] Italy (n=2),[35] Croatia (n=1),[36] Australia (n=1),[37] US (n=1),[38] Brazil (n=1),[39] and Senegal (n=1)[40] – as limited seawater monitoring data have been presented to date (Table S2). Remarkably, a significant correlation was also observed between the logarithmic values of the 11 pairs of seawaters and sea salts (r=0.68; p=0.021; excluding ID-S1 as an outlier) (Figure 2(c)), although the sampling dates and locations of the seawater and sea salt are different, as discussed further in section 4.2. A similar trend was also found between MP contents in bivalves and sea salts measured in the same coastal region (n=6) (Figure S7).

These findings suggest that MP pollution levels in environmental compartments of coastal regions, including seawater, sea salt, and marine organisms, are strongly related to one another and tend to increase proportionally to the amount of mismanaged plastic debris entering the sea.

4. Discussion

4.1. Comparison with other salt studies

MP contents found in the present study can be compared with those reported in previous studies, although the analytical methods and target brands are not identical. Yang *et al.*[14] found 550 – 681 n/kg in sea salts (n=5), 7 – 204 n/kg in rock/well salts (n=5), and 43 – 364 n/kg in lake salts from China. In the present study, Chinese sea, rock, and lake salts contained MPs of 120 – 718 n/kg (n=3), 0 – 14 n/kg (n=2), and 28 n/kg (n=1), respectively. It is notable that similar ranges for sea salts and a similar pattern of sea salts > lake salts > rock/well salts on average were found in both datasets, although the particle size ranges (0.045 – 4.3 mm versus 0.1 – 5 mm) were different. On the other hand, the amount of MPs in Italian and Croatian sea salt was more than two orders of magnitude higher in the study of Renzi and Blašković[19] than in the present study.

In terms of composition of MP polymers, there were remarkable differences among salts in different studies. In the work of Yang *et al.*,[14] cellophane was the predominant MP polymer type in lake and rock/well salts, and PET, polyester, PE, and cellophane were prevalent in sea salt, with similar proportions of each. However, cellophane particles were not found in the present study's Chinese salt samples (Figure 1) or in salts of previous studies.[15-17] Cellophane is made of regenerated cellulose. Notably, it is not easy to exactly distinguish manmade cellulose-based polymers, such as rayon (or viscose), from natural cellulose-based polymers, such as cotton, in FTIR spectra because the spectra of both groups of fiber samples appear similar at first glance.[41] In the present study, cotton fibers were not treated as MPs, and no rayon was found. PET was a predominant component (83.5%) in Spanish salts[16] but a relatively minor one in our salts (24%), as well as in the salts of Gündoğdu (<20%)[15] and Karami *et al.* (6.7%).[17]

In a previous study, the amount of MPs in Chinese salts were primarily determined by visual assessment according to their morphological features, and only some representative MP-like particles (n=152) were selectively analyzed for their polymer identity using FTIR.[14] Similarly, some randomly selected fibers found in Spanish salts[16] and only clear fibers in Croatian sea salts[19] were analyzed using FTIR. Previous studies confirmed that MPs in environmental samples can be significantly under- or overestimated using stereomicroscopic analysis.[20,21] Silva *et al.*[24] described the misidentification of approximately 20% of the particles initially identified as MPs by visual observation. Furthermore, 32% of visually counted MP particles below 100 μm were not confirmed as MPs by micro-Raman analysis, and FTIR analysis revealed that up to 70% of particles were erroneously identified as MPs. Meanwhile, in Raman spectroscopy analysis, additive and pigment compounds in MPs can act as signal confounding factors and interfere with the identification of the polymer types in environmental samples.[22,23] Notably, much small amounts of MPs[15,17] and significant

351 amounts of pigments (23.6%)[17] were found in the salts of two studies using Raman
352 spectroscopy.

353 Previous salt studies used various filters (e.g., cellulose nitrate filter, GF/D filter and
354 filter fiber disk) with different pore sizes of 0.2 – 149 μm.[14-19] Different minimum sizes of
355 targeted MPs might be another significant factor to have caused the differences among
356 studies. For example, the vast amount of MPs detected in Italian and Croatian sea salts of
357 Renzi and Blašković[19] might result from measurement of much smaller sized MP particles
358 (i.e., a minimum size = 4 μm), in addition to overestimation by visual observation method.
359 However, how substantially small sized MPs contributed to total MP content found in that
360 study is unclear because the data of size distribution of MPs found was not presented.

361

362 **4.2. Geographical distribution of sea salt MPs as an indicator of sea water MP pollution**

363 In this study, positive correlations were identified between MPs in sea salts and
364 riverine plastic emissions, MP contents in seawater, and MPs in marine organisms. However,
365 the interpretation of the results presented here needs attention due to limitations associated
366 with inconsistencies in the compared data.

367 First, the use of different units for MP content can cause inherent error in the case of
368 this study. The present study uses a number-based unit for sea salt, whereas a mass-based unit
369 was used for riverine plastic emissions. Number-based MP contents are important in exposure
370 and risk assessment because the toxicological effects vary depending on the morphological
371 features of individual MP particles;[42] however, this format could be inappropriate in
372 estimating the mass balance of plastic because a single high-mass piece of plastic litter can
373 generate numerous lighter MP particles. Nevertheless, the remarkably good correlation
374 between the two metrics established in the present research should not be overlooked.
375 Individual MP particles in sea salts were not weighed; thus, their weights were instead

estimated based on their original densities and measured volumes (see section S3 of supporting information). The roughly estimated mass-based MP amount for sea salts showed a nonsignificant but increasing trend with riverine plastic emissions (Figure S8(a)).

Second, it may be difficult to directly compare sea salt with other environmental compartments, such as seawater and marine organism, due to the different methods, locations, and seasons associated with the sampling. We collected all the currently available literature reporting MP pollutions in seawater and marine bivalves around the production sites of the sea salts investigated in the present study (Table S2 and Figure 2 and S7). Interestingly, the sea salts produced near areas with relatively high MP contents in seawater contained proportionally higher MP levels, and the predominant components of MPs were similar in several pairs in China, Indonesia, Italy, US, and Brazil. However, the compositions and the dominant components of MPs did not exactly match in other pairs. Different mesh sizes (153 to 505 μm), different seawater column depths (surface and subsurface water), and different monitoring seasons can cause potential differences among seawaters and between seawater and sea salts. Thus, a correlation obtained in the present study should be interpreted with caution as just an indirect evidence. Further study matching the pairs of source seawater and sea salt is expected to improve the uncertainty. Qu *et al.*[43] found a strong positive linear relationship (r^2>0.7) between MP levels in the pairs of seawater and associated mussels at 25 sites along the Chinese coastline. Data on MP pollution in bivalves for comparison with sea salts were available for only six monitoring sites in China (n=3), France (n=1), US (n=1), and UK (n=1) (Figure S7). Despite limitations in the available data, a general trend with a positively linear relationship was also present between bivalves and sea salt.

As a result, correlations observed in the present study suggest that MP pollution in various compartments is related to the plastic burden discharged to each coastal region and thus that sea salt can be a useful preliminary monitoring matrix.

401

402 **4.3. Indonesian sea salt, the outlier**

403 For Indonesian sea salt, our analysis results showed a very close similarity between
404 the replicates not only in the amounts with 14,932 and 12,326 n/kg (CV of 13.5%) but also in
405 the composition with a predominance of white-colored PP sheets (86% and 80%). Thus, the
406 uncertainty in the MP content of this salt might be negligible despite the limitation of the
407 analytical method for MP determination.

408 Indonesian seawater data (0.4 n/m^3),[34] collected on the Cilacap coast, was excluded in
409 our correlation because the sampling location is 700 km away from the production site of the
410 sea salt (ID-S1). However, it is notable that the composition patterns of identified MPs were
411 identical in terms of polymer type and color between the seawater and sea salt samples, as
412 shown in Table S2. Indonesia is considered to be the second-largest plastic emitter to the
413 ocean in the world after China.[1] A recent study predicted that Indonesian rivers, occupying
414 four of top 20 global plastic-discharging rivers, discharge 0.08 – 0.17 MMT of plastics
415 annually to the ocean, accounting for 7% of global river inputs.[2] Through comparison of the
416 amounts of plastic debris in the gastrointestinal tracts of fish, Rochman *et al.*[10] found that fish
417 purchased from local markets in Indonesia contained MP numbers dozens of times higher
418 than those in the US. These findings all support the hypothesis that the tremendously high MP
419 contents found in Indonesian salt could originate from the notorious pollution along its coast.
420 Therefore, the case of Indonesia, from which sea salt showed the highest MP content,
421 represents another proof that sea salt can be an alternative compartment for seawater
422 pollution monitoring.

423

424 **4.4. Implications for human exposure**

425 Considering a global mean salt-equivalent intake of 10.06 g/day,[44] the average adult

426 salt consumer worldwide consumes approximately 2,000 MPs per year (i.e., based on an
427 average MP content of 506 n/kg for all salts; n=39). The present study demonstrates that the
428 number of MPs in salt is strikingly different among different brands of salts and is especially
429 high in those consumed in Asian countries. Given the average MP content in salt brands
430 consumed in each country investigated in our study and the average salt-equivalent intake of
431 each country,[44] MP intakes via salt consumption range widely from 0 to 117 n/capita/day,
432 equivalent to 0 to 42,600 n/capita/year, with an average of 3,000 n/capita/year. Much higher
433 exposures are expected for the consumers of salts produced in Asia. However, it is important
434 to acknowledge that although consumed in the same country, different brands and types of
435 salt exhibited large ranges in MP content, varying from zero to several hundreds or thousands
436 of MPs per 1 kg salt, as depicted in Figure S9. Therefore, MP intakes can be very different
437 among different citizens in the same country depending on which brand of salt they consume.

438 MPs are also ingested by humans via other pathways, including inhalation, drinking,
439 and ingestion of MP-contained air, water, and foods, respectively. Recent studies also showed
440 the presence of MPs in indoor and outdoor air,[45] in tap water,[18] and in bivalves (see the
441 references in Figure S6). In an exposure scenario with the average MP concentrations
442 presented in the literature (i.e., 5.4 n/m^3 for indoor air, 0.9 n/m^3 for outdoor air, 5.45 n/L for
443 tap water, and 1.87 n/g for bivalves) and average exposure parameters (i.e., an inhalation rate
444 of 14.3 m^3/day with relative residence proportions of 0.89 indoors and 0.11 outdoors,[46] a
445 drinking rate of 1.4 L/day,[47] and a bivalve ingestion rate of 3.17 g/day (global average
446 consumption of mollusks)),[48] adults are expected to intake approximately 32,000 MP
447 particles annually via the four pathways concerned, with inhalation exposure representing the
448 dominant exposure pathway (approximately 80%). Salt ingestion might be a minor
449 contributor, accounting for approximately 6% of total exposure.

450 MP intake via salt consumption in the present study assumes that only the salt brands

investigated in this study are consumed and that sodium intake originates solely from salt consumption. Thus, annual MP intake via salt consumption could be much less than suggested in the present study, if other sources of sodium are considered and the outlier (ID-S1) is excluded. Furthermore, it should also be noted that most of the refined sea salt brands consumed in each country were not investigated in this study. Contrarily, only MP particles >100 μm were counted in the present study. As shown in Figure 1 and Figure S6, the number of MP particles tends to increase with decreasing particle size. Thus, our estimate for human exposure might be underestimated because uncounted smaller MPs are expected to be prevalent. MP exposure is thought to be a harmful threat to human because of injuries caused by the microparticles themselves and the absorption of harmful chemicals in or on plastics.[4,5] Our results suggest that human exposure via ingestion of marine products is strongly related to plastic emissions. Therefore, preventive measures to limit human exposure to MPs are required, such as controlling environmental discharge of mismanaged plastics and fundamentally reducing plastic waste.

To our knowledge, this paper is the first attempt to present a geographical distribution of MP pollution based on consistent and extensive sea salt monitoring data and to assess the relationships between environmental discharge and pollution levels of plastics.

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Supporting information. The details of the analytical procedure of MPs in salt samples (section S1), quality assurance/quality control (QA/QC) (section S2), and methods used to

estimate the weight of individual MP particles (section S3) are described. Furthermore, additional data are provided, including maps of the locations where each salt brand was purchased (Figure S1) or produced (Figure S2), details of the measurement method and an example picture of MPs in Indonesian salt (Figure S3), typological features and FTIR spectra of various MP particles found in salt samples (Figure S4), the rank of salt brands in order of MP content (Figure S5), comparison of average composition profiles of MPs among salt types (Figure S6), the relationship between MP contents in sea salts and bivalves (Figure S7), the relationship between mass-based MP contents in sea salts and riverine plastic emission (Figure S8), the variation in MP contents among different brands and types of salts that are consumed in the same country (Figure S9), data on riverine plastic emissions and sea salt-hosted MPs (Table S1), and a comparison of MP contents in seawater and sea salts originating from similar regions (Table S2).

Brief

The abundance of microplastics in sea salts produced from geospatially different countries showed linear correlations with riverine plastic emissions and microplastic pollution levels in seawater and marine organisms, suggesting the possibility that sea salt can be used as an indicator or alternative monitoring matrix for determining the magnitude of microplastic pollution in the surrounding marine environment.

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TABLE CAPTION

Table 1. Details of table salt products investigated in the present study.

FIGURE CAPTIONS

Figure 1. Abundance (A) and composition profiles by polymer type (B), shape (C), color (D), and size of MPs detected in sea salts (SS), rock salts (RS), and lake salts (LS). The vertical and error bars in (A) indicate the average and standard deviation of MP abundance, respectively. MPs were not detected (n.d.) in three brands, and thus, their composition profiles are not available (n/a). The stars in (A) indicate the refined salt brands.

Figure 2. Potential possibility of sea salt as an indicator of geographical distribution of MP pollution: Geographical distribution of salt-hosted MP abundance (A) and the relationships between MP abundance in sea salt and riverine plastic emissions (B) and the MP abundance in surface seawater near the sea salt production site (C). Each spot for salts in (A) represents the production site for each table salt brand and the

651 references (superscripts "a" and "b") quoted in (B) and (C) were presented in the
652 text or supplementary information.
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661 **Table 1.** Details of table salt products investigated in the present study.

NO.	Consumption location (17 countries)	Sample ID	Production location (21 countries)	Salt types	Manufacuring process	Packing material	
						description[a]	FTIR analysis[b]
1	Korea (KR)	KR-S1	Korea	sea salt	solar[d]	PE	whoe body -PE
2		KR-S2	Korea	sea salt	solar	PE	whoe body -PE
3		KR-S3	Korea	sea salt	solar	PE	whoe body -PE
4	China (CN)	CN-S1	China	sea salt	solar	—[i]	whoe body -PE
5		CN-S2	China	sea salt	solar	—	whoe body -PE
6		CN-S3	China	sea salt	solar	—	whoe body -PE
7		CN-R1	China	rock salt	refined[e]	—	whoe body -PE
8		CN-R2	China	rock salt	refined	—	whoe body -PE
9		CN-L1	China	lake salt	solar	—	whoe body -PE
10	Chinese Taipei (CT)	CT-S1	Chinese Taipei	sea salt	solar	—	whole body - Glass
11		CT-S2	Chinese Taipei	sea salt	refined	LDPE	whole body - PE
12		CT-S3	Chinese Taipei	sea salt	solar	—	whoe body -PE
13		CT-S4	Chinese Taipei	sea salt	solar	—	whole body - Glass
14	Thailand (TH)	TH-S1	Thailand	sea salt	solar	—	whoe body -PE
15		TH-S2	Thailand	sea salt	solar	—	whoe body -PE
16	Philippines (PH)	PH-R1	Philippines	rock salt	unrefined[f,g]	—	whoe body -PE
17	India (IN)	IN-S1	India	sea salt	solar	—	whoe body -PE
18		IN-S2	India	sea salt	solar	—	whoe body -PE
19		IN-S3	India	sea salt	solar	—	whoe body -PE
20	Vietnam (VN)	VN-S1	Vietnam	sea salt	solar	—	whoe body -PE
21		VN-S2	Vietnam	sea salt	solar	—	whoe body -PE
22	Indonesia (ID)	ID-S1	Indonesia	sea salt	solar	PP	whoe body -PP
23	France (FR)	FR-S1	France	sea salt	solar	PS	lid - PS body - Hard Paper
24	Italy (IT)	IT-S1	Italy	sea salt	solar	—	whole body - Hard Paper
25		IT-S2	Italy	sea salt	solar	—	whole body - Hard Paper
26		IT-R1	Italy	rock salt	unrefined	—	whole body - Hard Paper
27	United Kingdom (UK)	UK-S1	UK	sea salt	refined[h]	—	body inside - PE body outside - Hard Paper
28		AU-S1	Australia	sea salt	solar	plastic[j]	lid - PS body - Hard Paper
29	Germany (DE)	DE-R1	Germany	rock salt	refined	—	whole body - Hard Paper
30	Bulgaria (BG)	BG-S1	Bulgaria	sea salt	solar	PP	whole body - PP
31		BY-R1	Belarus (BY)	rock salt	refined	PP-PE	whole body - PE
32	Hungary (HU)	HU-R1	Hungary	rock salt	unrefined	LDPE	whole body - PE
33	Croatia (HR)	HR-S1	Croatia	sea salt	solar	PP, PE	lid - PP/PE body - Hard Paper
34	US	US-S1	USA	sea salt	solar	—	whole body - Hard Paper
35		BR-S1	Brazil (BR)	sea salt	solar	Plastic	whole body - PE
36		US-R1	USA	rock salt	unrefined	—	whole body - PE
37		PK-R1	Pakistan (PK)	rock salt	unrefined	plastic	body inside - PE body outside - Hard Paper
38	Senegal (SN)	SN-S1	Senegal[c]	sea salt	solar	—	whole body - PET
39		SN-L1	Senegal	lake salt	solar	—	whole body - PE

[a] polymer of packing material described on each package. [b] polymer of packing material identified by FTIR. [c] not confirmed due to no details available but assumed to be produced in PH and SN, respecitvley. [d] solar dry - brine evaporation by solar heat. [e] refined - brine evaporation by other heat source than solar, mostly purified thorugh electric dialysis. [f] unrefined - mined and mechanically crushed, mostly not purified. [g] iodized salt for PH-R1. [h] filtration, pan-heat & air-dry. [i] No information about packing material was presented on the package. [j] polymer-identity of packing material was not presented and only description was "plastic".

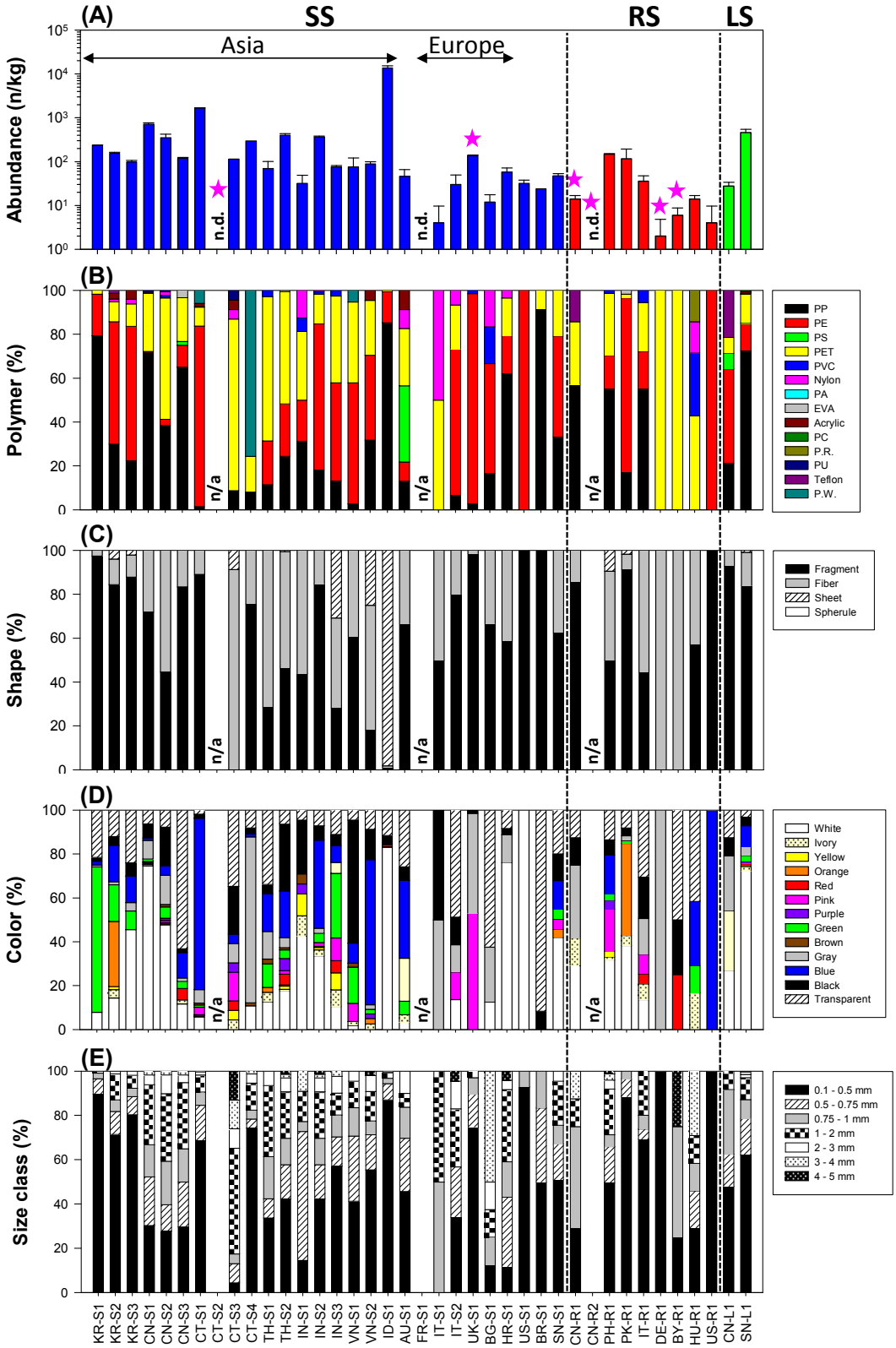
(A)
SS
RS
LS
Asia
Europe
Abundance (n/kg)
n.d.
(B)
Polymer (%)
n/a
PP
PE
PS
PET
PVC
Nylon
PA
EVA
Acrylic
PC
P.R.
PU
Teflon
P.W.
(C)
Shape (%)
Fragment
Fiber
Sheet
Spherule
(D)
Color (%)
White
Ivory
Yellow
Orange
Red
Pink
Purple
Green
Brown
Gray
Blue
Black
Transparent
(E)
Size class (%)
0.1 - 0.5 mm
0.5 - 0.75 mm
0.75 - 1 mm
1 - 2 mm
2 - 3 mm
3 - 4 mm
4 - 5 mm
KR-S1
KR-S2
KR-S3
CN-S1
CN-S2
CN-S3
CT-S1
CT-S2
CT-S3
CT-S4
TH-S1
TH-S2
IN-S1
IN-S2
IN-S3
VN-S1
VN-S2
ID-S1
AU-S1
FR-S1
IT-S1
IT-S2
UK-S1
BG-S1
HR-S1
US-S1
BR-S1
SN-S1
CN-R1
CN-R2
PH-R1
PK-R1
IT-R1
DE-R1
BY-R1
HU-R1
US-R1
CN-L1
SN-L1

Figure 1.

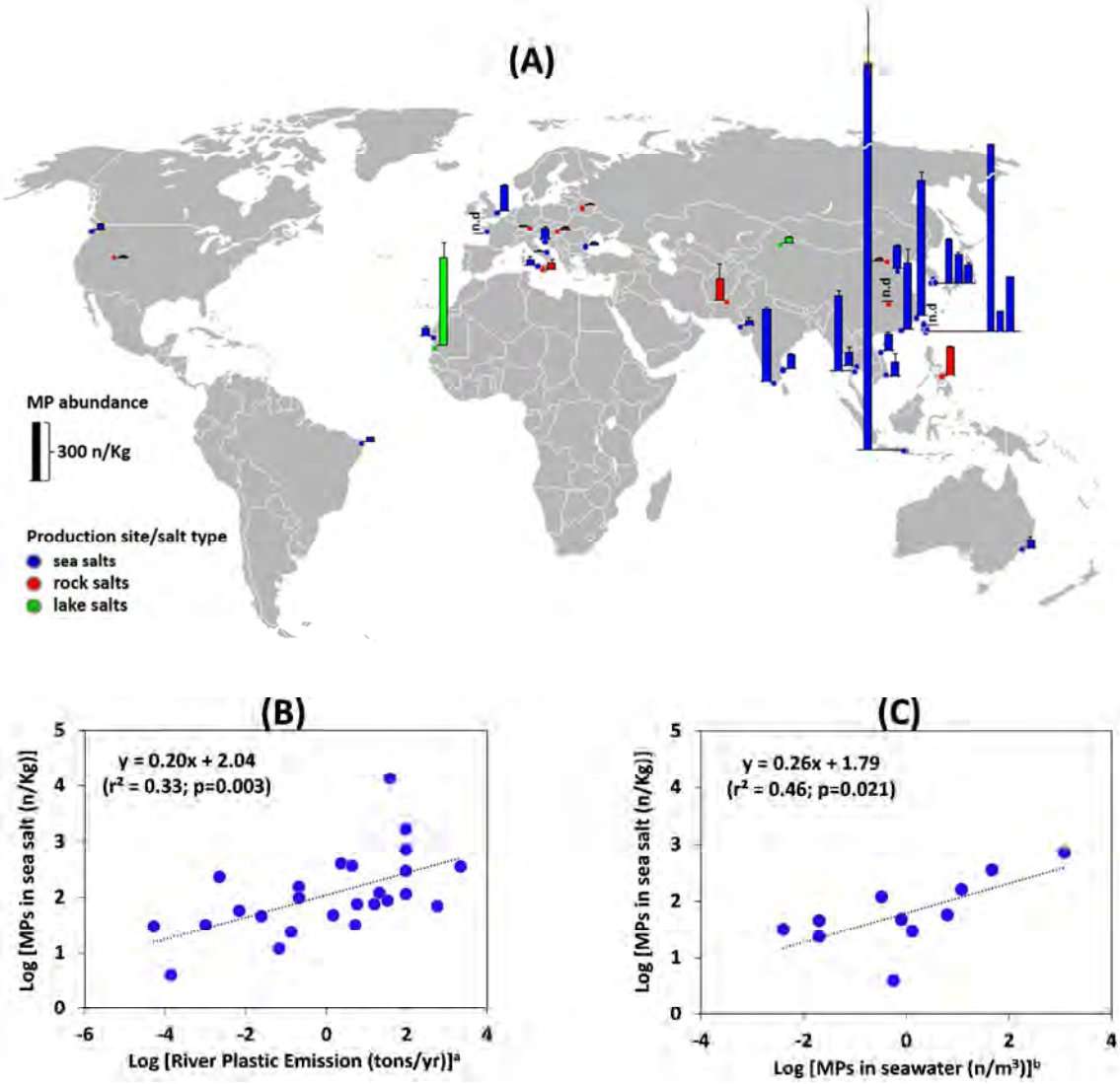
(A)
MP abundance
300 n/Kg
Production site/salt type
sea salts
rock salts
lake salts
n.d
(B)
y = 0.20x + 2.04
(r² = 0.33; p=0.003)
Log [MPs in sea salt (n/Kg)]
Log [River Plastic Emission (tons/yr)]ᵃ
(C)
y = 0.26x + 1.79
(r² = 0.46; p=0.021)
Log [MPs in sea salt (n/Kg)]
Log [MPs in seawater (n/m³)]ᵇ

Figure 2.